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RESEARCH ARTICLE

Chemical composition and antiradical capacity of essential oils from Lebanese medicinal plants

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Essential oils from five wild species growing in Lebanon and used in traditional medicine were obtained by hydrodistillation. Their chemical composition was determined by gas chromatography (GC) and GC–mass spectrometry (GC–MS), and the major constituents were α-pinene in *Juniperus excelsa* M. Bieb. (68.8–86.8%, according to different organs), carvacrol in *Thymbra spicata* L. (65.8%) and *Coridothymus capitatus* (L.) Rchb. f. (47%), pulegone in *Mentha spicata* L. subsp. *condensata* (Briq.) Greuter & Burdet (32.8%) and 1,8-cineole in *Salvia fruticosa* (Mill.) K. Schum. (48.7%). Antiradical capacity of the essential oils was measured *in vitro* by the 2,2′-azino-*bis*(3-ethylbenzothiazoline-6-sulfonic acid) ($ABTS^{\cdot+}$) radical cation and 2,2-diphenyl-picryl hydrazyl (DPPH·) radical assays. A good agreement between the two tests was recorded: *C. capitatus* and *T. spicata* oils showed the highest $ABTS^{\cdot+}$ and DPPH· radical-scavenging activity, possibly due to their high levels of carvacrol, whereas the lowest antiradical capacity was reported for *J. excelsa* oils.

Keywords: essential oils; antioxidant activity; antiradical capacity; Cupressaceae; Lamiaceae; Lebanon; ethnobotany

Introduction

During the past decade, traditional systems of medicine have become increasingly important in view of their safety. Current estimates suggest that, in many developing countries, a large proportion of the population relies heavily on traditional practitioners and medicinal plants to meet primary health care needs. Although modern medicine may be available in these countries, herbal medicines (phytomedicines) have often maintained popularity for historical and cultural reasons. Lebanon, with 2607 plant species, has a particularly plentiful rich and varied flora, which exceeds those of other countries of similar area in the number of genera, species and ecological types. This is due to the moderate climate of the country, which favors the growth of numerous endemic wild or cultivated species, and the varied topography of the land. A large number of taxa is autochthonous and many others were introduced into the region during the evolution of the botanic platform. Many of these plants have been used since ancient times for healing and curing ailments, even chronic diseases. Ethnobotanical and ethnopharmacological surveys showed that 529 species, distributed in 102 botanical families, are still currently used as herbal remedies in Lebanon (1), and other works on the ethnobotany of Lebanon have been published (2–6). In the present study, essential oils were obtained by hydrodistillation from five different medicinal plants collected in Lebanon: *Juniperus excelsa* M. Bieb. (Cupressaceae) and the Lamiaceae: *Thymbra spicata* L., *Coridothymus capitatus* (L.) Rchb.f., *Mentha spicata* subsp. *condensata* (Briq.) Greuter & Burdet (syn. *M. microphylla* K. Koch.) and *Salvia fruticosa* (Mill.) K. Schum. Their chemical compositions were analyzed by gas chromatography coupled to mass spectroscopy (GC–MS) and terpenes were identified. Finally, essential oils were screened for their *in vitro* antiradical capacity by the 2,2′-azino-*bis*(3-ethylbenzothiazoline-6-sulfonic acid) ($ABTS^{\cdot+}$) radical cation and 2,2-diphenyl-picryl hydrazyl (DPPH·) radical assays.

Experimental

Plant species and sample collection

The plant samples were collected from the wild flora of different localities in Lebanon and systematically identified by Dr. Marc El Beyrouthy according to Mouterde (7).

The samples were dried at room temperature (in the shade) for two weeks and deposited in the Herbarium of Botany, Medicinal Plants and Malherbology, Faculty of Agronomy of USEK University, Lebanon.

