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Utilization of *Lavandula angustifolia* Miller extracts as natural repellents, pharmaceutical and industrial auxiliaries

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Abstract: Essential oils, absolutes and concretes were prepared from the flowers and leaves of the plant *Lavandula angustifolia* Miller cultivated in the Bosphorus region of Istanbul, Turkey. The difference in the chemical composition of the mentioned extracts was investigated and compared by using a combination of capillary GC-MS with the aim of offering them as repellent, pharmaceutical and industrial auxiliaries. The IR-spectra, the yields and the physico-chemical data of the extracts were also analysed.
Keywords: *Lavandula angustifolia* Miller, essential oil, absolute, concrete, repellent, pharmaceutics, industrial.

INTRODUCTION

Essential oils are used in a variety of industries, such as paint, petroleum, mining-manufacturing, food processing-flavouring, drink, pharmaceutical products, perfumes-toiletries, hygiene products and pesticides, *etc*. Commercially, the *Lavandula* genus provides several important essential oils according to the soil and climate.[1,2] *Lavandula angustifolia* Miller is one of the important *Lavandula* species which is found in several regions of Turkey.[3] The essential oils of its flowers and leaves are colourless or light yellow liquids having slightly bitter tastes. The concretes of the flowers are dark green and of the leaves dark yellow solids. The absolutes prepared from the concretes are viscous liquids of green colour from the flowers and yellow from the leaves. Except from cosmetic aspects, they are used as drugs for their antiseptic, analgesic and anti-irritation effects.[4] A previous study[5] did not give well defined compositions of the essential oils, concretes and absolutes.

The aim of this work was to compare the composition and physical, chemical characteristics of the essential oils, concretes and absolutes belonging to the flowers and the leaves of *Lavandula angustifolia* Miller cultivated in Bosphorus, Istanbul, Turkey in order to establish the possibility of their utilization in different fields as natural products.

The essential oils were prepared by steam distillation and the concretes and absolutes by extraction of the flowers and the leaves of this *Lavandula* plant in full bloom. The yield of the steam distillation was 1.9 % for the flowers and 0.66 % for the leaves. The concretes were isolated in 2.15 % yield from the flowers and in 32.4 % yield from the leaves. The yields of the obtained absolutes were 60.5 % from the flowers and 45.2 % from the leaves.

The physical and chemical characteristics of the obtained oils, concretes and absolutes, such as refractive and acid indices, ester contents, optical rotations, were established. The chemical compositions were investigated by capillary GC-MS. All the experimental results are summarized in Tables I–IV.

The plant *Lavandula angustifolia* Miller used in this work is registered at the Faculty of Pharmacy of Istanbul University with the herbarium number A2(E) 59850.

RESULTS AND DISCUSSION

The yields of the essential oils, concretes and absolutes of the flowers and leaves of *L. angustifolia* Miller are given in Table I, the IR spectra in Table II and the physicochemical properties in Table III.

TABLE I. The yields of the essential oil, concrete and absolute of *L. angustifolia* Miller from the Bosphorus region of Turkey

L. angustifolia Miller	Essential oil yield / %	Concrete yield / %	Absolute yield / %
Flower fresh	1.9 (6)	2.15	60.5
Leaf fresh	0.66	32.4	45.2

TABLE II. IR spectra of the essential oil, concrete and absolute of *L. angustifolia* Miller

	IR Spectra (neat, cm^{-1})
Flower essential oil[6]	3445; 3086; 2967; 2928; 2729; 1738; 1674; 1643; 1452; 1413; 1375; 1306; 1236; 1211; 1167; 920; 837; 750; 688; 553; 520; 455; 447; 439
Flower concrete	3438; 3086; 2930; 2861; 1738; 1700; 1643; 1461; 1392; 1284; 1046
Flower absolute	3407; 3086; 2969; 2923; 1738; 1646; 1446; 1376; 1176; 1092; 1053; 1000; 923; 846; 692; 615; 553
Leaf essential oil	3438; 3086; 2969; 2876; 1746; 1684; 1461; 1376; 1230; 1176; 1084; 1061; 992; 923; 846
Leaf concrete	3430; 3086; 2961; 2930; 2861; 1738; 1653; 1461; 1384; 1176; 1123; 1000; 923; 846; 730; 692; 576
Leaf absolute	3384; 3086; 2976; 2938; 1738; 1700; 1645; 1461; 1384; 1330; 1276; 1092; 1046; 884; 676

