Comparative Study of *Lavandula angustifolia* Essential Oils Obtained by Microwave and Classical Hydrodistillation

ELENA DANILA[1,4], DURMUS ALPASLAN KAYA[2]*, MARIANA PATRASCU[3], MADALINA ALBU KAYA [1], SYLVIU KUMBAKISAKA[3]

[1]INCDTP - Division Leather and Footwear Research Institute, Collagen Department, 93 Ion Minulescu Str, 031215, Bucharest, Romania.

[2]Department of Field Crops, Faculty of Agriculture, Mustafa Kemal University, 31034, Antakya-Hatay, Turkey.

[3]Chemspeed SRL, 33 Catanoaia Str., 032903, Bucharest, Romania.

[4]University Politehnica of Bucharest, Faculty of Applied Chemistry and Materials Science, Bucharest, 1-7 Str. Gheorghe Polizu Str., 011061, Bucharest, Romania

Lavandula angustifolia which belong to Lamiaceae family with origins in the Mediterranean regionis one of the most important species of Lavandula. The most common method for obtaining the essential oil of lavender is described in the literature is steam hydrodistillation using a Neo-Clevenger, official method described in the European Pharmacopoeia. Another method described in the literature for obtaining oil of lavender is the extraction using microwave. The aim of this study was to obtain lavender essential oils bythese two different methods: classical hydrodistillation using Neo-Clevenger apparatus and microwave–assisted extraction. The yield of extraction was different for the two of methods 0.124 % for classical hydrodistillation and 0.6 % for microwave – assisted extraction. In order to establish and compare the chemical composition of lavender essential oils we used a gas chromatography coupled with mass spectrometry (GC-MS). In the classical hydrodistillation method 44 components with 99.20% yield and in microwave extraction, 46 components with 99.33% yields have been identified. The main compound of essential oil obtained was Linalool. But the other secondary main components are totally different. Even if the obtainedyield for lavender essential oil was bigger in the case of classical hydrodistillation, taking into account the time of extraction was lower for the oil obtained by microwave, it is recommended to continue with this modern method.

Keywords: Lavandula angustifolia, microwave assisted, Neo-Clevenger distillation, essential oil

Lavender belongs to the family *Lamiaceae*which contains 39 species, *Lavandulaangustifolia*being one of the most important species. *Lavandula angustifolia, Lavandula officinalis* or *Lavandula vera* is an evergreen plant with origins in the Mediterranean area (France, Spain, Italy).The name lavender comes from the latin verb *lavare* which means *to wash* or *to clean*. Medicinal properties of lavender have been known since ancient times as specified in the work of Dioscorides De MateriaMedica . The Romans used lavender as an additive for bath and lavender oil in the Middle Ages was one of the most valuable oils used in perfumes and soaps[1].The plant is used in traditional herbal medicine as a sedative, anxiolytic, carminative, antispasmodic, antimicrobial, anti-inflammatory; also presents antioxidant effect, repellent and has a positive effect on memory [2].

Lavender can reach heights of 40-60 cm and forms compact and regular bushes. The bottom of the stem is woody, while the top is green. Lavender has leaves linear or with lanceolate edged and very branched fibrous root. The purple flowers grow pale arranged in circles at the top of the stem and the raw material that contains essential oil (3%) and phenolic acids, flavonoids, anthocyanins, phytosterols, sugars and tannins. Most oil extracted from flowers, is contained in glands on the calyx[3].

The use of lavender essential oil for therapeutic benefit is not new. Lavender essential oils have long been considered to be natural remedies for various ailments. Lavender oil today is used in aromatherapy or massage [4], it has calming, antiflatulence, anticolicproperties [5]. Furthermore, some studies have shown that several constituents of lavender essential oil possess anticancer and antimutagenic properties. The components of essential oils can be encapsulated in different polymers as targeted drug delivery systems and local administrated to different types of cancer such as prostate, lung, liver ones [6 – 8].A multitude of clinical studies have quantified the potential of lavender essential oils in altering the behavior of patients suffering from dementia[9].And also are some studies about insecticidal activity of lavender essential oil [10].

