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Essential oils of *Hyssopus officinalis* L. cultivated in East Lithuania

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Hyssopus officinalis L. plants were collected in six localities of East Lithuania. Essential oils obtained by hydrodistillation were analysed using GC and GC/MS. Pinocarvone (21.1–28.1%) was the major component in four oils and isopinocamphone (16.8–33.6%) in two oils. Beside pinocarvone, notable amounts of isopinocamphone (11.5–15.9%), β-pinene (7.0–11.4%), germacrene D (3.7–5.5%) and hedycaryol (4.1–4.8%) were found in four oils. The other main constituents in two other oils were pinocarvone (9.0–13.6%), β-pinene (7.2–8.6%) and hedycaryol (4.0–9.1%). Compounds with the pinane carbon skeleton made up 43.6–56.9% of the essential oils. Oxygenated monoterpenes were the most characteristic components of the oils (36.0–50.7%). Sixty-three identified compounds comprised 91.0–97.8% of *Hyssopus officinalis* L. essential oils.

Key words: *Hyssopus officinalis* L., Lamiaceae, composition of essential oils, isopinocamphone, pinocarvone, β-pinene, hedycaryol

INTRODUCTION

Hyssopus officinalis L. belonging to the family Lamiaceae is a perennial herb native to the Mediterranean region, spread also in the European part of Russia, the Caucasus region, middle Asia. Hyssop is cultivated in Lithuania and in many other countries [1–3]. *Hyssopus officinalis* L. has been known as a culinary and medicinal herb for hundreds of years. Hyssop has been employed as a fragrance component in cosmetics. It has been used as a flavour ingredient in many food products and drinks [1, 2]. As a medicinal plant, hyssop has been used as a carminative, emmenagogue, expectorant, stimulant, tonic. Various hyssop preparations have been used as remedies for asthma, rheumatism and some other diseases [2]. Extracts from *H. officinalis* L. possess a slight antioxidant [4] and anti-HIV activity [5]. The essential oils of hyssop have antiseptic properties against bacteria, fungi and viruses [2, 6–9] and show a spasmolytic action [10].

The bioactivity of *H. officinalis* essential oils has usually been attributed to certain components of its oil. The chemical composition can be related to many factors, such as geography, climate, stages of development. Hyssop oils of different phenotypes show a great variability in their chemical composition. Moreover, the composition of oils varies even for the same subspecies [3, 6, 7, 9–24].

Essential oils of hyssops (growing wild or cultivated in various countries) contain large amounts of compounds with a pinane carbon skeleton: mostly β-pinene (2.9–39.6%), pinocamphone (3.2–>62%) and isopinocamphone (3.0–50.5%) [3, 4, 7, 9–24]. Some plant oils (*H. Officinalis* L. ssp. *aristatus, decumbers, angustifolius*) contain 1,8-cineole (40–53%), methyleugenol (38–44%), myrtenol (33%) and linalool (49.0%) as the main constituents [3, 10, 12]. Pinocarvone (26.2–36.3%) is the predominant component in other oils (the subspecies were not indicated) [20, 22].

Authors from Finland characterized hyssop phenotypes by the colour of the corolla (blue, red, white and mixed) [22]. The main components of oils obtained from plants with white, blue and red corollas were, respectively, pinocamphone (46.2–48.7%), isopinocamphone (29.2–32.6%) and pinocarvone (25.3–26.2%). The phenotype with mixed-coloured flowers showed a higher variability in the content of these major components [22].

According to [3], the dominant components in essential oils of cultivated plants of hyssop are pinocamphone (up to 62%), isopinocamphone (up to 43%), β-pinene (up to23%) and limonene (up to 12%). Pinocarvone, germacrene D, α- and β-phellandrene, β-caryophyllene, sabinene, myrcene,

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bourbonene were also determined. The quality of essential oil of hyssop (*Hyssopus officinalis* L. ssp. *officinalis*) is specified by ISO 9841. This standard requires 5.5 to 17.5% of pinocamphone, 34.5 to 50% of isopinocamphone and 13.6–23% of β-pinene. Only hyssop oils cultivated in Serbia and Montenegro [7] and in Bulgaria [12] satisfied ISO requirements.

