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CHEMICAL CHARACTERIZATION OF ESSENTIAL OIL OF LOCAL ENDEMIC *LAMIUM BILGILII* CELEP (LAMIACEAE)

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Abstract

In this study, the chemical constitution of the essential oil of *Lamium bilgilii* Celep, a local endemic species of Turkey, was studied by GC-MS for the first time. The essential oil was obtained by hydrodistillation method and forty-three compounds were successfully identified representing 98.2% of the oil. Bicyclogermacrene (14.22%) and linalyl acetate (8.33%) and linalool (7.86%) were the most abundant components of the essential oil. *L. bilgilii* essential oil was rich in sesquiterpenes (35.8%) and oxygenated monoterpenes (26.6%). The results showed that bicyclogermacrene was found, for the first time, as a most abundant component in the *Lamium* oils.

Key words: *Lamium bilgilii,* Lamiaceae, essential oil, bicyclogermacrene, GC-MS

Introduction. Depending on the circumscription of the genus, *Lamium* L., the type genus of Lamiaceae (Mint family), consists of 16–40 species [1,2] and contains both annuals and perennials that are widely distributed in the Mediterranean region and temperate Eurasia, where its diversity centre lies in the phytogeographic regions of Mediterranean and Irano-Turanian [3]. There are 24 recognized *Lamium* species, as well as several infraspecific taxa in the World Checklist

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of Lamiaceae and Verbenaceae, according to GOVAERTS et al. [4]. In addition, *L. bilgilii* Celep which is closely related to the *L. garganicum* complex found in Turkey was described in 2017 by CELEP [5].

Some species of *Lamium* are commonly used as food supplement for alimentary, pharmacological and cosmetic purposes [6–9]. *Lamium* species are commonly used in folk medicine worldwide for their antiviral, antispasmodic, astringent, antiseptic, anti-inflammatory, uterotonic properties and are also used in the treatment of many disorders such as leukorrhea, hypertension, paralysis, chronic bronchitis, menorrhagia, prostate and scrofula [7,8,10–14]. In this context, the aim of our study is to find out the composition of essential oil of local endemic *L. bilgilii*. The information concerning the chemical composition of local endemic *L. bilgilii* essential oil has not been reported previously. The current research also provides a comparison of the composition of the oil within the genus *Lamium*.

Material and methods. ***Plant material.*** In 2018, the aerial parts of *L. bilgilii* were collected from the Dirmil-Burdur from its type locality (Fig. 1). Voucher specimens were deposited in the Chemical Ecology Laboratory of Pamukkale University (No: GSE-2016).