Hydrodistillation by Clevenger

The volatile oils were distilled for 3 hours according to the standard procedures described in the *European*

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Pharmacopoeia, using a Clevenger-type apparatus. All the oil samples, lighter than water, were dried over anhydrous sodium sulfate and stored under refrigeration (4°C). Oil yields (volume to weight) were determined on an oven-dry weight basis (48 hours at 70°C).

Essential oil analysis

GC analysis

Analytical GC was carried out on a Thermo Electron Corporation gas chromatograph fitted with a HP-5 MS capillary column (30 m × 0.25 mm), 0.25 μm film thickness.

Helium was the carrier gas (0.8 mL/minute). The column temperature was initially kept at 40°C for 5 minutes, then gradually increased to 250°C at 2°C/minute, held for 15 minutes and finally raised to 310°C at 10°C/minute. Flame ionization detection (FID) was performed at 280°C. The analysis was also run using a fused silica HP Innowax polyethylenglycol capillary column (50 m × 0.20 mm, 0.25 μm film thickness). In both cases, helium was used as carrier gas (1.0 mL/minute).

GC–MS analysis

GC/MS was performed using an Agilent gas chromatograph 6890 coupled with Mass Detector 5975. The 7683 B auto sampler injected 1 μL of oil sample each time. GC/MS analysis was carried out using a fused silica Factor DB-5 MS capillary column, measuring 30 m × 0.25 mm internal diameter, film thickness 0.1 μm; the oven temperature program adopted was 35°C with an increase of 5°C/minute until 85°C (20 minutes) and then to 300°C (10 minutes) with an increase of 10°C/minute. Mass spectra were recorded at 70 eV, ion source temperature 310°C, transfer line 320°C, acquisition: full scan 50–400 amu.

GC and GC/MS analysis were also run using a fused silica HP Innowax polyethylenglycol capillary column (50 m × 0.20 mm), 0.20 μm film thickness. Again, helium was used as carrier gas.

Qualitative and quantitative analyses

Most constituents were identified by GC by comparison of their retention indices (RIs) with those of the literature (8, 9) or with those of authentic compounds available in our laboratories. The RIs were determined in relation to a homologous series of *n*-alkanes (C_8–C_{24}) under the same operating conditions. Further identification was made by comparison of their mass spectra on both columns with those stored in NIST and Wiley 275 Libraries, and our home-made library or with mass spectra from the literature (9, 10). Standards of some essential oils of known composition (such as that of *Rosmarinus officinalis* L.; Phytosun' Aroms, Plelo, France) have been injected in similar conditions to check the retention times and the mass spectra. Component relative concentrations were calculated based on GC peak areas without using correction factors.

In vitro *antiradical capacity*

ABTS·$^+$ radical-scavenging assay

The ABTS·$^+$ radical cation-scavenging capacity was determined following Vitalini et al. (11) with slight modifications. The ABTS·$^+$ radical cation was produced by reacting 7 mM ABTS with 2.45 mM potassium persulfate (final concentration) and maintaining the mixture in the dark at room temperature for at least 6 hours before use. The ABTS·$^+$ solution was diluted with ethanol to an absorbance of 0.7 (± 0.02) at 734 nm and equilibrated at 30°C. After the suitable dilution of each sample, 10 μL of essential oil were mixed for 30 seconds with 1 mL of diluted ABTS·$^+$ solution. Ethanol and a standard solution of the synthetic antioxidant 6-hydroxy-2,5,7,8-tetramethychroman-2-carboxylic acid (Trolox) were the negative and positive control, respectively. Their absorbance was read at 734 nm, at room temperature, 20 seconds after the end of the mixing. The percentage of inhibition was calculated $[(ABS_{control\ 734nm} - ABS_{sample\ 734\ nm}/ABS_{control\ 734\ nm}) \times 100]$ and reported as Trolox equivalent antioxidant capacity (TEAC, μmol eq Trolox/mL essential oil).