The most common method for obtaining the essential oil of lavender described in the literature is steam hydrodistillation using a Neo-Clevenger, official method described in the European Pharmacopoeia. Using this method M. BelhadjMostefai et al. describe obtaining the essential oil of Lavender flowers: 100 g of fresh plant material was distilled for 3 hours to give a yellow oil, fragrant with a yield of 2%[11].

Another method described in the literature for obtaining oil of lavender is the extraction using microwave: lavender flowers are first subjected to an enzymatic treatment with a Buffer (Na_2HPO_4 . $2H_2O$, 0.2 M and acetic acid, 0.1 M) in different proportions keeping the *p*H constant. Mixtures were stirred in an open vessel for one hour at the desired temperature and then subjected to microwave assisted hydrodistillation (MWHD) to remove the essential oil. During the experiment, the time, temperature, pressure and power were controlled by an operating console. The steam produced in the reactor which carries the essential oil of lavender is directed to Neo Clevenger modified with a siphon tube gradually. The separated oil was stored at 4° C until analysis[12].

Characterization of the chemical composition of lavender oil using GC-MS can lead to different results

* email: dak1976@msn.com

depending on the method of production. The ISO standard 3515: 2002 defines the acceptable ranges for the major components of the essential oil of *L. angustifolia*: 1,8-cineol, 0-15%; Limonene is 0-0.5%; trans-β-ocimene 2-6%; cis-β-ocimene 4-10%; 3-octanone 0-2%; camphor 0-0.5%; linalool 25-38%; linaliyl acetate 25-45%; terpinene-4-ol 2-6%; lavandulol minimum 0.3%; lavandulol acetate minimum 2.0%; α-terpineol 0-4%[13].

In this study we obtain lavender essential oil by two different methods: classical hydrodistillation using Neo-Clevenger apparatus and microwave - assisted hydrodistillation and we characterized the obtained by GC-MS in order toestablish and compare the chemical composition.

Experimental part

Materials and methods

Plant Material

Lavender (*Lavandula angustifolia*) flowers was suppliedbyMEV LOGISTICS CONSULTING SRL, Prahova, beingcollected in July 2016, it was dried at room temperature and shredded before used. Harvesting was performed in optimum phenophases of plant development for extraction of volatile oil.

Obtaining lavender essential oils

Lavandula angustifolia essential oils were obtained by two different methods:

Classical hydrodistillation using Neo-Clevenger apparatus (CLO)

Hydrodistillation was made by a vegetable material dried at room temperature. It was weighed and placed in a round bottom flask with a volume of distilled water (the solvent); mixture was refluxed about 3-4 h, during which the oil was collected in the side arm of the system (having a density less than water, oil separates out of the water). The installation was allowed to stand for about half an hour to prevent the oil to reach room temperature, the oil was dried over anhydrous sodium sulphate and then stored in dark colour (amber) glass bottles and keep to refrigerator (about 4°C) until use for GC/MS analysis. The obtained volatile oil is a clearly liquid, slightly yellowish and with characteristic smell.

2. *Microwave extraction(MLO)*using a microwave - assisted equipment,Minilabotron 2000 consisting of a 2.45 GHz microwave multimode resonant cavity equipped with a built-in CW (continuous wave) industrial generator (switch mode power supply and magnetron) with output power adjustable up to 2,000 W.A single extraction was performed by lavender flowers: the plant was weighed and placed in a round bottom flask with a volume of distilled water; the mixture was refluxed about 15 min, during which the oil is collected on the side arm of the system. During the experiment, the continuous control and monitoring ofthetime, forwardpower, reflectedpower & temperature are achieved via an integrated PLC/digital display: t = 15 min., P = 0.5 kW, T = 96°C. The steam produced in the reactor carrying the essential oil of lavender is directed to a condenser with a Clevenger system. The separated oil was dried over anhydrous sodium sulfate and then stored in dark color glass bottles andkeep in the refrigerator at 4°C until GC/MS analysis.