Isolation and analysis of essential oil. Using a Clevenger-type apparatus,

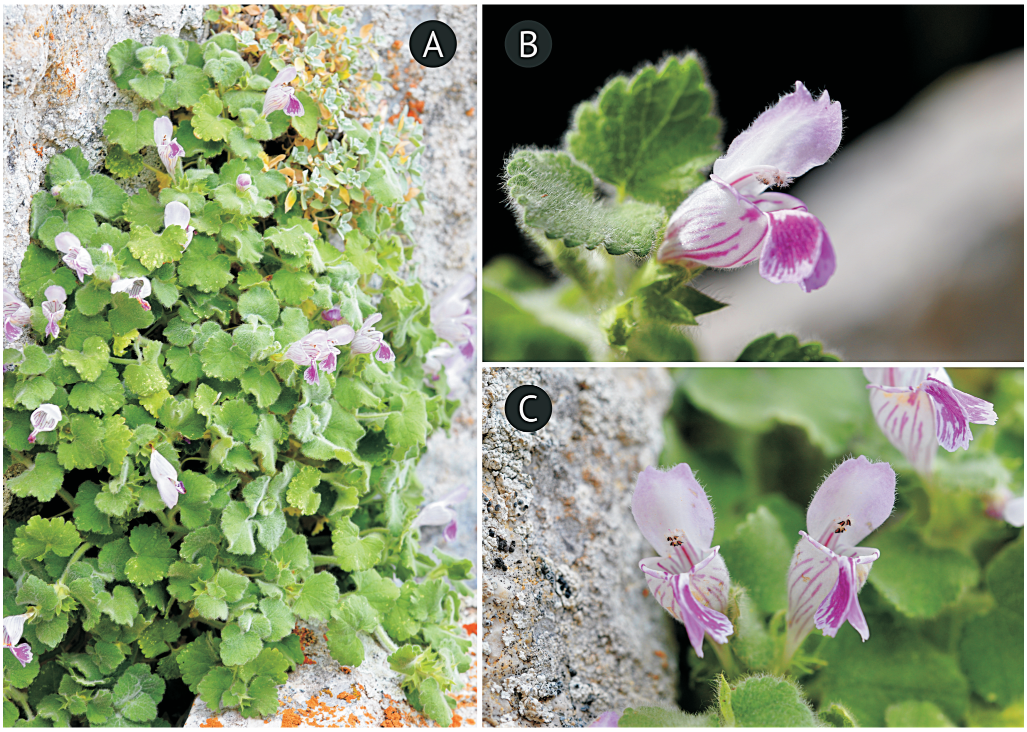

Fig. 1. General appearance of *Lamium bilgilii* in natural habitat at flowering stage. A: Habitat of *L. bilgilii* on calcareous rocks at the type locality; B: Flower; C: Plant with inflorescence

the air-dried plant parts (100 g) were subjected to hydro-distillation for 3 hours to obtain the oil. The obtained essential oil was dried over anhydrous magnesium sulphate and then, the oil was kept in glass vial and stored at 4 °C until analysis.

GC-MS analysis of the oil. The essential oil was analysed using Agilent Technologies HP 7820A GC equipped with 5975 inert MSD in Pamukkale University, Chemical Ecology Laboratory. Fifteen µl diluted oil (1:100 v/v in hexane) was analysed on HP-5 MS capillary column (Hewlett Packard 30 m × 0.25 mm ID, film tickness 0.25 mm). The column temperature was programmed at 50 °C for 3 min then raised to 250 °C at a rate 5 °C/min and kept constant at 250 °C for 5 min. Helium was used as carrier gas, split flow rate was 1.2 mL/min and SCAN technique was used. A series of *n*-alkanes were used to calculate relative retention indices (RRI). The MS ionization energy was 70 eV and a mass range of 30–350. Component identification was performed using WILEY 7 MS and NIST02 libraries. The percentages of the compounds were calculated from GC peaks with normalization method [15].

Results and discussion. The chemical profile of *L. bilgilii* oil was characterized using GC-MS analyses. The essential oil yield of *L. bilgilii* was found as 0.1% (v/w) based on dry weight and the oil was pale yellow. The result of the analysis of *L. bilgilii* essential oil was presented in Table 1. Forty-three components were identified, representing 98.2% of the oils. Bicyclogermacrene (14.22%), linalyl acetate (8.33%) and linalool (7.86%) were determined as major constituents of the oil. The other minor components detected in the essential oil in more than 3% were *trans*-α-bergamotene (6.83%), geranyl acetate (6.04%), *trans*-β-caryophyllene (5.73%), dodecanal (4.52%), spathulenol (4.42%), α-humulene (4.28%) and neryl acetate (3.43%). The results showed that the *L. bilgilii* essential oil was richer in sesquiterpene constituents than monoterpenes and sesquiterpene hydrocarbons were the most abundant components in the essential oil accounting for 35.77%, followed by oxygenated monoterpenes indicated as 26.58%. Considering that there are nearly 25–40 species of *Lamium* in the world, the essential oil composition of genus *Lamium* has not been well explored for all species. A review of the current literature on essential oils of the *Lamium* species revealed the presence of a few studies. The results of our analysis showed that the proportion of compounds presented with the other *Lamium* species had qualitative and quantitative differences and the chemical composition of *L. bilgilii* differed from other *Lamium* species studied. The oil obtained from *L. amplexicaule* L. from Iran was reported to contain a high amount of *trans*-phytol (44.8%) while it was only 1.86% in our study [16]. Another study investigated other species of *Lamium*, rich in germacrene D in *L. purpureum* L. (35.4%), *L. hybridum* Vill. (39.0%), *L. bifidum* Cirillo (34.9%), and *trans*-chrysanthenyl acetate in *L. amplexicaule* (41.1%) [17]. According to MORTEZA-SEMNANI et al. [18], 6,10,14-trimethyl-2-pentadecanone (10.2%) and 4-hydroxy-4-methyl-2-pentanone (9.1%) were among the abundant components of *L. album* L. JONES et al. [19] also studied the chemi-