DPPH· radical-scavenging assay

The DPPH· radical-scavenging capacity was performed following Vitalini et al. (11) with some modifications. Briefly, 10 μL of each sample were added to a 0.07 mM MeOH solution of DPPH· free radical reaching a final volume of 2 mL. After a reaction time of 30 minutes in the dark, at room temperature, the decrease in absorbance at 517 nm was measured by a spectrophotometer (Jenway 6310). A DPPH· solution without essential oil was used as control. The reduction of DPPH· radical was determined as RSA (radical scavenging activity) % = $[(ABS_{control} - ABS_{sample})/ABS_{control}] \times 100$.

Statistical analysis

All experiments were carried out in triplicate and data were expressed as means ± standard deviation (SD). Differences were evaluated by the one-way analysis of variance (ANOVA), and comparison among means was determined according to Fisher's least significant difference (LSD) test. The differences were considered significant at $p<0.05$ and represented by different letters.

Results and discussion

The yields of essential oils obtained by hydrodistillation ranged from 0.38% (v/w, relative to dry weight material) to 1.30%, in *J. excelsa* twigs and *C. capitatus* flowering tops, respectively (Table 1). Both oils of *J. excelsa* twigs and leaves were characterized by the presence of three components, representing 89% and 88.7% of the total identified compounds, respectively, whereas five main constituents were identified in berries, which accounted for 97% of the total oil composition, respectively (Table 1). In all these samples, α-pinene was the most abundant compound, ranging from 68.8% to 86.8%, with cedrol, δ-3-carene and myrcene the second most abundant metabolites in twigs (8.1%), leaves (11.2%) and berries (3.2%), respectively (Table 1). Three constituents characterized the *T. spicata* oil obtained from leaves, representing the 91.15% of the total identified components, with carvacrol (65.8%), γ-terpinene (15.8%) and *p*-cymene (9.5%) the most abundant compounds (Table 1). In essential oil of *M. spicata* subsp. *condensata* flowering tops, six metabolites accounted for 85.7% of the total oil composition, with pulegone (32.8%), *cis*-piperitone oxide (19.2%), piperitenone (13.2%), *p*-menthone (7.7%) and 1,8-cineole (3.7%) the main constituents (Table 1). Six compounds represented 91.3% of the oil of *S. fruticosa* leaves, containing 1,8-cineole (48.7%) and β-caryophyllene (30.8%) the major components (Table 1). Other important metabolites, in this sample were aromadendrene (3.3%), β-pinene (3.2%), α-humulene (2.8%) and δ-3-carene (2.5%) (Table 1). Finally, *C. capitatus* oil obtained from flowering tops was characterized by five compounds accounting for 81.4% of the identified components. The most abundant constituents were carvacrol (47%), thymol (19.9%), γ-terpinene (6.7%) and *p*-cymene (5.7%) (Table 1).

The use of plants in traditional medicine is well reported (1) and their biological activity was investigated in terms of antioxidant power (Table 2). A high variability in the antiradical capacity of the assayed essential oils was reported (Table 2), as well as a good correlation between the two tests ($R^2 = 0.6125$, $p<0.05$; Figure 1). In fact, *C. capitatus* oil showed the highest ABTS· and DPPH· radical-scavenging power, followed by that of *T. spicata* (Table 2). In general, the antioxidant activity of essential oils increased in the order: *J. excelsa* (berries) < *J. excelsa* (leaves) < *J. excelsa* (twigs) < *S. fruticosa* (leaves) < *M. spicata* subsp *condensata* (flowering tops) < *T. spicata* (leaves) < *C. capitatus* (flowering tops), as indicated by both assays (Table 2).

