

Chemical composition of essential oils of *Origanum vulgare* L. growing in Lithuania

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Twenty essential oils of all aerial parts, inflorescences and leaves of cultivated *Origanum vulgare* L. were analyzed by capillary gas chromatography and mass spectrometry (GC/MS). The main constituents in 6 out of 7 samples of inflorescence oils were sabinene (8.7–19.5%), β -caryophyllene (15.4–24.9%) and germacrene D (12.3–16.0%) and in the same quantity of the leaf oils – β -caryophyllene (15.9–21.3%), germacrene D (12.1–15.7%) and caryophyllene oxide (4.7–11.1%). The amount of phenols, thymol and carvacrol, was 0–3.9 %. The chemical composition with a low concentration of phenols of the above oregano disagreed with the data on high concentrations of these compounds reported in some books describing Lithuanian medical and aromatical plants. The major part (60.5–90.5%) of the oils (except commercial plants) consisted of mono- and sesquiterpene hydrocarbons. The identified constituents made up 79.5–98.0% of the essential oils.

Key words: *Origanum vulgare* L., composition of essential oil, sabinene, β -caryophyllene, germacrene D, caryophyllene oxide

INTRODUCTION

Data on the chemical composition of some essential oils of *Origanum vulgare* L. given in some Lithuanian books are inaccurate. For example: the books “Medicinal Plants” (1973) [1], “Aromatic Plants” (1989) [2]. “The Minor Forest Resources” (1992) [3], “Plants – Our Life” (1996) [4] and “Handbook of Medicinal Plants” (2002) [5] informed us that the essential oils of oregano growing in Lithuania contained up to 40–44% of phenols (thymol and carvacrol). Analysis of oils from plants of Lithuanian botanical gardens (1980) showed that the content of thymol and carvacrol was 2.3% and their derivatives comprised 1.9% [6]. Nearly the same amount of phenols was found in the oil from oregano plants grown in the experimental garden of Lithuanian Institute of Horticulture (1996; Table 1, B) [7]. The major constituents of the above oil were sabinene, 1,8-cineole and caryophyllene oxide.

Only one species of oregano *O. vulgare* ssp. *vulgare* is growing wild in Lithuania [8]. Essential oils of oregano plants grown wild in Eastern Lithuania did not contain phenols, either, and the main constituents were β -ocimene, β -caryophyllene and germacrene D (Tables 1 and 2, W, IW, LW) [9, 10].

More than half of essential oils from 70 clones of *O. vulgare* ssp. *vulgare* cultivated in France (seeds from different European countries) contained 0–1.4% of thymol and only several samples contained \leq 14.5% [11]. The amount of carvacrol did not ex-

ceed 0–0.95% in all 70 investigated oils. The major constituents of the above leaf oils were mono- (sabinene, cis-sabinene hydrate, β -ocimene, terpinen-4-ol) and sesquiterpenoids (germacrene D, β -caryophyllene, caryophyllene oxide). Essential oils of the same subspecies in India contained 62.0% of γ -murolene [12].

Inflorescence oils of four chemotypes (β -caryophyllene, thymol, terpinen-4-ol, p-cymene + β -caryophyllene) from *Origanum vulgare* ssp. *vulgare* cultivated in Italy were investigated [13]. Low contents of thymol (1.1%) and carvacrol (0.3–0.6%) were found in the oils of terpinen-4-ol and p-cymene + β -caryophyllene chemotypes. A higher content of carvacrol (7.6%) was determined in the oil of β -caryophyllene chemotype (seeds from USA).

Carvacrol (37.7–83.8%) and thymol (41.3–79.4%) were the major constituents in oils of *O. vulgare* ssp. *hirtum* (7 samples), *O. vulgare* ssp. *glandulosum* (2 samples) and *O. vulgare* ssp. *grasile* (1 sample) cultivated in Italy [13]. Two phenol chemotypes of the oils of *O. vulgare* ssp. *hirtum* were also found by other investigators [14–22]. Thymol (61.0–69.1%) was the main constituent in the oil of *O. vulgare* ssp. *viridulum* from Greece [23]. Some essential oils in Finland from wild *O. vulgare* L. (without specification) contained carvacrol as a dominant constituent [24].