The aim of this work was to determine the composition of the essential oil of *H. officinalis* cultivated for many years in East Lithuania and compare our results to those previously published.

MATERIALS AND METHODS

Overground parts of cultivated *H. officinalis* L. (blue flower) were collected in July 2008 at full flowering stage in six localities: A – Rinkiškis (Vilnius district), B – Šeškinė (Vilnius), C – Salininkai (Vilnius), D – Rastinėnai (Vilnius district), E – Pavilnis (Vilnius), F – Molėtai district.

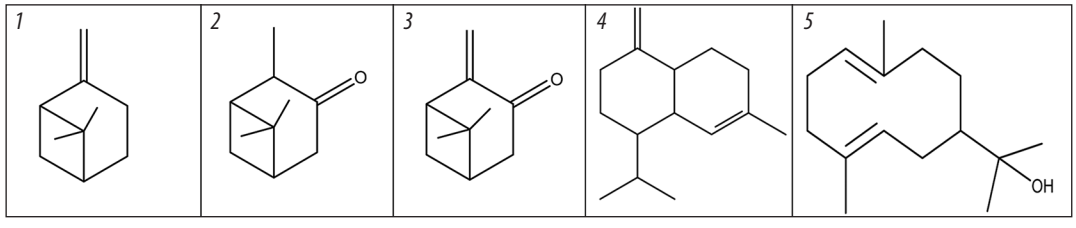
The oils were isolated by hydrodistillation (3 h) of samples dried at room temperature (~20 °C) in an apparatus according to [25] and collected in 2 mL of the hexane : diethyl ether mixture (1 : 1). The plant material and water ratio was 1 : 10. The results are represented as mean values obtained from at least two hydrodistillations of plant material.

Analysis of the essential oils was carried out by GC and GC/MS. An HP 5890 II chromatograph equipped with a FID and a capillary column HP-FFAP (30 m × 0.25 mm, film thickness 0.3 μm) was used for the quantitative analysis. The GC oven temperature was programmed as follows; from 60 °C (isotermal for 3 min) increased to 160 °C at a rate of 5 °C/min (isotermal for 1 min) and then increased to 250 °C at the rate of 10 °C/min, and the final temperature was kept for 3 min. The temperature of the injector and the detector was 250 °C. The flow rate of the carier gas (helium) was 1 mL/min. The GC/MS analysis was performed using HP-5890 gas chromatograph interfaced with a HP 5971 mass spectrometer (ionization voltage 70 eV, scan time 0.6 s, scan range 35–400 Da) and equiped with a DB-5 capillary column (50 m × 0.32 mm × 0.25 μm). The chromatographic conditions were the same.

The percentage composition of essential oils was computed from GC peak areas without the correction factor. The qualitative analysis was based on a comparison of retention times, retention indexes and mass spectra with the corresponding data in the literature [26] and the Willey and NBS 54K computer mass spectra libraries.

RESULTS AND DISCUSSION

The essential oil content in air-dried overground parts of the hyssop, determined by hydrodistillation, did not exceed 0.36%. The results of chemical analysis of hyssop oils are presented in Table. The main compounds in all *H. Officinalis* L. oils was β-pinene, isopinocamphone, pinocarvone, γ-muurolene and hedycaryol (Scheme). Pinocarvone (21.1–28.1%) dominated in four samples (A–D, Table). Isopinocamphone (11.5–15.9%), β-pinene (7.0–11.4%), hedycaryol (4.1–4.8%) and germacrene D (4.1–5.5%) were the other main compounds in these oils. Isopinocamphone was the second and β-pinene was the third main constituent in A–D hyssop oils, similarly as in the solvent extract of hyssop with red corolla from Finland [22]. Pinocarvone (36.3%) as the main constituent was found in oils of *H. officinalis* ssp. *angustifolius* (blue flowers) growing wild in Turkey 1 800 m above sea level [20] and was the dominant compound (25.3–26.2%) in the solvent extract of red-flowered hyssop cultivated in Finland [22].