In the past decade, aromatic herbs and spices have received increasing attention for their health-promoting effects, including antioxidant activity mainly ascribed to some their constituents (12–15). Our results showed that the essential oils of *C. capitatus* and *T. spicata*, two species belonging to the Lamiaceae family, were the most bioactive in terms of antiradical capacity among the studied plants. *Coridothymus capitatus* is widely distributed in the Mediterranean area, known in the trade as 'Spanish origanum'. Its essential oil was characterized by four main components – carvacrol, thymol, γ-terpinene and *p*-cymene – as previously reported by other authors (16). Three different chemotypes of *C. capitatus* were individuated according to the thymol:carvacrol ratio, the carvacrol-type, thymol-type and mixed-type, with our essential oil belonging to the first group (17). In accordance with our results, antioxidant activity was previously documented in *C. capitatus* collected in different Mediterranean areas, including Greece, Tunisia and Morocco (18–20). Interestingly, essential oil of *C. capitatus* showed antioxidant activity when tested by two different methods, the β-carotene bleaching test and determination of the oxidative stability of lard (18). Carvacrol and thymol, the main constituents of the oil, were equally effective as antioxidants, independent of the assayed method (18). *Thymbra spicata*, another carvacrol-containing plant known as 'black thyme', grows wild in eastern Mediterranean countries, where dried leaves are used as a spice or herbal tea. Antioxidant activity of *T. spicata* essential oil grown in Turkey has been documented (21).

The existence of different chemotypes, based on qualitative differences within a taxon, is a common trait of most *Mentha* species and hybrids. As a result, mints produce a number of commercially valuable essential oils, i.e. spearmint (*M. spicata*) oil, peppermint (*Mentha* × *piperita*) oil and pennyroyal (*M. pulegium*) oil used in perfumery, cosmetics, pharmaceutical and food industries as active ingredients, or as flavor and fragrance mainly in toothpastes, mouthwashes, cigarettes and alcoholic drinks. The global mint market exceeds billions of dollars, and oil production of this species in the USA is around 500 tons annually (22, 23). The main chemotypes are characterized by the dominant occurrence of linalool, carvone, piperitone oxide and/or piperitenone oxide and pulegone (24–26). As a result of our analyses, we found a pulegone/piperitone-rich chemotype of *M. spicata* subsp *condensata*. Interestingly, pulegone and piperitone are the main components of pennyroyal and peppermint oils, respectively. In previous works, carvone was found to be the main constituent of *M. spicata* subsp *condensata* oil from Turkey, as well as of mints belonging to the Spicatae group (22, 23), even if a Turkish pulegone/piperitone-rich spearmint was reported (27, 28). It is noteworthy that the essential oil composition of aromatic plants depends on their genetic traits, as well as climatic and environmental factors, which may affect aroma constituents (27, 28). *Salvia fruticosa*, commonly known as 'Greek sage', is an aromatic perennial

Table 1. Chemical composition (%) and yield of essential oils obtained by hydrodistillation from different organs of wild Lebanese medicinal plants collected in diverse localities (compounds are listed according to their order of elution).