Essential oils of *O. vulgare* ssp. *virens* cultivated (seeds of different origin) in Italy were grouped into two chemotypes: linalool (3 samples) and terpi-

Table 1. The chemical composition of the essential oils of aerial parts of *Origanum vulgare* L.^a

Compound	RI	A	B [7]	C	D	E [31]	W [9]
α -Thujene	931	0.0–0.9	0.4–1.2	tr-0.5	–	0.0–0.1	0.2–1.1
α -Pinene	939	0.4–0.5	0.8–1.0	1.0–1.6	0.2–0.3	0.0–0.1	0.3–1.0
Sabinene	976	19.2–22.9	14.0–14.1	8.5–10.6	0.1–1.5	0.3–1.6	6.4–14.2
β -Pinene	980	–	3.0	0.4–1.4	0.6–1.9	0.0–0.4	0.5–1.2
1-Octen-3-ol	984	0.5	3.0	0.1–0.4	–	0.1–0.2	0–0.9
Myrcene	991	0.9	0.2–0.3	1.4	–	0.0–0.2	1.1–2.0
α -Terpinene	1018	0.4–0.9	0.8–0.9	0.4–1.4	tr	0.1–0.2	0.6–1.6
<i>p</i> -Cymene	1026	0.6–4.3	2.5–3.9	0.4–1.0	0.1–0.7	0.1–1.0	0–0.9
β -Phellandrene	1032	0.7–0.9	–	3.0–6.5	–	0.0–0.8	4.7–7.9
1,8-Cineole	1033	tr-3.6	9.5–11.6	3.1–6.9	0.1–0.3	0.5–2.9	4.7–7.9
(<i>Z</i>)- β -Ocimene	1040	5.4–6.1	2.1–5.3	5.0–5.9	0.9–1.1	0.2–1.4	6.2–11.5
(<i>E</i>)- β -Ocimene	1050	0.8–3.1	1.0–3.3	5.1–5.9	tr	0.5–1.3	7.0–11.5
γ -Terpinene	1062	1.2–3.2	1.6–1.9	1.9–2.8	–	0.3–0.9	0–2.6
<i>cis</i> -Sabinene hydrate	1068	tr-1.5	0.9–1.0	0.0–0.2	–	–	0–0.4
Terpinolene	1088	–	0.3–0.4	0.4–0.5	–	0–0.1	0–0.6
Linalool	1099	0.9–1.2	1.7–2.1	0.9–1.0	tr	0.3–1.4	0.6–2.9
Terpinen-4-ol	1177	1.2–1.9	1.2–1.6	0.9–1.2	1.4–4.2	0.5–1.5	0–3.0
β -Terpineol	1187	tr-0.2	1.5–1.7	0.9–1.0	0.1–0.3	0.2–1.5	0.8–2.4
Thymol	1290	tr-0.2	1.4–3.2	0.0–0.6	1.6–1.7	0–0.2	–
β -Bourbonene	1387	0.6–0.8	1.0	1.2–1.6	1.3–1.6	1.9–2.5	0.5–1.0
β -Elemene	1391	–	–	0.0–0.3	tr-0.3	0.6–0.8	0–0.4
β-Caryophyllene	1418	18.2–18.8	5.5–7.7	14.2–15.4	11.9–12.9	24.6–31.3	10.8–15.4
β -Gurjunene	1432	–	0.2	0.3–0.9	0.1–0.3	–	0–0.2
α -Humulene	1454	2.3–3.1	0.8–1.3	2.1–3.3	0.2–0.6	5.0–5.3	1.5–2.6
<i>allo</i> -Aromadendrene	1461	0.4–0.5	–	tr-0.4	0.3–0.7	1.7–2.3	0–0.6
Germacrene D	1480	9.6–10.5	3.8–5.5	11.0–11.6	11.6–12.8	13.7–19.2	10.6–16.9
Bicyclgermacrene	1494	0.6–3.5	–	tr-3.2	1.0–1.7	2.8–3.7	1.4–4.5
α -Muralene	1499	0.3–3.0	–	0.4–0.5	tr-0.5	0.9–1.1	0–0.8
α -Farnesene+	1508	1.1–5.5	0.5–1.0	3.2–6.0	5.0–7.1	8.4–11.5	4.1–6.9
β -bisabolene	1509						
δ -Cadinene	1524	tr-0.9	0.3–0.5	0.7–1.5	1.7–3.0	3.5–5.0	1.3–3.2
Spathulenol	1576	3.9–4.9	1.5–3.1	0.9–3.2	9.3–12.1	1.2–1.7	1.4–3.2
Caryophyllene oxide	1581	8.6–8.7	9.6–10.6	4.3–8.0	21.9–23.4	5.0–5.9	0.9–7.1
α -Cadinol	1653	0.4–1.4	0.4–0.6	0.2–0.6	0.5–1.0	2.0–3.1	0–1.2
Total		91.6–97.4	89.6–90.7	88.4–92.9	79.5–80.0	89.3–97.5	85.6–98.0
Monoterpene							
Hydrocarbons		34.7–39.1	30.8–34.9	33.9–35.6	2.9–4.9	1.6–8.2	29.5–41.8
C ₁₀							
Monoterpenoids		35.9–46.0	54.9–59.4	40.1–46.2	5.9–12.0	4.9–10.6	39.9–52.0
Hydrocarbons		36.2–38.5	12.0–17.8	35.7–39.9	33.9–35.5	65.6–80.4	38.5–46.8
C ₁₅							
Caryophyllane skeleton		26.8–26.9	14.3–16.1	18.5–23.4	35.3–35.8	29.6–37.2	21.5–30.9