Characterization by GC-MS of lavender essential oils

Analysis of the lavender essential oils obtained by classical hydrodistillation and the microwave was performed using a Thermo Scientific Focus gas chromatograph equipped with a mass spectrometer, autosampler and TR-5MS (5% phenyl-polisilfenilenesiloxan, 30 m x 0.25 mm inner diameter, film thickness 0.25). The carrier gas was helium (99.9%) with a flow rate of 1 mL/min; ionization energy was 70 eV. Mass range m/z 50-650 amu. Data acquisition was scanning mode. Transfer line temperature of the mass spectrometer was 220° C,the temperature of orifice injection was 220° C. The samples were injected with a split ratio of 250. The injection volume was 1 mL. The temperature of oven was programmed in the range of 50 to 220° C at 3 °C/min. The structure of each compound was identified by comparison of their mass

Table 1

THE EXPERIMENTAL DATA OBTAINED FROM THE EXTRACTION OF OIL OF LAVENDER BY DIFFERENT METHODS

Vegetable material	The method of obtaining	Practical weight (g)	Volume of distillated water (L)	Volume of essential oil (mL)	%
lavender flowers	classical hydrodistillation	500.00	2	0.62	0.124
	microwave extraction	500.00	1.0	3.00	0.600

Characteristic	Value		Method
	CLO	MLO	
Appearance	Clear liquid	Clear liquid	cf. PharmacopeiaBritannica
Color	Pale yellow	Pale yellow	cf. PharmacopeiaBritannica
Specific gravity, g/cm³ at 20°C	0.88	0.876	ISO 279:1998
Refractive index, n_D^{20}	1.449	1.468	ISO 280:1998
Flash point, °C	76	65	ISO/TR 11018:1997

Table 2

PHYSICAL - CHEMICAL CHARACTERISTICS OF LAVENDER ESSENTIAL OIL

spectra (Wiley 9 library). The data were processed using Xcalibur software.

Results and discussions

Yield of extraction lavender essential oils

Essential oil rates obtained by different methodsare given in table 1. The relative rate of essential oil was calculated as equation 1:

$$Essential\ oil\ rate\ (\%) = \frac{g\ of\ extract}{g\ of\ dried\ material} \times 100 \quad (1)$$

The main physical - chemical characteristics of both oils obtained by two extraction methods are shown in table 2.

The literature specified obtaining a lavender essential oil yellow, with a yield of 1.32 % for lavender flowers[14], respective 3% for all aerial parts[15].

The differences from the data specified in the literature for both lavender essential oil can be generated by a number of factors such as soil and climatic conditions, time of harvest, subspecies, and differences related to the extraction method applied (different parameters).

Composition of lavender essential oils

Lavender oil chromatograms obtained by two different methods are shown in figures 1 and 2, and the chemical compositions of essential oilsare shown in table 3:

Table 3 shows that the main components of lavender oil extracted by classical hydrodistillation are: Linalool (44.65%), Eucalyptol (15.44%), Linalyl acetate (13.23%), Camphor (10.63%) and Borneol (4.99%); the different main components are found for microwave-assisted extraction: Linalool (42.62%), β-Fenchyl acetate (18.20%), Linalool oxide (4,26%), Lavandulol(3.66%) and . The same major components are found in the literature data, which demonstrates a high purity of the essential oil obtained by the two methods.

In figure 3 was performed a comparison between main components for both essential oils.

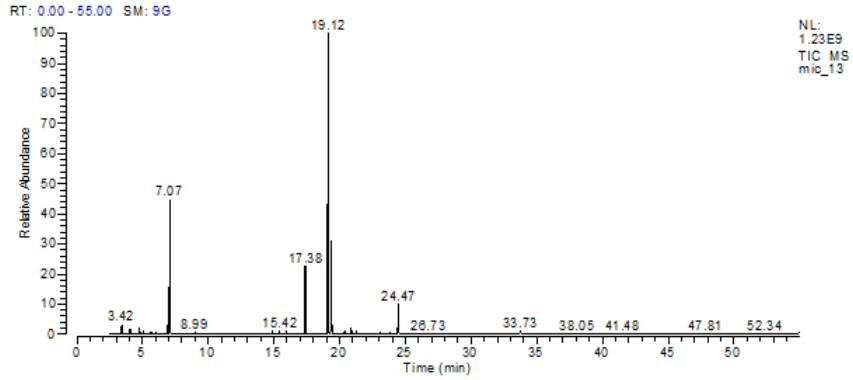

Fig. 1. GC-MS chromatogram of *Lavandula angustifolia*oil obtained by classical distillation

