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Composition of the essential oils and in vivo emission of volatiles of four *Lamium* species from Italy: *L. purpureum*, *L. hybridum*, *L. bifidum* and *L. amplexicaule*

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Abstract

The essential oils and the volatiles emitted in vivo by flowers, leaves and bracts of *Lamium purpureum*, *L. hybridum*, *L. bifidum*, *L. amplexicaule* (Lamiaceae) were analyzed by GC-MS and SPME, respectively. All the essential oils were characterized by their high contents of germacrene D. In *L. purpureum* (35.4%), *L. hybridum* (39.0%) and *L. bifidum* (34.9%), it was the main compound, while in *L. amplexicaule* (28.9%), the main constituent was *trans*-chrysanthenyl acetate (41.1%). The SPME analyses showed a pattern typical of volatiles for both the different species and the single plant parts analyzed.

Keywords: *Lamium purpureum*; *L. hybridum*; *L. bifidum*; *L. amplexicaule*; Lamiaceae; Essential oil composition; SPME analysis

1. Introduction

Lamium is a genus belonging to the Lamiaceae family, characterized by annual or perennial herbaceous plants. Some species of this genus are traditionally used as food in some countries, i.e. *Lamium amplexicaule* is one of the ingredients of the so-called "seven spring herbs", a rice porridge traditionally consumed in Japan during the New Year holidays. The oil obtained from the seeds of the same species showed strong antioxidant properties, so its use as a food additive has been proposed (Picuric-Jovanovic, Milovanovic, Budincevic, & Vrbaski, 1997). Also, the hydro-alcoholic extract of another species, *L. album*, showed interesting antioxidant, anti-inflammatory and anti-proliferative properties (Trouillas et al., 2003). Finally, in local folk medicine, all the species are employed as useful remedies in menorrhagia and intermenstrual bleeding, in the treatment of scrofula and for the regulation of sebaceous secretions (Mazza, 2000).

L. purpureum L. (purple dead-nettle) is a herbaceous plant, about 20 cm long, with opposite petiolated leaves; the inflorescence is a dense verticillaster of pinkish flowers; the bracts typically show a dark purple stripe. *L. hybridum* Vill. (cut-leaved dead-nettle) is an ascending branched herb, leafless below the flowers. The bracts are deltoid and obtuse and subtend the crimson flowers. *L. bifidum* Cyr. is an annual herb with erect stems and white flowers with a deeply bifid lower lip. *L. amplexicaule* L. (common henbit, giraffe head) has an ascending stem, purplish basally and greenish above; the lowest leaves are petiolate while the upper ones are reduced to sessile bracts. The pale pink to purple flowers are grouped in verticillasters in the apical half of the stem (Pignatti, 1982).

All these species live on waste ground, lawns, pastures and roadsides.

The essential oils obtained from *Lamium* species are scarcely studied. The most recent papers were published

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in 1996 and 1993: but the latter analyzed the diethyl ether extract of the leaves of *L. maculatum* (Abuzeina, Handjieva, Popov, & Evstatieva, 1993) while the former examined the oil of *L. garganicum* ssp. *laevigatum* (Roussis, Chinou, Perdetzoglou, & Loukis, 1996). The principal constituents were 1,8-cineole, citronellal and isoeugenol. Another paper studied *L. purpureum,* but it dated back to 1976 and non-terpenic derivatives, mainly alcohols and phenols, where found to be the main components of its essential oil (Kurihara & Kikuchi, 1976).

2. Materials and methods

All the samples were collected in the locality Pergole (Arcidosso Municipality, South Tuscany, Italy) at about 600 m above sea level. Here, *Lamium* ssp. grow, together with *Avena fatua*, *Agropyron repens* and *Medicago sativa*. *L. amplexicaule* was found mainly in sunny places, while the other three species preferred shady places.

The flowering aerial parts were collected at the end of March 2003 and were hydrodistilled the next day in a Clevenger-type apparatus for two hours.

The GC analyses were accomplished with a HP-5890 Series II instrument equipped with HP-WAX and HP-5 capillary columns (30 m×0.25 mm, 0.25 μm film thickness), working with the following temperature programme: 60 °C for 10 min, ramp of 5 °C/min up to 220 °C; injector and detector temperatures 250 °C; carrier gas nitrogen (2 ml/min); detector dual FID; split ratio 1:30; injection of 0.5 (μl). The identification of the components was performed, for both the columns, by comparison of their retention times with those of pure authentic samples and by means of their linear retention indices (l.r.i.) relative to the series of *n*-hydrocarbons.