^a tr – traces; some more constituent with low concentration beside given in table were in [7, 31] essential oils.

RI – retention indexes on nonpolar column, **A** – Rokiškis district (Aleknos) and Vilnius (Deðkinė), **B** – data of P.R. Venskutonis [7], **C** – Vilnius district (Nemenėnė and Siponys), **D** – bought in the chemist shops, **E** – plants with pink flowers (seed from Moscow botanical garden) [31], **W** – aerial part of wild plants from Vilnius district [9].

Table 2. Chemical composition of essential oils of *Origanum vulgare* L. inflorescences (F, G, H, J, IW) and leaves (F', G', J', H', LW)^a

Component	RI	Leaves					Inflorescences				
		F	G	H	J	IW[10]	F'	G'	H'	J'	LW[10]
α-Thujene	931	tr	0.4–0.5	0.5–1.7	0.4	tr-0.8	tr	tr	0–0.3	tr	tr-0.3
α-Pinene	939	0.1	tr-1.7	tr-0.6	0.5	0.5–1.2	tr	tr	tr-1.1	0.4	0.4–0.9
Sabinene	976	5.8	8.7–19.5	14.4–15.7	17.1	10.5–15.8	3.8	3.4–4.5	6.2–8.1	7.7	6.7–9.8
β-Pinene	980	0.1	0.6–0.8	1.1–1.5	1.1	1.0–1.6	1.4	tr	tr-0.4	0.6	0.6–1.0
Myrcene	991	0.4	–	tr-0.7	1.0	0.4–1.3	–	tr	tr-1.1	2.7	2.4–3.5
α-Terpinene	1018	–	0.2–0.5	0.6–0.8	1.2	0–1.4	0.2	–	0.1–0.3	0.1	tr-0.6
p-Cymene	1026	–	0.7	0.5–0.7	0.2	0–2.0	0.1	tr	0.4–1.7	0.3	tr-0.3
β-Phellandrene	1032	0.2	0–0.2	tr-1.1	1.3	3.2–9.5	–	tr	0.1–3.9	0.3	1.7–4.6
1,8-Cineole	1033	3.7	tr	2.7–4.5	7.8	3.2–9.5	3.4	tr	0.9–4.1	5.6	1.7–4.6
(Z)-β-Ocimene	1040	1.1	1.5–2.0	4.7–5.2	5.8	2.8–5.4	5.1	0.9–1.0	3.6–5.5	7.8	14.0–16.6
(E)-β-Ocimene	1050	4.3	1.7–2.8	6.1–6.4	6.9	9.8–12.7	3.5	0.3–0.4	2.1–2.9	6.1	6.6–8.5
γ-Terpinene	1062	0.8	0.5–1.3	1.5–2.3	1.1	0.6–2.4	0.3	tr	0.2–2.9	0.7	0.3–1.0
cis-Sabinene hydrate	1068	tr	–	0.1–0.5	0.2	0–0.6	0.3	–	0–0.3	tr	0–0.1
Terpinolene	1088	tr	–	0.2–0.6	0.2	tr-0.4	0.2	tr	0–0.1	tr	0–0.2
trans-Sabinene hydrate	1096	tr	–	0–0.3	tr	tr-1.0	–	–	–	–	tr-0.2
Linalool	1099	0.4	0.7–1.2	0.7–1.6	1.1	0.3–5.9	0.5	tr-0.1	tr-1.1	0.9	tr-2.1
allo-Ocimene	1129	–	–	–	–	–	3.4	tr-0.6	0–1.9	–	–
Terpinen-4-ol	1177	0.6	tr-0.6	0.6–1.5	0.3	tr-1.2	0.3	0–0.2	0.1–0.9	1.2	0–0.6
α-Terpineol	1187	0.2	0.1–0.8	0.6–1.4	0.9	0.5–3.7	0.1	0.2–0.7	0.1–1.2	0.2	0–0.6
Thymol	1290	1.2	0.1–0.2	0.2–1.4	0.4	–	1.6	0.5–0.8	0.1–1.7	–	–
Carvacrol	1298	–	tr	0–0.2	–	–	1.3	–	0–0.2	–	–