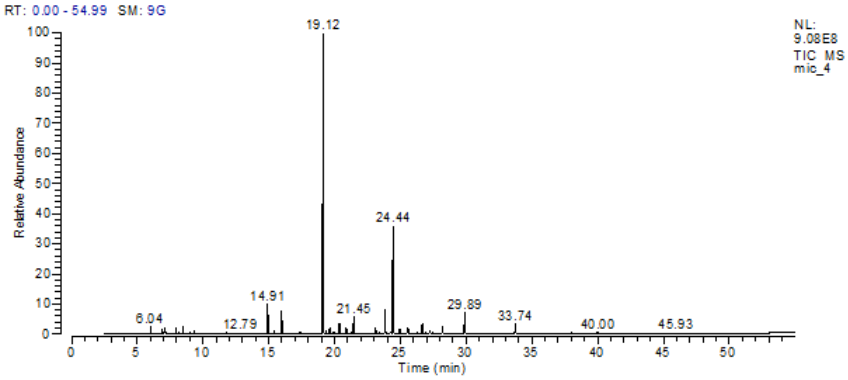

Fig. 2. GC-MS chromatogram of *Lavandula angustifolia* oil obtained by microwave extraction

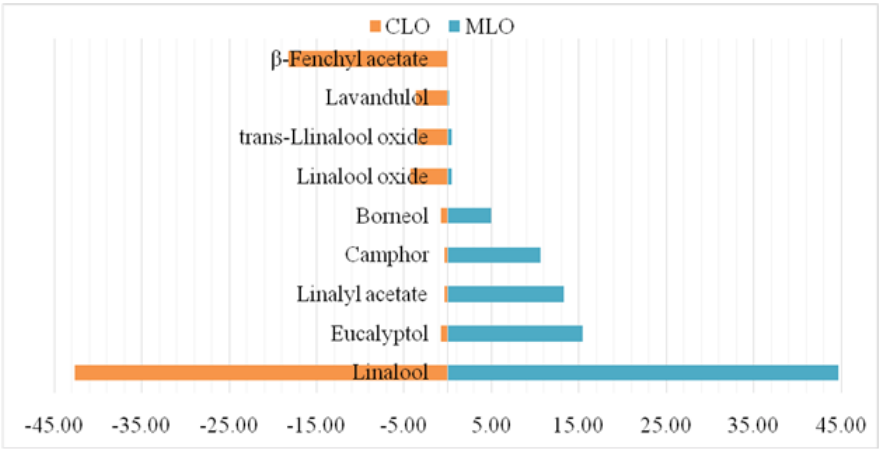

Fig. 3. The main components of *Lavandula angustifolia* oil obtained by CLO (classical hydrodistillation) and MLO (microwave extraction)

Table 3

COMPARISON BETWEEN CHEMICAL COMPOSITION OF *LAVANDULA ANGUSTIFOLIA* OIL OBTAINED BY CLASSICAL HYDRODISTILLATION (CLO) AND MICROWAVE EXTRACTION (MLO)