T a b l e 1

Chemical composition of *Lamium bilgilii* essential oil

No	Compounds[a]	RI[b]	% of the oil
1	α-pinene	932	0.17
2	camphene	944	0.03
3	sabinene	972	0.03
4	β-pinene	978	0.12
5	1-octen-3-ol	986	0.07
6	myrcene	990	1.55
7	α-phellandrene	1007	0.04
8	α-terpinene	1020	0.07
9	d-limonene	1032	0.57
10	*trans*-β-ocimene	1052	1.98
11	γ-terpinene	1063	0.12
12	α-terpinolene	1088	0.35
13	linalool	1098	7.86
14	geyrene	1135	1.55
15	α-terpineol	1181	2.73
16	decanal	1204	0.40
17	*cis*-geraniol	1249	0.92
18	linalyl acetate	1254	8.33
19	prejeijerene	1287	1.20
20	undecanal	1306	1.06
21	δ-elemene	1338	1.79
22	neryl acetate	1360	3.43
23	geranyl acetate	1381	6.04
24	β-elemene	1392	0.47
25	dodecanal	1408	4.52
26	*trans*-β-caryophyllene	1422	5.73
27	*trans*-α-bergamotene	1436	6.83
28	aromadendrene	1456	0.83
29	α-humulene	1461	4.28
30	germacrene-D	1482	1.61
31	bicyclogermacrene	1497	14.22
32	β-curcumene	1514	0.99
33	δ-cadinene	1520	1.25
34	germacrene-B	1558	0.95
35	spathulenol	1576	4.42
36	caryophyllene oxide	1581	1.05
37	viridiflorol	1590	0.78
38	humulene epoxide II	1604	1.65
39	isospathulenol	1633	2.58

Table 1

Continued

40	β-selinene	1716	1.00
41	isobutyl-o-phthalate	1866	2.01
42	manoyl oxide	1993	0.76
43	phytol	2110	1.86
Monoterpene hydrocarbons			5.03
Oxygenated monoterpenes			26.58
Sesquiterpene hydrocarbons			35.77
Oxygenated sesquiterpenes			13.28
Others			17.54
Total identified (%)			**98.20**

[a]Compounds are listed according to their elution by HP-5MS column.
[b]RI: Retention indices calculated against n-alkanes

cal compositions of *L. purpureum* and *L. amplexicaule* essential oils collected from Alabama (USA) and stated that the major compounds found in both species were α-pinene, β-pinene and 1-octen-3-ol. Celep [5] showed that *L. garganicum* L. was the closest species to *L. bilgilii* in terms of morphological characters. However, when evaluated in terms of essential oil constituents, there are obvious differences between the chemical contents of the two species. That is, while *L. garganicum* L. subsp. *laevigatum* Arcangeli has a large proportion of 1,8-cineole (47.5%), citronellal (25.1%) and isoeugenol (11.8%) compounds [20], our study showed that *L. bilgilii* did not have these compounds at all. Germacrene D was frequently reported as the most abundant compound in the essential oils from *Lamium* species. More differences in the content and percentages of major compounds of essential oils of *Lamium* species growing in different regions are shown in Table 2. The differences in chemical composition by different studies may be explained by different ecological parameters, extraction procedure, edaphic factors, genetic features or developmental stage of the plant.