Bourbonene	1387	0.2	0.7–0.8	0.5–0.9	0.6	tr-0.4	0.7	0.6–3.2	1.7–5.9	2.9	0.4–1.4
β-Elementene	1391	0.7	0.4–0.8	0.7–1.2	0.5	–	0.8	0.7–0.9	0-tr	0.4	–
β-Caryophyllene	1418	11.6	21.9–24.9	15.4–17.0	18.7	10.2–14.5	11.0	20.3–21.3	15.9–18.3	16.1	9.3–13.7
β-Gurjunene	1432	1.6	1.5–1.7	0.6–1.8	0.5	tr-1.3	1.5	1.6	0.9–1.4	0.5	tr-0.3
α-Humulene	1454	2.5	4.7–5.4	2.5–3.7	3.2	0.5–2.7	2.3	5.0–5.3	2.3–3.1	3.1	0.8–2.6
allo-Aromadendrene	1461	1.5	tr-1.1	0–0.3	tr	0.5–0.9	1.7	0-tr	0–0.8	tr	–
γ-Muurolene	1477	–	–	–	–	–	tr	1.1–1.5	tr-0.2	0.1	–
Germacrene D	1480	16.0	12.9–14.6	12.3–13.7	11.4	9.5–15.9	14.3	15.1–15.7	12.1–13.8	14.0	12.7–15.7
Bicyclgermacrene	1494	6.4	5.4	0.4–2.1	1.7	1.4–3.5	6.0	5.5–6.7	tr-1.2	2.0	2.6–4.1
α-Muurolene	1499	–	–	0–0.8	tr	tr-0.7	tr	–	0.1–3.9	0.1	0.2–0.5
α-Farnesene	1508	–	tr-0.1	4.1–6.8	4.9	6.1–9.4	–	tr-5.5	0.4–4.1	2.3	2.5–3.6
β-Bisabolene	1509	5.5	4.3–5.0	0.7–2.0	1.9	6.1–9.4	2.4	2.4–4.7	1.6–4.1	1.6	2.5–3.6
γ-Cadinene	1524	1.3	tr	tr-0.1	0.2	–	1.4	0.1–0.4	tr-0.1	tr	–
δ-Cadinene	1524	4.3	2.0–4.0	0.6–1.6	0.7	1.6–2.5	3.7	0.4–1.2	0.1–0.9	0.6	1.7–2.4
α-Cadinene	1538	–	–	–	–	–	0.4	tr-2.8	0.2.6	1.1	–
Germacrene D-4-ol	1574	0.3	0.1–0.3	tr-1.9	1.2	1.7–3.4	0.1	0.3–0.5	0–1.1	0.7	2.6–4.9
Spathulenol	1576	6.5	2.7–3.0	0.1–0.9	1.3	–	5.0	4.5–5.3	0.5–3.1	0.3	–
Caryophyllene oxide	1581	1.3	2.9–3.1	1.6–3.6	1.7	1.2–4.8	2.7	7.6–8.9	4.7.11.1	5.2	1.4–4.1
epi-γ-Eudesmol	1619	2.2	0.7–0.9	0.2–0.6	0.4	–	0.3	0.5–1.4	0.1–0.3	0.1	–
epi-α-Muurolol	1642	3.6	1.5–2.4	0.3–0.9	0.5	0.4–1.6	2.3	0.7–1.1	0.2–0.5	0.4	0.4–2.1
α-Cadinol	1653	1.0	0.3–0.5	1.0–1.8	1.2	0.3–3.1	0.4	0.4	0.8–0.5	0.4	0.4–2.1
Total		85.4	86.5–96.4	89.1–95.1	97.9	93.0–97.8	82.5	84.3–84.5	82.4–85.7	86.7	87.2–95.0
Hydrocarbons C ₁₀		14.0	15.4–29.2	33.0–35.3	37.0	33.9–39.9	20.9	5.3–6.3	17.2–24.6	26.7	34.9–43.2
Monoterpenoids		18.9	16.7–31.3	38.3–45.6	47.3	45.2–59.9	25.5	6.3–7.3	18.3–33.0	34.6	38.7–48.3
Hydrocarbons C ₁₅		51.6	56.3–61.2	43.3–45.7	44.3	34.1–44.0	46.2	61.3–62.3	43.3	44.8	36.0–43.2
Caryophyllane skeleton		12.9	24.8–28.0	17.5–20.3	20.4	14.9–16.7	13.7	28.9–29.2	20.6–29.4	21.3	12.0–16.7

^a tr – traces, RI – retention indexes on nonpolar column.