The chemical compositions of the essential oil of the leaves, the concretes and the absolutes of the flowers and the leaves are summarized in Table IV. Identifica-

tion of the compounds in all the GC-chromatograms was based on the comparison of the retention times with those of authentic samples, comparing their Kovats indices,[7] and on computer matching of their mass spectra with those in Wiley and Nist Libraries.

TABLE III. The physical and chemical characteristics of *L. angustifolia* Miller and their comparison with literature values[5]

	Experimentally obtained values			Literature values (French origin)[5]		
	Oil	Concrete	Absolute	Oil	Concrete	Absolute
Refractive index						
Flowers	1.4519*	1.6759**	1.6755**	1.4601	–	1.467–1.486
Leaves	1.4485*	1.656***	1.675**	–	–	–
Acid index						
Flowers	2.8	15	23.6	0.5	5.6–28.0	4.8–15.0
Leaves	5.2	10.4	12.53	–	–	–
Ester index						
Flowers	42.9	71	89.6	76–136	76–124	157.6
Leaves	35.5	95.2	50.18	–	–	–
Optical rotation						
Flowers	– 0.5° (neat)	+21º13'[(a)]	+45º45'[(b)]	–	–	–
Leaves	+16º25'[(c)]	+8º[(d)]	+3º75'[(e)]	–	–	–

*) 20 ºC, **) 31 ºC, ***) 90 ºC
a (*c* = 0.44), *b* (*c* = 0.22), *c* (*c* = 0.08), *d* (*c* = 0.04), *e* (*c* = 0.4) (all in $CHCl_3$)

TABLE IV. GC-Chromatograms of *L. angustifolia* Miller

RT min	KI	Compound	1 %RA	2 %RA	3 %RA	4 %RA	5 %RA
5.05	940	α-Pinene	0.77	–	–	–	–
5.33	951	α-Fenchene	0.38	–	–	–	–
5.56	975	Sabinene	2.11	–	–	–	–
6.76	980	β-Pinene	0.57	–	–	–	–
7.53	1009	Δ-3-Carene	2.13	–	–	–	–
8.26	1033	1,8-Cineole	49.23	15.99	8.15	9.48	38.43
10.83	1099	Linalool	–	53.73	–	50.00	–
12.24	1141	Camphor	34.67	22.60	9.44	24.10	28.3
12.8	1156	Isoborneol	4.60	2.56	1.72	3.95	1.52
13.1	1160	4-Methyl-1-(methylethyl)-3-cyclohexen-1-ol	–	2.98	–	10.27	–
14.6	1192	α-Terpineol	–	–	–	–	0.32

TABLE IV. Continued

RT min	KI	Compound	1 %RA	2 %RA	3 %RA	4 %RA	5 %RA
16.83	1258	5-Hexenoic acid methyl ester	–	–	–	–	0.91
20.51	1339	trans-Carvylacetate	0.74	–	–	–	0.61
20.9	1344	1,7-Dimethyl-7-(4-methyl-3-pentenyl)-tricyclo[2.2.1.0(2,6)]heptane	0.78	–	–	–	0.45
21.13	1356	6-Methyl-2-methylene-6-(4-methyl-3-pentenyl)-bicyclo[3.1.1]heptane	0.38	–	–	–	–
23.33	1401	2-Isopropyl-5-methyl-9-methylene-bicyclo-1-decene(4.4.0)	1.92	–	–	–	3.96
25	1447	2-Methyl-5,7-dimethylene-1,8-nonadiene	0.38	–	–	–	–
26.1	1477	γ-Muurolene	0.96	–	1.72	–	10.67
27.5	1515	γ-Bisabolene (*Z*)	0.38	–	1.28	–	5.80
40.25	1871	Hexanedioic acid dioctyl ester	–	–	1.07	–	–
42.44	1929	2,4-Dimethyl-7-ethyl-6,8-dioxabicyclo[3.2.1]oct-3-ene	–	–	48.49	–	–
45.66	2000	Eicosane	–	–	1.07	–	–
51.75	2200	Docosane	–	–	9.01	–	–
57.2	2400	Tetracosane	–	–	4.72	–	–
69.1	2800	Octacosane	–	–	0.85	–	–
74.9	3000	Triacontane	–	–	12.45	–	–