Conclusions. This work is the first report on the essential oil composition of local endemic *L. bilgilii* growing in Turkey. Our study showed that essential oils of *L. bilgilii* was rich in sesquiterpene hydrocarbons and oxygenated monoterpenes. In addition, our results are the first report of the chemical composition of the essential oil obtained from *L. bilgilii* and will also be useful for the chemotaxonomy of *Lamium* species that has not been done yet. Also, the results showed that bicyclogermacrene was found, for the first time, as a most abundant component in the *Lamium* oils. The determination of possible biological activities of the essential oil from *L.bilgilii* could be the next step for further studies.

T a b l e 2

Major components of essential oils from different species of *Lamium* genus (Compiled and modified from SALEHI et al. [6] with some additions)

Plant species	Major components (%)	Subjected parts	Extraction method	Country	Reference
Lamium album L.	α-Terpeniol, linalool, squalene	Leaves	N/A	Ukraine	[21]
	6,10,14-trimethyl-2-pentadecanone(10.2 %), 4-hydroxy-4-methyl-2-pentanone (9.1 %), *n*-Tetracosane (3.9%)	Aerial	Hydro-distillation	Iran	[18]
Lamium album L. subsp. *album*	Germacrene-D (21.2%), caryophyllene oxide (8.2%), (E)-β-caryophyllene (6.6%)	Flower	Hydro-distillation	Poland	[22]
Lamium amplexicaule L.	*trans*-phytol (44.8%), octadecanol (12.0%), hexadecanoic acid (11.8 %)	Aerial	Hydro-distillation	Iran	[16]
	Germacrene-D (18.5–34.9%), (E)-caryophyllene (2.5–11.9%), α-pinene (2.2–16.2%)	Aerial	Hydro-distillation	USA	[19]
	Isophytol (14.8%), 9,12,15-ocadecanoic acid methyl ester (19.2%), 6,10,14-trimethyl-2-pentadecanone (8.0%)	Leaves	N/A	Egypt	[23]
	Germacrene-D (22.3%), camphor (18.1%), Isophytol (14.8%)	Aerial	Hydro-distillation	Iran	[24]
Lamium bifidum Cyr.	Germacrene D (34.9%), sabinene (12.4%), β-caryophyllene (11.5%)	Flowering aerial	Hydro-distillation	Italy	[17]
Lamium bilgilii Celep	Bicyclogermacrene (14.22%), linalyl acetate (8.33%), linalool (7.86%)	Aerial	Hydro-distillation	Turkey	*

T a b l e 2

Continued

Plant species	Major components (%)	Subjected parts	Extraction method	Country	Reference
Lamium garganicum L. subsp. *laevigatum* Arcangeli	1,8-cineole (47.5%), citronellal (25.1%) and isoeugenol (11.8%)	Aerial	Hydro-distillation	Greece	[20]
Lamium hybridum Vill	Germacrene D (39.0%), (Z)-ocimene (8.7%), methyl salicylate (7.5%)	Flowering aerial	Hydro-distillation	Italy	[17]
Lamium maculatum L.	β-caryophyllene (14.8%), caryophyllene oxide (13.8%), Z, E-α-franesene (10.1%)	Aerial	Hydro-distillation	Egypt	[25]
Lamium purpureum L.	Germacrene D (15.0–46.3%), α-pinene (4.1–15.3%), β-pinene (6.3–16.3%)	Aerial	Hydro-distillation	USA	[19]
	Germacrene D (35.4%), β-pinene (26.8%), α-pinene (13.4%)	Flowering aerial	Hydro-distillation	Italy	[17]
	1-Octen-3-ol, *cis*-3-hexen-1-ol, phenethyl alcohol	Aerial	Steam-distillation	Japan	[26]
	Trans-anethole (59.61%), fenchone (6.54%), α-pinene (6.14%)	Aerial	Hydro-distillation	Serbia	[27]

N/A – Not Available, * – In this study.

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