	Compound Name	SI	RSI	Cas #	CLO	MLO
3,42	α -Pinene	990	994	80-56-8	0.89	-
4,06	Camphene	964	986	79-92-5	0.44	0.13
4,66	Cyclohexane	983	989	110-82-7	0.08	-
4,78	β-Pinene	977	994	127-91-3	0.66	-
5,06	β -Phellandrene	979	983	555-10-2	0.25	-
6,04	β-Myrcene	981	985	123-35-3	0.13	0.77
6,87	Limonene	986	993	5989-54-8	0.90	0.51
7,06	Eucalyptol	986	993	470-82-6	15.44	0.81
7,25	3-Dodecyne	888	895	6790-27-8	-	0.18
7,97	cis-Ocimene	987	992	6874-10-8	0.06	0.66
8,18	3-Dodecyne	883	892	6790-27-8	-	0.20
8,54	3-Octanone	952	987	106-68-3	0.11	-
8,47	trans-β-Ocimene	980	983	3779-61-1	-	0.87
9,00	Benzene, methyl(1-methylethyl)	955	979	25155-15-1	0.23	0.28
9,35	α-Terpinolene	972	980	586-62-9	-	0.32
11,43	Propanoic acid, 2-methyl-, hexyl ester	952	991	2349-07-7	0.07	-
11,79	1-Hexanol	962	981	111-27-3	0.07	0.16
12,79	1-Octen-3-yl acetate	963	988	2442-10-6	0.08	0.16
14,07	Butanoic acid, hexyl ester	974	984	2639-63-6	0.19	-
14,47	Hexyl 2-methyl butyrate	978	994	10032-15-2	0.12	-
14,91	Linalool oxide	979	993	5989-33-3	0.45	4.26
15,42	1-Octen-3-ol	983	985	3391-86-4	0.52	0.50
15,80	cis-Sabinenehysrate	970	987	15826-82-1	0.20	-
15,98	trans-Linalool oxide	965	969	NA	0.43	3.44
17,38	Camphor	973	978	76-22-2	10.63	0.37
19,12	Linalool	983	984	78-70-6	44.65	42.62
19,36	Linalyl acetate	978	988	115-95-7	13.23	0.34
19,63	Isosativene	905	910	NA	0.20	0.85
19,94	Endobornyl acetate	982	985	76-49-3	0.07	0.29
20,16	trans-α-Bergamotene	842	902	13474-59-4	0.06	0.19
20,38	trans-Caryophyllene	968	986	87-44-5	0.52	1.67
20,88	Terpinen-4-ol	984	984	562-74-3	0.94	1.05
21,27	Lavandulyl acetate	966	981	25905-14-0	0.38	0.28
21,45	Hotrienol	959	967	20053-88-7	-	2.62
21,68	Hexyl tiglate	912	969	16930-96-4	0.07	-
23,11	Cryptone	957	960	500-02-7	0.34	1.00
23,39	trans-β-Farnesene	933	958	502-60-3	0.16	0.32
23,50	α-Terpineol	882	895	10482-56-1	0.13	-
23,84	Lavandulol	963	963	498-16-8	0.17	3.66
24,20	Eucarvone	946	950	503-93-5	-	0.33
24,44	β-Fenchyl acetate	981	989	470-08-6	-	18.20
24,47	Borneol	980	986	10385-78-1	4.99	0.79
24,94	Phellandral	919	987	23963-70-4	-	1.05
25,57	Neryl acetate	975	980	141-12-8	0.08	0.16
26,30	trans-Caryophyllene	856	872	87-44-5	0.08	-

26,31	Germacrene-D	931	944	23986-74-5	-	0.31
26,67	Geranyl acetate	988	988	105-87-3	0.08	1.65
26,73	Geranylisovalerate	919	936	109-20-6	0.12	-
26,91	Linalool oxide	892	974	5989-33-3	-	0.15
27,24	Cumaldehyde	931	937	122-03-2	0.09	0.46
28,21	Nerol	977	979	106-25-2	-	1.29
29,03	trans-Anethole	945	971	4180-23-8	0.08	-
29,89	Geraniol	971	972	106-24-1	0.08	3.37
31,75	Junipene	803	809	475-20-7	-	0.12
33,74	Caryophyllene oxide	970	984	1139-30-6	0.66	1.87
34,67	Perilla alcohol	851	873	536-59-4	-	0.14
38,06	Cuminic alcohol	886	952	536-60-7	0.07	0.26
40,00	Isoledene	919	929	95910-36-4	-	0.41
40,09	trans-Isoeugenol	814	847	5932-68-3	-	0.13
44,79	Aromadendrenepoxide	881	904	85710-39-0	-	0.13

** RT = retention time, SI = Isotope patterns, RSI value = Reverse match, CAS = Chemical Abstracts Service*

Conclusions

In the classical hydrodistillation method 44 components with 99.20% yield and in microwave extraction, 46 components with 99.33% yields have been identified. The main compound of essential oil obtained were Linalool. But the other secondary main components were totally different.