1: Leaf essential oil 2: Flower concrete 3: Leaf concrete 4: Flower absolute 5: Leaf absolute
RT: retention time on a DB-5 column in minutes. KI: Kovats retention indices as determined on a DB-5 column using the homologous series of *n*-hydrocarbons. RA: relative area (peak area relative to total peak area).

The plant "Lavandula" is best known for its flowers and the essential oil from its flowers. The essential oil is generally preferred to the concrete and absolute and the lavandula leaves gain, unfortunately, no attention according to our literature studies.

In this work, the aim was to study the plant *Lavandula angustifolia* Miller with the essential oils, concretes and absolutes of its flowers and leaves in terms of natural and industrial aspects.

According to our previous study,[6] the main components of the essential oil of the flowers were 45.09 % linalool, 13.32 % camphor, 8.82 % terpinen-4-ol, 5.81 % 1,8-cineole, 5.22 % borneol, 3.08 % linalyl acetate. In the essential oil of the leaves, however, 49.23 % 1,8-cineole, 34.67 % camphor, 4.60 % isoborneol, 2.13 % Δ-3-carene and 2.11 % sabinene were the main components. Linalool and linalyl acetate, existing in the flowers, were not detected in the leaves. Besides, the increase of the contents of 1,8-cineole and camphor in the leaves compared to the flowers is very characteristic.

The chemical compositions of the prepared concretes and absolutes were changed and the lighter components found in the essential oils were lacking. The flowers gave a

concrete composition of 53.73 % linalool, 22.60 % camphor, 15.99 % 1,8-cineole, 2.98 % 4-methyl-1-(methylethyl)-3-cyclohexen-1-ol and 2.56 % isoborneol and an absolute composition of 50.00 % linalool, 24.10 % camphor, 10.27 % 4-methyl-1-(methylethyl)-3-cyclohexen-1-ol, 9.48 % 1,8-cineole and 3.95 % isoborneol as the main components. The concrete and absolute of the flowers were similar to each other. The lack of linalyl acetate, which is responsible for the characteristic odor, decreases the cosmetic value of these extracts, but their medicinal and insecticidal uses can become important due to the increased percentages of camphor and 1,8-cineole.[8]

The main components of the concrete of the leaves are 48.49 % 2,4-dimethyl-7-ethyl-6,8-dioxabicyclo-[3.2.1]-3-octene, 12.45 % triacontane, 9.44 % camphor, 9.01 % docosane and 8.15 % 1,8-cineole and of the absolute 38.43 % 1,8-cineole, 28.3 % camphor, 10.67 % γ-muurolene, 5.80 % γ-bisabolene. 1,8-Cineole, camphor, isoborneol, 2,4-dimethyl-7-ethyl-6,8-dioxabicyclo-[3.2.1]-3-octene and triacontane are common components of the essential oil, concrete and absolute of the leaves. The increase of the contents of γ-muurolene and γ-bisabolene could make the absolute useful in the pharmaceutical industry.[9] The concrete of the leaves shows a quite different composition with its oxide and long chain hydrocarbon contents, as can be seen in Table IV. It could be utilized for cosmetic and cleaner auxiliaries.[9]

The IR-spectra of the essential oils, concretes and absolutes are similar to each other, all indicating the presence of free alcohols, ether and ester compounds. The alkane, alkyne and alkene stretchings are due to the terpenoid hydrocarbons.