F, F' – Vilnius (Antakalnis).

G, G' – Vilnius (Deðkinė and Salininkai).

H, H' – Vilnius district (Nemenėnė) and Rokiškis district (Aleknos, plants with pink and rose-pink flowers).

J, J' – Aleknos, plants with white flowers.

IW, LW – inflorescences and leaves of wild *Oregano* from Eastern Lithuania [10].

neol (3 samples) [13]. Linalool dominated in the oil of the above subspecies in Portugal [25], sabinene + germacrene D in France [26] and carvacrol + γ -terpinene + p-cymene in Moldavia [27]. The oil of *O. vulgare* L. (without specification) also contained linalool as a main constituent in India [28]. Linalyl acetate dominated in the oil of *O. vulgare* ssp. *viride* in Iran [29].

Essential oils of *O. vulgare* L. (without specification) growing wild in West Siberia were of β -ocimene type [30] as the oils of wild *O. vulgare* ssp. *vulgare* in Eastern Lithuania [9, 10]. Oils of *O. vulgare* L. cultivated in Leningrad region (seeds from the Moscow Botanical Garden) contained β -caryophyllene as a major constituent [31]. The cultivated plants with white flowers synthesized more monoterpenoids than plants with pink and rose-pink flowers [31].

The most bioactive compounds in essential oils of *O. vulgare* L. are the phenols carvacrol and thymol. Carvacrol exhibits antimicrobial [15, 30, 32–34], antioxidative [32], cytotoxic [15], genotoxic [18], insecticidal [18, 35], acaricidal [35] and antifungal [36, 37] activity. The antimicrobial [30] and antifungal [37] activity of thymol is lower than that of carvacrol. The essential oil of oregano with a low concentration of thymol (0.2%) also is antimicrobially active [30]. The main constituent of the oils under study, β -caryophyllene, is bioactive [34, 35]. It exhibited cytotoxicity against two human carcinoma cell lines and enhanced activity of indole and indole-3-carbinol [38]. β -Caryophyllene is used as an additive for food in USA [38]. Caryophyllene oxide can change the behavior of corn borers [39]. Antimicrobial activity was noticed for essential oils containing large amounts of terpinen-4-ol [34]. Antimicrobial activity [40], allelopathic effect [41] and other forms of influence of 1,8-cineole [42] were investigated.

An analysis of twenty essential oils produced by oregano cultivated in East Lithuania was done in order to determine which major volatile compounds were biosynthesized by plants.

MATERIALS AND METHODS

Plants of cultivated *Origanum vulgare* were collected at full flowering in the following gardens: A – Rokiškis district (Aleknos) and Vilnius (Šeškinė); C – Vilnius district (Nemenėinė and Siponys); D – bought in chemist's shops; F, F' – Vilnius (Antakalnis); G, G' – Vilnius (Deūkinė and Salininkai); H, H' – Vilnius district (Nemenėinė) and Rokiškis district (Aleknos, plants with pink and rose-pink flowers); J, J' – Rokiškis district (Aleknos, plants with white flowers). F, G, H, J – indicated inflorescences; F', G', H', J' – leaves. Earlier investigated plants: B – data of P. R. Venskutonis [7]; E – plants with pink flowers [31]; W – aerial part of wild plants

from Vilnius district [9]; IW, LW – inflorescences and leaves of wild oregano from Eastern Lithuania [10]. Essential oils from each garden were analyzed separately and grouped according to the obtained data.

The samples of plants with pink flowers whose essential oils are presented in A and C columns of Table 1 were collected in August 2000. The plants in Table 2 F – J and F' – J' columns were collected in August 2001.

All samples were dried at room temperature (20–25 °C). Essential oils were prepared by hydrodistillation of 20–100 g of air-dried plants.

Analysis of essential oils was carried out by GC and GC–MS. An HP 5890 II chromatograph equipped with FID and capillary column HP-FFAP (30 m \times 0.25 mm) was used for quantitative analysis. The GC oven temperature was set at 70 °C for 10 min and then programmed from 70 to 210 °C at a rate of 3 °C min⁻¹, using He as the carrier gas (0.7 ml min⁻¹). The injector and detector temperatures were 200 and 250 °C, respectively. Analyses by GC–MS were performed using a chromatograph interfaced to a HP 5971 mass spectrometer (ionization voltage 70 eV) and equipped with a CP-Sil 8 CB capillary column (50 m \times 0.32 mm). The oven temperature was held at 50 °C for 2 min, then programmed from 50 to 180 °C at a rate 3 °C, held for 1 min, then programmed from 180 to 250 °C at a rate 20 °C min⁻¹ and isothermal at 250 °C for 2 min, using He as the carrier gas (1.0 ml min⁻¹). The injector and detector temperatures were 250 °C.