In fact, microwave - assisted extraction of essential lavender oil is environmentally friendly, with low solvent rate and the process with lower cost, shorter extraction times and better yields than classical hydrodistillation. Microwave energy may enhance the release of the volatile compounds from the matrix, and high energy flux may enhance their co-distillation. Microwaves cause the glandular walls to quickly rupture, resulting in high extraction efficiency in a shorter time.

From the qualitative point of view, microwave -assisted extraction technique is recommended for obtaining lavender oil having high a percentage of linalool at the expense of low percentages of other monoterpene alcohols.

Scale - up of theproposed technique in industry should also be evaluated from different points of view including cost and energy consumption.

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References

1.PRUSINOWSKA, R., ŒMIGIELSKI, K. B., Herbapolonica, **60**, nr. 2, 2014, p. 56-66

2.LUNGU, C., CORCIOVA, A., SPAC, A., CIOBANU, C., IVANESCU, B., Analele Stiintifice ale Universitatii Al. I. Cuza Iasi, **60**, nr. 2, 2014, p. 11-19

3.SEIDLER-LOZYKOWSKA, K., MORDALSKI, R., KUCHARSKI, W., KÊDZIA, B., BOCIANOWSKI, J.,ActaScientiarumPolonorum Horticulture,**13,** nr.6, 2014, p. 173-183

4.CAVANAGH, H. M. A.,WILKINSON, J. M.,Australian Infection Control, **10**, nr. 1, 2005, p. 35-37

5.GYLLENHAAL C., MERRIT S. L., PETERSON S. D., BLOCK K. I., GOCHENOUR T., Sleep Medicine Reviews, **4**, nr. 2, 2000, p. 1-24

6.BOCAN, E.V., MEDERLE, O, SÂRB, S., MINCIU, R., AGAPIE, D., RAICA, M., Romanian Journal of Morphology and Embryology, **52**, nr. 4, 2011, p. 1215-1218.

7.DJESKA, I.S., CEAUSU, R.A., GAJE, P.N., CIMPEAN, A.M., MEDERLE, O., NICODIN, A., TUDORACHE, V., RAICA, M., Archives of Biological Sciences, **65**, nr. 4, 2013, p. 1599-1604.

8.RALUCA, B.A.,CIMPEAN, A.M., CIOCA, A., CRETU, O., MEDERLE, O., CIOLOFAN, A., GAJE, P., RAICA, M., Asian Pacific Journal of Cancer Prevention, **16**, nr. 11, 2015, p. 4549-4553.

9.WORONUK, G., DEMISSIE, Z., RHEAULT, M.,MAHMOUD, S., PlantaMedica,**77**, 2011, p. 7–15

10.ATTIA, S., LOGNAY, G., HEUSKIN, S., HANCE, T.,Journal of Entomology and Zoology Studies, **4**, nr. 1, 2016, p. 118-122

11.BELHADJ MOSTEFA, M., KABOUCHE, A., ABAZA, I., ABURJAI, T., TOUZANI, R., KABOUCHE, Z., Journal of Materials and Environmental Science, **5**, nr. 6,2014, p. 1896-1901

12.CALINESCU, I.,GAVRILA, A. I., IVOPOL, M., IVOPOL, G. C., POPESCU, M., MIRCIOAGA, N.,Central European Journal of Chemistry,**12**, nr. 8, 2014, p. 829-839

13.***http://www.iso.org/iso/home/store/catalogue_tc/catalogue_detail.htm?csnumber=36253

14.KAYA, D.A., MEMET, I., ELIFE SULTAN, G., SALIHA, K., Rev.Chim.(Bucharest), **63**, no. 8, 2012,p. 749-753

15.INAN, M., KAYA, D.A. ALBU, M. G, Rev.Chim.(Bucharest), **64**, no. 9, 2013, p. 1037.

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