All types of *Lavandula* have been the most favoured aromatics from the time of the Romans until today. *Lavandula* is mostly recommended for its fragrance, as well as its calming and relaxing effects.[10] The essential oil of the flowers in this study with its low linalyl acetate content can not be of great importance in the perfume industry, but with their high percents of linalool, the essential oil, the concrete and the absolute of the flowers could be a great equaliser of the nervous and emotional systems. Therefore, they could be utilized as natural sedatives. This calming property of lavender has been used in folk medicine for insomnia[10] and we also felt this effect during our examination. The essential oil and the absolute of the leaves with their high 1,8-cineole content can act stimulating and can be also expectorant and mucolytic.[8]

All the extracts in this work could be used as antiseptic, antifungal, antiviral, antiinflammatory, analgesic and bactericidal agents, because they are rich in terpenes, alcohols and so the unwanted effects of synhtetic medications could be avoided. These pharmaceutical properties are also known in folk medicine.[10]

The essential oils, concretes and absolutes of the flowers and the leaves could be employed as natural repellents according to their camphor contents. They could be utilized instead of many insecticidal products. In this way the environment could be protected. *Lavandula* has been used since ancient times in sachets or odor lamps against moths and nicotin odor.[11]

Only, the essential oils in this study are rich in constituents, but the mentioned methods can alter the chemical compositions and so the prepared extracts could be offered for several usages.

The *Lavandula angustifolia* Miller used in this study can be utilized as an extraordinarily versatile plant.

EXPERIMENTAL

The plant material was collected from *Lavandula angustifolia* Miller in full bloom cultivated in the Bosphorus region of Istanbul/Turkey. The essential oil of the leaves was obtained by steam distillation for about 60 minutes with a yield of 0.66 %. The *Lavandula* concretes were prepared from fresh flowers and leaves by extraction with petroleum ether (40–60 ºC) for 2 hours in a Soxhlet apparatus. The petroleum ether was evaporated and greenish-yellow solid concretes were obtained. These concretes were refluxed with absolute ethanol for two hours, left at –20 ºC for two days and then filtered from the insoluble organic compounds. The filtrates were evaporated and *Lavandula* absolute of greenish-yellow colour remained as a viscous residue.

The ester and acid indices were established by usual medhods.[12] The IR spectra were recorded using a Jasco FT/IR - 5300 instrument. The refractive index was measured with a Carl-Zeiss instrument model 15884 and the optical rotation with a Bellingham-Stanley Limited Model D Polarimeter.

The chemical compositions of the essential oil, concretes and absolutes were analysed using a 3400 Varian GC-Instrument (FID) with a DB-5-HT column (27 m × 0.25 mm ID) combined with a Finnigan SSQ 7000 MS system. The operating conditions were as follows: injection temperature 230 ºC. Helium carrier gas flow: 1.6 ml/min. Split ratio: 1/100. E. I.:70 eV. Temperature programming: 50 ºC (2 min) – 250 ºC at 2 ºC/min, hold 20 min.

ИЗВОД

КОРИШЋЕЊЕ ЕКСТРАКТА *Lavandula angustifolia* Miller КАО ПРИРОДНОГ РЕПЕЛАНТА И ФАРМАЦЕУТСКЕ И ИНДУСТРИЈСКЕ СИРОВИНЕ

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Добијена су етарска уља од цвета и листа биљке *Lavandula angustifolia* Miller из босфорског региона Истанбула. Испитиване су разлике у хемијским саставима и међусобно упоређене, коришћењем капиларне GC-MS са циљем проучавања могућности њиховог коришћења као репеланта и фармацеутске и индустријске сировине. Анализирани су такође и њихови ИЦ спектри, приноси и физичкохемијске карактеристике екстраката.

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