The relative proportions of the essential oil constituents were percentages obtained by FID peak-area normalisation, all relative response factors being taken as one.

GC/EIMS analyses were performed with a Varian CP-3800 gas-chromatograph equipped with a DB-5 capillary column (30 m×0.25 mm; coating thickness 0.25 μm) and a Varian Saturn 2000 ion trap mass detector. Analytical conditions were injector and transfer line temperatures 220 and 240 °C, respectively, oven temperature programmed from 60 to 240 °C at 3 °C/min, carrier gas helium, at 1 ml/min, injection of 0.2 μl (10% hexane solution), split ratio 1:30. Identification of the constituents was based on comparison of the retention times with those of authentic samples, comparing their l.r.i. relative to the series of *n*-hydrocarbons, and by computer matching against commercial (NIST 98 and ADAMS) and home-made library mass spectra built up from pure substances and components of known oils and MS literature data (Adams, 1995; Davies, 1990; Jennings & Shibamoto, 1980; Massada, 1976; Stenhagen, Abrahamsson, & McLafferty, 1974; Swigar & Silverstein, 1981). Moreover, the molecular weights of all the identified substances were confirmed by GC/CIMS, using MeOH as CI ionizing gas.

For SPME analyses, Supelco SPME devices coated with polydimethylsiloxane (PDMS, 100 μm) were used for sampling the headspace of living flowers, bracts and leaves inserted into a 4 ml glass septum vial and allowed to equilibrate for 20 min. After the equilibration time, the fibre was exposed to the headspace for 15 min at room temperature. Once sampling was finished, the fibre was withdrawn into the needle and transferred to the injection port of the GC and GC/MS system, operating under the same conditions as above both for quantification and identification of the constituents, except that the splitless injection mode was used and the injector temperature was 250 °C.

3. Results and discussion

One hundred and five compounds were identified, which accounted for 92.5–99.8% of the total composition. The results are reported in Table 1. All the essential oils obtained from the four *Lamium* species were characterized by their high content of germacrene D. In *L. purpureum* (35.4%), *L. hybridum* (39.0%) and *L. bifidum* (34.9%) it was the main compound, while in *L. amplexicaule* (28.9%) it was the second constituent; in this species the principal compound was *trans*-chrysanthenyl acetate (41.1%). Other important substances were β-pinene (26.8%), and α-pinene (13.4%) in *L. purpureum*, *(Z)*-ocimene (8.7%), methyl salicylate (7.5%) and β-caryophyllene (6.1%) in *L. hybridum*, sabinene (12.4%), β-caryophyllene (11.5%) and α-humulene (6.8%) in *L. bifidum*, and α-pinene (6.8%) in *L. amplexicaule*.

The essential oil of *L. purpureum* was characterized by the high content of hydrocarbons (about 91%), nearly equally divided between monoterpene (43.7%) and sesquiterpene (47.1%) hydrocarbons. Terpene alcohols (linalool and 4-terpineol) were present only in trace amounts; the same is true for non-terpene alcohols, ketones and hydrocarbons (1.1%, 0.8% and 1.5%, respectively). Non-terpene aldehydes reached in this species, globally, 4.1%, while in all the other species they were in lower amounts (0.8–1.5%).

Also, in the essential oil of *L. bifidum*, terpene hydrocarbons were the most represented compounds (91.4%) but, in this case, sesquiterpene hydrocarbons (60.1%) prevailed over monoterpenes (31.4%). Monoterpene alcohols (linalool, 4-terpineol and α-terpineol) summed 1.3%, while non-terpene aldehydes reached 1.1% and non-terpene hydrocarbons 0.6%. Notably, myrcene here constituted 3.9% of the essential oil while, in the other species, it was only a minor constituent (tr-0.8%).

Table 1
SPME and essential oil compositions[a] of *Lamium* ssp.