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 indexes on both columns and mass spectra with corresponding data in the literature [43–45] and computer mass spectra libraries (Wiley and NBS 54K).

RESULTS AND DISCUSSION

Essential oils from the plants of cultivated *O. vulgare* L. collected at full flowering in six gardens in August 2000 and 2001 contained low levels of phenols (Tables 1 and 2: thymol 0–3.2%, carvacrol 0–1.3%) than in earlier investigated oils of oregano cultivated in botanical and experimental gardens (Table 1, B) [6, 7]. Experimental data showed that the amount (<5%) of phenols in essential oils of cultivated (Tables 1 and 2) [6, 7] and wild plants [9, 10] markedly differed from the data given in some books (phenols 44%) [1–5].

The major constituent, β -caryophyllene, in more than half of the oils under study was from aerial parts of *O. vulgare* L. plants (Table 1, C), inflorescences (Table 2, G, H) and leaves (G', H', J'). A small habitat of wild plants producing the

β -caryophyllene chemotype essential oil was found in Sapiėginė [9]. Seeds of the wild plants with oil of the above chemotype might originate from plants cultivated in gardens. β -Caryophyllene was the major constituent of some oils from cloned plants in France [11] and in plants cultivated in Leningrad region (Table 1, E) [31]. Volatile oils from Russia contained a notably lower amount of sabinene than did oils of all aerial parts and inflorescences of plants under study (Tables 1 and 2, compare E with C, G, H, J). The chemical composition of the oils under study was rather like that in France.

Sabinene was the main constituent in the oils of aerial parts of oregano plants collected in two gardens (Table 1, A) and caryophyllene oxide – in crushed aerial parts of plants bought in chemist's shops (D). Caryophyllene oxide in the above crushed plants might be synthesized not in the growing plants but during storage in chemist's shops, in paper boxes. Part of volatile monoterpenoids might also be evaporated during storage. The chemical composition of oils from commercial plants (Table 1, D) was most similar to that of oils from the plants with pink flowers collected in a garden of Leningrad region (Table 1, E) [31]. The amounts of the compounds with the caryophyllane skeleton (β -caryophyllene + caryophyllene oxide) were 35.3–35.8% in the oils D under study and 29.6–37.2% in the oils E from Russia. The above oils contained the close quantities of monoterpenoids (Table 1, D – 5.9–12.0% and E – 4.9–10.6%).

Essential oils of inflorescences (Table 2, F) and leaves (F') of the plants from a garden in Antakalnis contained germacrene D as a major constituent (Table 2). The oil of the same type was found in plants growing wild in a small habitat in Rokantiškės (the edge of the forest) at a distance of ~2.5 km from this garden [9]. These wild plants might originate from cultivated plants.

The main constituents in essential oils of the oregano aerial parts collected in 2000 (Table 1, A, C) were sabinene (8.5–22.9%), β -caryophyllene (14.2–18.8%), germacrene D (9.6–11.6%) and caryophyllene oxide (4.3–8.7%). The inflorescence oils (Table 2, G, H, J) contained the same major constituents as aerial part oils. β -Caryophyllene (15.9–21.3%), germacrene D (12.1–15.7%) and caryophyllene oxide (4.7–11.1%) prevailed in the leaf oils G', H' and J'. The oils H, H' and J' contained a larger amount of β -ocimene (10.8–13.9%) than G and G' oils (1.2–4.8%). The main compounds were germacrene D (14.3–16.0%), β -caryophyllene (11.0–11.6%), bicyclgermacrene (6.0–6.4%) and spathulenol (5.0–6.5%) in F and F' oils.

The content of sabinene in the volatile inflorescence oil (Table 2, 5.8–19.5%) was several times

higher than in the leaf oil (3.4–8.1%) of plants from the same garden. An opposite correlation was found for caryophyllene oxide 1.3–3.6% and 2.7–11.1%, respectively. The amount of compounds with caryophyllane skeleton in the oil from plants of the same garden was also larger in leaf than in inflorescence oils (Table 2). The plants under study synthesized more monoterpene hydrocarbons and monoterpenoids in inflorescences than in leaves, except one garden (F, F'). The oils presented in Table 2 contained more sesquiterpene (43.3–62.3%) than monoterpene (5.3–37.0%) hydrocarbons. The largest amount of sesquiterpene hydrocarbons was found in G' leaf oils (61.3–62.3%). The above hydrocarbons made up more than half of the inflorescence oils in three gardens (F, G, 51.6–61.2%).