Scheme. The main compounds of *H. officinalis* essential oils: *1* – β-pinene, *2* – isopinocamphone, *3* – pinocarvone, *4* – γ-muurolene, *5* – hedycaryol

Table. **The chemical composition (%) of *H. officinalis* essential oils***

Compounds	RI	A	B	C	D	E	F
α-Thujene	930	0.1	tr	tr	0.1	0.1	tr
α-Pinene	939	0.8	0.5	0.5	0.5	0.5	0.4
Camphene	954	–	–	tr	–	0.1	–
Sabinene	975	2.5	2.1	1.0	0.9	0.9	0.8
β-Pinene	979	8.5	7.0	10.2	11.4	7.2	8.6
Myrcene	990	2.0	1.9	1.8	1.0	1.2	1.0
Limonene	1029	0.8	1.0	1.1	0.5	0.5	0.7
β-Phellandrene	1030	3.6	3.1	3.4	2.5	1.7	1.5
(Z)-β-Ocimene	1037	–	–	tr	–	1.2	0.3
γ-Terpinene	1059	–	–	–	tr	tr	–
trans-Linalool oxide	1072	–	tr	–	tr	–	–

Table (continued)

Compounds	RI	A	B	C	D	E	F
Terpinolene	1088	0.1	tr	tr	tr	0.2	tr
Linalool	1096	2.5	2.5	2.3	1.3	0.9	1.0
cis-Thujone	1102	–	–	–	–	0.1	0.1
trans-Thujone	1114	–	–	–	–	0.1	tr
allo-Ocimene	1132	–	tr	–	tr	tr	–
trans-Pinocarveol	1139	1.0	1.2	1.1	1.0	0.3	0.5
Pinocarvone	1164	28.1	26.9	24.5	21.1	9.0	13.6
Isopinocamphone	1175	15.5	15.9	14.2	11.5	33.6	16.8
α-Terpineol	1188	0.3	0.4	0.4	0.3	0.1	0.3
Myrtenol	1195	2.6	2.8	3.0	2.7	2.5	2.4
trans-hydroxy-Pinocamphone	1250	tr	0.1	0.1	–	0.3	0.2
(2E)-Decenal	1263	tr	tr	tr	–	–	–
2-Undecanone	1292	–	–	–	0.4	–	–
Methyl myrtenate	1294	tr	0.4	0.3	0.4	0.4	0.4
Myrtenyl acetate	1326	–	–	–	–	3.2	0.7
δ-Elemene	1338	2.6	2.6	2.4	2.5	2.4	3.1
Eugenol	1359	tr	0.1	–	0.1	–	–
α-Copaene	1376	tr	0.1	0.1	tr	0.1	0.1
β-Bourbonene	1388	1.1	1.1	1.5	1.2	1.2	1.5
(Z)-Jasmone	1392	0.2	0.4	0.1	–	–	–
α-Gurjunene	1409	0.6	0.5	0.6	0.8	1.0	1.1
(E)-Caryophyllene	1419	2.9	2.3	3.1	2.2	1.1	4.0
β-Ylangene	1420	0.2	0.3	0.3	0.5	0.3	0.4
β-Gurjunene	1433	0.6	–	–	0.8	0.7	0.8
Neryl acetone	1436	tr	tr	tr	tr	tr	tr
Aromadendrene	1441	0.3	0.5	0.3	0.4	0.5	0.6
trans-Muurola-3,5-diene	1453	–	0.3	–	–	–	–
α-Humulene	1454	1.0	0.8	1.0	–	–	1.1
allo-Aromadendrene	1460	–	–	–	–	–	3.7
9-epi-(E)-Caryophyllene	1466	–	–	–	–	0.6	–
γ-Muurolene	1479	2.7	2.4	2.7	3.0	4.8	6.2
ar-Curcumene	1480	–	–	–	0.9	–	–
Germacrene D	1485	3.8	3.7	4.1	5.5	–	–
β-Selinene	1490	0.3	–	0.4	1.4	–	–
γ-Amorphene	1495	–	0.3	–	–	0.4	0.5
Bicyclogermacrene	1500	2.1	2.5	2.4	2.4	2.4	3.3
γ-Patchoulene	1502	0.1	–	0.4	0.2	–	–
γ-Cadinene	1513	–	–	–	0.1	tr	0.2
δ-Cadinene	1523	–	0.1	–	0.5	0.5	0.7
Hedycaryol	1548	4.2	4.3	4.1	4.8	4.0	9.1
Spathulenol	1578	2.2	2.3	2.0	3.0	2.2	2.1
Caryophyllene oxide	1583	1.5	1.7	1.5	–	0.6	1.2
Viridiflorol	1592	0.1	0.2	0.1	1.9	0.1	0.1
Ledol	1602	0.6	0.7	0.6	1.1	0.7	0.5
10-epi-γ-Eudesmol	1623	0.3	0.4	0.3	0.7	0.7	0.2
γ-Eudesmol	1632	0.5	0.6	0.6	0.8	0.5	1.3
epi-α-Cadinol	1640	0.1	0.1	0.5	0.8	0.8	0.8
β-Eudesmol	1650	1.1	1.3	1.1	1.1	0.6	1.4
Germacra-4(15),5,10(14)-trien-α-ol	1686	0.1	0.1	0.2	0.4	0.2	0.2
Cypertundone	1695	–	–	–	0.3	–	–
Hexahydrofarnesyl acetone	1842	0.4	0.1	0.1	0.4	0.6	0.2
Total		97.8	95.6	94.2	93.3	91.0	93.6
Monoterpene hydrocarbons		18.4	15.6	18.0	16.9	13.5	13.4
Oxygenated monoterpenes		50.2	50.7	45.9	38.4	50.4	36.0
Sesquiterpene hydrocarbons		18.2	17.6	19.2	22.4	16.0	27.2
Oxygenated sesquiterpenes		11.0	11.8	11.2	15.2	11.1	17.1
Pinane skeleton		56.4	54.8	53.7	48.7	56.9	43.6