Constituents	l.r.i.[b]	*Lamium purpureum*				*Lamium hybridum*				*Lamium bifidum*				*Lamium amplexicalule*			
		SPME			Essential oil	SPME			Essential oil	SPME			Essential oil	SPME			Essential oil
		Flowers	Bracts	Leaves		Flowers	Bracts	Leaves		Flowers	Bracts	Leaves		Flowers	Bracts	Leaves	
Furfural	833	–	–	–	tr[c]	–	–	–	–	–	–	–	–	–	–	–	–
(E)-2-Hexenal	855	–	–	–	–	–	–	–	–	–	–	–	0.2	–	–	–	0.5
(E)-3-Hexen-1-ol	856	3.4	–	–	0.8	tr	0.5	–	1.5	1.0	0.2	tr		0.7	0.8	0.7	–
(E)-2-Hexen-1-ol	862	–	–	–	–	–	–	–	–	0.4	–	–		0.4	0.3	–	–
n-Nonane	900	–	–	–	–	–	–	–	–	–	–	–	0.5	–	–	–	–
Heptanal	903	–	–	–	1.5	–	–	–	0.2	–	–	–	0.2	–	–	–	–
α-Thujene	928	tr	–	–	0.1	tr	tr	tr	tr	0.3	–	–	1.1	0.5	0.2	–	0.3
α-Pinene	941	27.6	2.8	13.7	13.4	28.3	2.1	8.8	5.0	3.6	–	tr	4.3	11.0	5. 3	tr	6.8
Camphene	955	0.2	–	–	tr	0.1	–	–	–	–	–	–	–	tr	–	–	–
(Z)-2-Heptenal	964	–	–	–	tr	–	–	–	–	–	–	–	–	–	–	–	–
Verbenene	969	–	–	–	–	–	–	–	–	–	–	–	–	1.3	0.7	tr	0.5
Benzaldehyde	971	–	–	–	tr	–	–	–	tr	–	–	–	–	–	–	–	–
Sabinene	979	0.4	–	–	0.1	0.4	1.8	0.3	4.6	11.0	0.1	0.6	12.4	0.4	0.3	–	0.3
1-Octen-3-one	981		–	–	0.2	–	–	–	tr	–	–	–	tr	–	–	–	–
β-Pinene	983	47.7	4.7	22.0	26.8	39.0	1.3	2.3	3.5	0.7	tr	tr	1.2	0.6	0.4	tr	1.0
3-Octanone	989	0.2	–	tr	0.4	–	–	tr	0.2	tr	–	–	tr	0.3	0.4	–	–
Mmyrcene	992	0.5	–	–	0.8	0.5	0.2	tr	0.4	47.2	1.1	0.2	3.9	0.2	tr	–	tr
3-Octanol	994	0.8	–	–	0.3	0.3	–	–	tr	–	–	–	–	–	–	–	–
n-Decane	1000	–	–	–	–	–	–	–	–	–	–	–	0.1	–	–	–	–
Octanal	1002	–	–	–	–	–	–	–	tr	–	–	–	tr	–	–	–	–
(E)-3-Hexenyl acetate	1004	7.9	14.1	3.1	–	1.6	2.9	0.2	–	2.1	2.9	1.1	–	3.6	8.6	6.7	–
α-Phellandrene	1007	–	–	–	–	–	–	0.4	–	–	–	–	–	–	–	–	–
Hexyl acetate	1011	0.3	tr	1.1	–	–	tr	–	–	tr	tr	tr	–	tr	0.8	0.6	–
(E)-2-Hexenyl acetate	1017	–	–	–	–	–	–	–	–	0.3	–	–	–	0.7	1.1	tr	–
(E,E)-2,4-Heptadienal	1017	–	–	–	0.1	–	–	–	–	–	–	–	–	–	–	–	–
α-Terpinene	1020	–	–	–	–	–	–	–	tr	–	–	–	0.1	–	–	–	–
p-Cymene	1027	–	–	–	–	–	tr	0.3	–	tr	–	–	–	tr	–	–	–
Limonene	1032	1.8	0.4	4.5	3.0	0.5	0.1	0.3	0.3	0.7	0.2	–	0.5	1.2	0.8	1.2	0.6
β-Phellandrene	1034	–	–	–	–	–	–	–	tr	–	–	–	0.1	–	–	–	–
(Z)-Ocimene	1041	–	0.4	–	1.6	3.8	3.8	1.7	8.7	5.5	1.1	1.4	4.8	1.1	–	–	tr
(E)-Ocimene	1052	tr	tr	–	1.3	0.7	1.8	0.9	2.9	1.0	0.7	1.2	2.8	–	0.1	–	0.8
Phenylacetaldehyde	1053	–	–	–	tr	–	–	–	–	–	–	–	–	–	–	–	–
γ-Terpinene	1063	–	–	–	–	–	–	–	0.2	0.1	–	–	0.2	–	–	–	–
cis-Sabinene-hydrate	1070	–	–	–	–	–	–	–	tr	–	–	–	tr	–	–	–	–
Terpinolene	1090	0.2	–	–	tr	–	–	–	tr	tr	–	–	tr	0.1	tr	–	–
1-Undecene	1092	–	–	–	–	–	–	–	–	tr	–	–	–	–	–	–	–