The largest amount of monoterpene hydrocarbons (26.7–37.0%) and monoterpenoids (34.6–47.3%) contained oils (J, J') from plants with white flowers. The above correlation had been noticed in Russia during investigation of oregano oils from aerial parts of plants [31].

Thirty-three identified constituents of essential oils from the aerial parts of cultivated *O. vulgare* L. plants made up 79.5–97.4%. The amount of 42 identified compounds in inflorescence oils was 85.4–97.9% and in the leaf oils 82.5–86.7%. Mono- and sesquiterpene hydrocarbons made up the major part in essential oils from inflorescences and leaves (except plants bought in chemist's shops) – 65.6–90.4% and 60.5–71.8%, respectively.

Cultivated oregano plants growing in Eastern Lithuania biosynthesized essential oils with sabinene, caryophyllene, caryophyllene oxide or germacrene D as the main constituents and without or with small amounts of phenols than in the earlier investigated plants from the Kaunas Botanical Garden [6] and the experimental garden of Lithuanian Institute of Horticulture [7]. The chemical composition of oregano essential oils under study differed from that presented in the books [1–5], where the content of phenols ($\leq 44\%$) was indicated without any references.

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References

1. Jaskonis J. in Vaistiniai augalai (red. Pipinys J, Jaskonis J, Vaiėiūnienė J). Vilnius, Mintis, 1973: 261–3.
2. Jaskonis J. Aromatiniai augalai. Vilnius, Mokslas, 1989: 104–6.
3. Butkus V, Jaskonis J, Urbonas V, Ėervokas V. Maėpieji miėko turtai. Vilnius, Mokslas, 1987: 144–5.
4. Jaskonis J. Augalai – mūsė gyvenimas. Vilnius, Algi-mantas, 1996: 178–80.
5. Sasnauskas V. Vaistinių augalė ėinynas. 2002: 226–33.
6. Станкявичене НА, Юкнявичене ГК, Моркунас АБ. Тезисы докладов III симпозиума