* RI – retention indexes on nonpolar column; tr – traces; A–F indicate localities of growing.

This component was also identified among the major components of some other oils [22, 24].

Isopinocamphone was a predominant compound in essential oils E and F (33.6 and 16.8%) as well as in the oils from cultivated plants from Serbia and Montenegro [7], Italy 9, 11], blue-flowered plants from Finnland [22]. The content of pinocarvone in oil F (13.6%) was very close to that of isopinocamphone and was about four times lower in oil E (9.0%). β-Pinene (7.2 and 8.6%), hedycaryol (4.0 and 9.1%) and γ-muurolene (4.8 and 6.2%) were among considerable constituents of essential oils E and F.

Thujones (tr – 0.1%) were found only in E and F oils. Germacrene D (4.1–5.5%) was present only in A–D oils (Table). The latter essential oils differed from E and F oils by a higher content of β-phellandrene and linalool and nearly a half as low content of γ-muurolene (Table).

Sixty-three of the identified components made up 91.0–97.8% of the total content of the oil. The main constituents of essential oils were oxygenated monoterpenes (36.0–50.7%). Compounds with a pinane carbon skeleton (α-and β-pinene, trans-pinocarveol, pinocarvone, isopinocamphone, myrtenol, trans-hydroxy-pinocamphone, methyl myrtenate and myrtenylacetate) comprised the largest part of essential oils (43.6–56.9%). All (A–F) oils differed essentially from the ISO standard.

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S a n t r a u k a

Kultūrinė juozažolė (*Hyssopus officinalis* L.) surinkta šešiose vietovėse Rytų Lietuvoje. Eteriniai aliejai išgauti hidrodistiliacijos būdu, analizuoti dujų chromatografijos ir dujų chromatografijos–masių spektrometrijos metodais. Keturiuose aliejuose pagrindinis komponentas buvo pinokarvonas (21,1–28,1 %), o dviejuose – izopinokamfonas (16,8–33,6 %). Kartu su pinokarvonu keturiuose aliejuose rasta daug izopinokamfono (11,5–15,9 %), β-pineno (7,0–11,4 %), germakreno D (3,7–5,5 %) ir hedikariolio (4,1–4,8 %). Kituose dviejuose aliejuose svarbūs komponentai buvo pinokarvonas (9,0–13,6 %), β-pinenas (7,2–8,6 %) ir hedikariolis (4,0–9,1 %). Junginiai, turintys pinano skeletą, sudarė 43,6–56,9 % aliejaus. Deguonį turintys monoterpenai (36,0–50,7 %) buvo būdingiausi eterinių aliejų komponentai. Šešiasdešimt trys identifikuoti junginiai sudarė 91,0–97,8 % juozažolės eterinio aliejaus.