(*continued on next page*)

Table 1 (*continued*)

Constituents	l.r.i.[b]	*Lamium purpureum*				*Lamium hybridum*				*Lamium bifidum*				*Lamium amplexicalule*			
		SPME			Essential oil	SPME			Essential oil	SPME			Essential oil	SPME			Essential oil
		Flowers	Bracts	Leaves		Flowers	Bracts	Leaves		Flowers	Bracts	Leaves		Flowers	Bracts	Leaves	
p-Cymenene	1093	–	–	–	–	–	–	–	–	–	–	–	–	tr	tr	–	–
Linalool	1098	–	–	–	tr	0.3	–	–	2.8	0.1	–	–	0.8	–	–	–	tr
n-Undecane	1100	–	tr	–	0.4	–	–	–	–	–	–	–	–	0.6	0.4	tr	0.3
α-Pinene oxide	1103	–	–	–	–	–	–	–	–	–	–	–	–	–	tr	–	–
Nonanal	1104	–	0.4	1.8	2.0	tr	tr	tr	0.6	–	–	tr	0.4	–	tr	0.8	0.8
allo-Ocimene	1132	–	–	–	–	0.3	0.4	0.5	–	0.7	0.1	0.2	–	–		–	–
trans-Pinocarveol	1141	–	–	–	–	–	–	–	–	–	–	–	–	–	tr	–	–
Pinocarvone	1165	0.3	–	–	–	–	–	–	–	–	–	–	–	–	0.1	–	–
4-Terpineol	1179	–	–	–	tr	–	–	–	0.4	–	–	–	0.4	–	–	–	–
α-Terpineol	1191	–	tr	tr	–	–	–	–	1.3	–	–	–	0.1	–	–	tr	–
Methyl salicylate	1193	–	–	–	–	tr	–	–	7.5	–	–	–	–	–	–	–	–
Methyl chavicol	1197	–	–	–	–	–	–	0.8	–	0.4	0.1	–	–	–	–	–	–
n-Dodecane	1200	–	tr	tr	–	–	–	–	–	–	–	tr	–	0.2	0.1	tr	0.2
Decanal	1204	–	0.8	4.1	0.3	0.2	tr	0.4	–	–	tr	0.1	0.2	–	0.2	1.0	0.2
β-Cyclocitral	1223	–	–	–	0.2	–	–	–	–	–	–	–	–	–	–	–	tr
trans-Chrysanthenyl acetate	1237	–	–	–	–	–	–	–	–	0.1	–	–	–	46.1	52.5	37.8	41.1
(E)-2-Decenal	1264	–	tr	3.6	tr	–	–	–	–	–	–	–	–	0.9	–	–	–
(E)-Anethole	1285	–	–	–	–	–	–	0.9	–	–	tr	–	–	–	–	–	–
trans-Verbenyl acetate	1292	–	–	–	–	–	–	–	–	–	–	–	–	0.2	0.2	tr	–
1-Tridecene	1292	0.3	–	–	tr	–	–	–	–	tr	–	–	–	–	–	–	–
Dihydroedulan IA	1292	–	–	–	–	–	–	–	0.2	–	–	–	–	–	–	–	–
trans-Pinocarvyl acetate	1298	–	–	–	–	–	–	–	–	–	–	–	–	0.1	0.2	tr	0.4
n-Tridecane	1300	0.9	0.5	1.9	0.3	0.1	–	–	–	tr	tr	tr	–	0.9	1.0	1.7	0.5
Undecanal	1308	–	–	–	–	–	–	–	–	–	–	–	0.1	–	–	–	–
4-Vinyl guaiacol	1309	–	–	–	–	–	–	–	0.4	–	–	–	–	–	–	–	–
δ-Elemene	1338	0.2	0.5	tr	0.2	–	1.0	0.9	–	–	tr	tr	–	–	–	–	–
α-Cubebene	1352	–	tr	–	–	–	tr	0.2	–	–	tr	0.1	–	tr	tr	–	–
α-Ylangene	1373	–	–	–	–	–	tr	–	–	tr	tr	tr	–	tr	tr	–	–
α-Copaene	1378	0.2	1.7	0.9	0.8	0.2	0.8	0.7	0.4	0.2	1.6	2.2	0.4	0.7	0.4	1.0	0.6
(E)-b-Damascenone	1382	–	–	–	0.2	–	–	–	0.3	–	–	–	–	–	–	–	–
β-Bourbonene	1386	–	0.6	1.6	0.4	0.3	0.9	1.7	0.8	0.3	0.9	0.5	0.3	0.6	0.8	0.8	1.2
β-Cubebene	1390	–	–	–	tr	tr	–	–	tr	–	–	–	–	0.2	tr	tr	0.3
β-Elemene	1392	0.3	5.0	5.3	1.9	0.2	2.4	3.3	0.7	0.3	3.1	5.2	2.8	0.6	0.6	1.7	0.3
n-Tetradecane	1400	tr	–	0.8	–	–	–	–	–	–	–	–	–	tr	tr	tr	–
α-Gurjunene	1409	–	–	–	–	–	0.2	0.2	tr	–	–	–	–	–	–	–	–
β-Caryophyllene	1420	0.3	2.3	–	0.4	7.9	5.6	6.0	6.1	11.8	7.9	7.0	11.5	4.1	3.8	1.0	3.6
β-Cedrene	1421	tr	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–