- “Актуальные вопросы изучения и исследования эфиромасличных растений и эфирных масел”. Симферополь, 1980: 252–3.
7. Venskutonis PR. I krajoje simposium “Naturalne i sintetyczne produkty zapachowe”. Lodz, 1996: 139–49.
 8. Lekavičius A. Lietuvos TSR Flora. T. 5 (red. M. Natkevičiūtė-Ivanauskienė). Vilnius, Mokslas, 1976: 305–6.
 9. Mockutė D, Bernotienė G, Judpientienė A. Phytochemistry 2001; 57: 65–9.
 10. Mockutė D, Bernotienė G, Judpientienė A. J Biochem Syst Ecol 2003; 31: 269–78.
 11. Chalchat JC, Pasquier B. J Essent Oil Res 1998; 10: 119–25.
 12. Pande C, Mathela CS. J Essent Oil Res 2000; 12: 441–2.
 13. Melegari M, Severi F, Bertoldi M, Benvenuti S, Circeta G, Fortunato IM, Bianchi A, Leto C, Carrubba A. Rivista Italiana EPPOS 1995; 16: 21–9.
 14. Leto C, Carrubba A, Trapani P. Atti del Convegno Internazionale: Coltivazione e miglioramento di piante officinali. Trento, Italy. 1994: 343–54.
 15. Sivropoulou A, Papanikolou E, Nikolou C, Kokkini S, Lanaras T, Arsenakis M. J Agric Food Chem 1996; 44: 1202–5.
 16. Kokkini S, Korousou R, Dardioti A, Krigas N, Lanaras T. Phytochemistry 1997; 44: 883–6.
 17. Bocchini P, Russo M, Galletti GC. Rapid Commun Mass Spectrom 1998; 12: 1555–63.
 18. Karpouhtsis I, Paradali E, Feggou E. J. Agric Food Chem 1998; 46: 1111–5.
 19. Skoula M, Gotsiou P, Naxakis G, Johnson CB. Phytochemistry 1999; 52: 649–57.
 20. Karomanoli K, Vokou D, Menkissoglu U, Canstantinidou HI. J Chem Ecol 2000; 26: 2035–48.
 21. D’Antuono DF, Galletti G, Bocchini P. Annal Botany 2000; 86: 471–8.
 22. Mastelic J, Milos M, Jerkovic I. Flav Fragr 2000; 15: 190–4.
 23. Arnold N, Bellomaria B, Valentini G. J Essent Oil Res 2000; 12: 192–6.
 24. Nykanen I, Lebenszen Z. Z. Lebensm Untersuch Forsch 1996; 183: 267–72.
 25. Alves-Pereira IMS, Fernandes-Fereira M. Phytochemistry 1998; 48: 795–9.
 26. Chalchat J, Pasquier B. J Essent Oil Res 1999; 10: 119–25.
 27. Бодруг МВ, Драгалин ИП, Влад ПФ. V Всесоюзный симпозиум “Основные направления научных исследований по интенсификации эфирно-масличного производства”. Кишинев, 1990: 161.
 28. Kaul VK, Singh B, Sood RP. J Essent Oil Res 1996; 8: 101–3.
 29. Afsharypour S, Sajjadi SE, Mahbloombeh EM. Planta Medica 1997; 63: 179–80.
 30. Казаринова НВ, Ткаченко КГ, Мызыченко ЛМ, Сафонова НМ, Ткачев АВ, Каролюк ЕА. Растительные ресурсы 2002; 38(2): 99–103.
 31. Ткаченко КГ, Ткачев АВ. Растительные ресурсы 2002; 38(1): 97–101.
 32. Baratta MT, Dorman HJD, Deans GS. J Essent Oil Res 1998; 10: 618–27.
 33. Ultee A, Gorris LGM, Smid EJ. J Appl Microbiol 1998; 85: 211–8.
 34. Aligianis N, Kalpoutzakis E, Mitaku S, Chinou IB. J Agric Food Chem 2001; 49: 4168–70.
 35. Ahn Y-J, Lee S-B, Lee H-S, Kim G-H. J Chem Ecol 1998; 24: 81–90.
 36. Ferhout H, Bohatier J, Guillot J, Chalchat JC. J Essent Oil Res 1999; 11: 119–29.
 37. Voda K, Boh B, Vrtacnik M, Pohleven F. Inter Biodeter Biodegr 2003; 51: 51–9.
 38. Kubo I, Morimitsu Y. J Agric Food Chem 1995; 43: 1626–28.
 39. Binder BF, Robbins JC. J Agric Food Chem 1997; 45: 980–4.
 40. Clark G, Stuart C. Perfum Flavor 2000; 25: 312–5.
 41. Romagni JG, Allen SN, Dayan FE. J Chem Ecol 2000; 26: 303–13.
 42. Lawler IR, Stapley J, Foley WJ, Eschler BM. J Chem Ecol 1999; 25: 401–15.
 43. Adams RP. Identification of Essential Oil Components by Gas Chromatography / Mass Spectrometry. Allured Publishing Corp., Carol Stream, IL. 1995.
 44. Chung TY, Eiserich JP, Shibamoto T. J Agric Food Chem 1993; 41: 1693–7.
 45. Зенкевич ИГ. Растительные ресурсы 1996; 32(1–2): 45–8.
 46. Зенкевич ИГ. Растительные ресурсы 1997; 33(1): 16–28.

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LIETUVOJE AUGANĖIŲ PAPRASTŲJŲ RAUDONĖLIŲ (*ORIGANUM VULGARE* L.) ETERINIŲ ALIEJŲ CHEMINĖ SUDĖTIS

Santrauka

Dujų chromatografijos ir masių spektrometrijos metodais tirta 20 eterinių aliejų, išskirtų iš kultūrinio paprastųjų raudonėlių pėdų, lapų ir visos antžeminės dalies. Pagrindiniai komponentai (dešimties iš septynių tirtų pėdų aliejų) buvo sabinenas (8,7–19,5%), β-kariofilenas (15,4–24,9%) ir germakrenas D (12,3–16,0%), tokiame pat iš lapų išskirtų eterinių aliejų skaičiuje – β-kariofilenas (15,9–21,3%), germakrenas D (12,1–15,7%) ir kariofileno oksidas (4,7–11,1%). Minėti junginiai vyravo ir tirtuose dešiuose antžeminės augalo dalies eteriniuose aliejuose. Didžioji dalis komponentų (60,5–90,5%) buvo mono- ir seskviterpeniniai angliavandeniliai. Monoterpeninių fenolių timolio ir karvakrolio kiekis lakiuose aliejuose buvo nedidelis (0–3,9%). Šie rezultatai skiriasi nuo pateiktų duomenų švairiose lietuviškose knygose apie vaistąpoles, kur teigiama, kad fenolių kiekiai siekia 44% raudonėlio eterinio aliejaus.

Trisdešimt vienas identifikuotas komponentas sudarė 79,5–97,4% raudonėlio antžeminės dalies eterinio aliejaus. Keturiasdešimt du junginiai, identifikuoti pėdų ir lapų aliejuose, sudarė atitinkamai 85,4–96,4% ir 82,5–86,7%.