β-Ylangene	1421	–	–	–	–	–	–	–	–	–	–	–	–	–	–	0.9	–
β-Gurjunene	1433	–	1.3	tr	tr	0.2	1.4	2.6	0.2	0.7	1.2	1.5	–	0.6	0.6	1.7	0.3
trans-α-Bergamotene	1436	–	–	–	–	–	–	–	–	–	–	–	–	tr	–	–	–
(E)-Geranylacetone	1453	–	tr	tr	–	–	tr	0.2	–	–	–	–	–	–	tr	tr	–
α-Humulene	1455	tr	–	–	–	1.8	1.0	1.2	2.2	5.9	3.6	2.2	6.8	1.4	1.7	0.6	2.0
(*E*)β-Farnesene	1458	0.4	3.7	tr	0.9	6.3	1.2	3.4	0.9	0.6	2.5	3.0	0.8	0.2	0.2	–	0.2
Farnesane	1461	–	–	–	0.8	–	tr	–	–	–	–	–	0.3	–	–	–	1.1
Alloaromadendrene	1462	–	–	–	–	–	2.2	2.2	1.3	–	–	–	–	–	–	–	–
cis-Muurola-4(14),5-diene	1464	–	0.9	–	–	–	–	–	–	–	0.7	0.8	–	0.2	0.2	0.6	–
γ-Muurolene	1478	–	–	–	–	–	0.1	0.3	tr	–	0.3	0.3	–	tr	tr	tr	–
Germacrene D	1482	4.5	54.3	12.6	35.4	5.3	54.4	47.0	39.0	4.2	67.8	67.7	34.9	17.3	12.9	30.9	28.9
β-Selinene	1489	–	–	–	–	–	–	–	–	–	–	–	0.1	–	–	–	–
(Z,E)-α-Farnesene	1491	–	–	–	–	0.2	1.9	1.7	1.1	–	–	–	–	–	–	–	0.7
1-Pentadecene	1492	tr	–	0.8	–	–	–	–	–	–	–	–	–	–	–	–	–
trans-Muurola-4(14),5-diene	1494	–	0.3	–	–	–	–	–	–	–	0.4	0.4	–	tr	0.1	–	–
Valencene	1495	–	–	3.8	–	–	–	–	–	–	–	–	–	–	–	–	–
Bicyclogermacrene	1497	–	tr	–	0.2	–	6.7	5.1	1.4	–	0.3	0.4	0.3	tr	tr	–	0.3
α-Chamigrene	1499	–	–	tr	–	–	–	–	–	–	–	–	–	–	–	–	–
n-Pentadecane	1500	1.0	2.1	5.9	0.8	–	–	–	–	–	–	–	–	0.2	0.3	1.2	0.2
(*E*,*E*)-α-Farnesene	1505	tr	0.5	–	0.4	0.8	1.4	1.2	0.7	0.1	0.2	0.2	0.5	0.3	0.3	0.9	0.6
β-Bisabolene	1507	–	–	–	–	–	–	–	–	–	–	–	–	–	tr	–	–
α-Bulnesene	1508	–	tr	–	2.1	–	–	–	0.8	–	tr	tr	–	–	–	–	–
γ-Muurolene	1511	–	–	–	–	–	0.2	0.4	tr	–	0.2	0.3	–	tr	–	–	–
δ-Cadinene	1521	–	–	–	0.2	0.1	0.4	0.4	0.8	–	0.5	0.6	0.3	0.1	tr	tr	0.5
β-Sesquiphellandrene	1524	–	–	–	–	–	0.3	0.5	–	–	–	–	–	–	–	–	–
Cadina-1,4-diene	1533	–	–	–	–	–	tr	–	–	–	tr	–	–	–	–	–	–
Selina-3,7(11)-diene	1543	–	–	–	–	–	tr	tr	–	–	tr	0.1	–	–	–	–	–
Germacrene B	1560	–	–	–	–	–	0.3	1.0	–	0.5	–	–	1.0	0.1	0.2	0.7	1.7
Caryophyllene oxide	1583	–	–	–	–	–	–	–	0.2	–	–	–	0.1	–	–	–	–
α-Cadinol	1656	–	–	–	–	–	–	–	tr	–	–	–	–	–	–	–	–
n-Heptadecane	1700	–	–	–	tr	–	–	–	–	–	–	–	–	–	–	–	–
Total identified (%)		99.4	97.3	87.5	98.3	99.4	97.3	98.0	97.6	99.8	97.7	97.3	94.5	97.7	96.6	92.5	97.1

[a] Percentages obtained by FID peak-area normalisation.
[b] Linear retention indices (HP-5 column).
[c] tr < 0.1%.

In *L. hybridum*, terpene hydrocarbons reached 82.3% of the whole essential oil; this time sesquiterpene hydrocarbons were twice more abundant than monoterpenes. With respect to the essential oils of *L. purpureum* and *L. bifidum*, in *L. hybridum* the monoterpene alcohols, linalool, 4-terpineol and α-terpineol, reached higher percentages (4.5%). This species is also characterized by the presence of methyl salicylate (7.5%). Another feature of this species is the highest sum of ocimenes, 11.6% vs 2.9%, 0.8% and 7.6% of *L. purpureum*, *L. amplexicaule* and *L. bifidum*, respectively.

The essential oil of *L. amplexicaule* showed the lowest percentage of terpene hydrocarbons (52.6%), constituted by 10.3% of monoterpenes and 42.3% of sesquiterpenes. This species was characterized by the presence of high amounts of *trans*-chrysanthenyl acetate, a monoterpene derivative not detected in the other *Lamium* species.

SPME is a fast, solventless technique that permits the establishment of an equilibrium between the sample matrix, the headspace above the sample, and a stationary phase coated on a fused silica fibre. The adsorbed analytes are then thermally desorbed from the fibre in the injector port of a gas-chromatograph. This technique permits the sampling of the volatiles emitted by living plants in a fast and easy way. By means of this technique we have found that the volatiles emitted by the different living plants parts and the different species were very different (Table 1).

In *L. purpureum*, the three organs analyzed can be distinguished by their different contents of α- and β-pinene: 75.3% in the flowers, 35.7% in the leaves and 7.5% in the bracts. In this organ, the main constituent was germacrene D (54.3%).

Bracts and leaves of *L. bifidum* showed a profile characterized by the common presence of germacrene D, β-caryophyllene, α-humulene and β-elemene among the main produced volatiles. On the otherhand, flowers revealed a different emission pattern because of the presence of high percentages of myrcene (47.2%), β-caryophyllene (11.8%) and sabinene (11.0%).

In the volatiles of *L. hybridum*, pinenes globally reached 67.3% of the total volatiles, while, in bracts and leaves, germacrene D was the main emitted constituent (about 50%).

trans-Chrysanthenyl acetate and germacrene D were the main volatiles emitted by flowers, bracts and leaves of *L. amplexicaule:* altogether they always exceeded 60%. *trans*-Chrysanthenyl acetate was also the main constituent of the essential oil of this species, whereas it was not detected, either in the oils or in the SPME analyses of the other *Lamium* species.

Summarizing, the volatiles emitted by the bracts of *L. bifidum, L. hybridum* and *L. purpureum* were constituted (by more than 50%) of germacrene D; on the otherhand, the bracts of *L. amplexicaule* showed more than 50% of *trans*-chrysanthenyl acetate.

In the flowers of *L. hybridum* and *L. purpureum*, pinenes represented more than 65% of total volatiles; in *L. amplexicaule* and *L. bifidum*, the main chemicals were *trans*-chrysanthenyl acetate and myrcene, respectively.

In the leaves of *L. hybridum* and *L. bifidum*, germacrene D was detected as the principal constituent (47.0% and 34.9%, respectively), whereas *L. amplexicaule* and *L. purpureum* emitted mainly *trans*-chrysanthenyl acetate (37.8%) and α- and β-pinene (35.7%), respectively.

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