				Plant name						
				Juniperus excelsa	*Juniperus excelsa*	*Juniperus excelsa*	*Thymbra spicata*	*Mentha spicata* subsp. *condensata*	*Salvia fruticosa*	*Coridothymus capitatus*
Organs				Twigs	Leaves	Berries	Leaves	Flowering tops	Leaves	Flowering tops
Harvest time (2011)				October	October	October	May	July	May	July
Locality				Kartaba	Kartaba	Kartaba	Nahr Ibrahim	Faraya	Nahr Ibrahim	Anfeh
Yield % (v/w)				0.38	1.23	1.17	1.05	0.8	0.9	1.3
R_i[a]	R_i[b]	Compound ID	Identification[c]							
929	1035	*α*-Thujene	R_i, MS, CoGC	–	–	–	1.6	–	–	0.8
938	1076	*α*-Pinene	R_i, MS, CoGC	**78.3**	**68.8**	**86.8**	–	0.4	0.8	0.7
953	1076	Camphene	R_i, MS, CoGC	0.8	1.3	–	0.1	0.2	–	0.2
973	1132	Sabinene	R_i, MS, CoGC	0.4	–	–	–	–	–	–
980	1118	*β*-Pinene	R_i, MS, CoGC	0.6	0.6	**2.5**	0.1	0.9	**3.2**	0.2
993	1174	Myrcene	R_i, MS, CoGC	0.6	0.5	**3.2**	0.8	0.3	1.4	1
1013	1159	*δ*-3-Carene	R_i, MS	**2.6**	**11.2**	**2.4**	0.1	t	**2.5**	0.3
1025	1280	*p*-Cymene	R_i, MS, CoGC	–	–	–	**9.5**	–	–	**5.7**
1030	1203	Limonene	R_i, MS, CoGC	1.1	1.1	**2.2**	1.8	t	–	1.4
1034	1213	1,8-Cineole	R_i, MS, CoGC	–	–	–	–	**3.7**	**48.7**	–
1057	1255	*γ*-Terpinene	R_i, MS, CoGC	t	0.3	0.3	**15.8**	–	–	**6.3**
1094	1402	*α*-Fenchol	R_i, MS	0.3	0.2	–	–	–	–	–
1098	1553	Linalool	R_i, MS	–	–	–	–	–	–	0.7
1105	1430	*α*-Thujone	R_i, MS	–	–	–	–	–	0.4	–
1140	1663	*cis*-Verbenol	R_i, MS	0.1	–	–	–	–	–	–
1143	1532	Camphor	R_i, MS, CoGC	0.3	0.3	t	0.2	t	1.1	–
1152	1683	*trans*-Verbenol	R_i, MS	t	t	t	–	–	–	–
1154	1475	Menthone	R_i, MS	–	–	–	–	**7.7**	–	–
1162	1654	*trans*-Pinocarveol	R_i, MS	0.4	0.2	–	–	–	–	–
1165	1587	Pinocarvone	R_i, MS	0.1	t	t	–	–	–	–
1167	1719	Borneol	R_i, MS, CoGC	–	–	–				1
1176	1611	Terpinen-4-ol	R_i, MS	t	–	–	–	1.1	–	0.6
1182	1864	*p*-Cymen-8-ol	R_i, MS	t	t	t	–	–	–	–
1189	1706	alpha-terpineol	R_i, MS	1	0.5	0.4	–	–	–	–
1217	1725	Verbenone	R_i, MS	0.1	0.1	0.1	–	–	–	–
1217	1845	*trans*-Carveol	R_i, MS	0.1	–	–	–	–	–	–
1233	1662	Pulegone	R_i, MS, CoGC	–	–	–	–	**32.8**	–	–
1248	1758	Piperitone	R_i, MS, CoGC	–	–	–	–	**19.2**	–	–
1284	1597	Bornyl acetate	R_i, MS, CoGC	0.2	0.1	t	–	–	–	–
1293	2198	Thymol	R_i, MS, CoGC	–	–	–	0.1	0.2	–	**19.9**

(*Continued*)

Table 1. (*Continued*)

				Plant name						
				Juniperus excelsa	*Juniperus excelsa*	*Juniperus excelsa*	*Thymbra spicata*	*Mentha spicata* subsp. *condensata*	*Salvia fruticosa*	*Coridothymus capitatus*
Organs				Twigs	Leaves	Berries	Leaves	Flowering tops	Leaves	Flowering tops
Harvest time (2011)				October	October	October	May	July	May	July
Locality				Kartaba	Kartaba	Kartaba	Nahr Ibrahim	Faraya	Nahr Ibrahim	Anfeh
Yield % (v/w)				0.38	1.23	1.17	1.05	0.8	0.9	1.3
R_i[a]	R_i[b]	Compound ID	Identification[c]							
1299	2239	Carvacrol	R_i, MS, CoGC	0.1	–	–	**65.8**	–	–	**47**
1343	1874	Piperitenone	R_i, MS	–	–	–	–	**13.2**	–	–
1352	1466	*α*-Cubebene	R_i, MS	–	–	–				
1369		Piperitenone oxide	R_i, MS					**9.1**		
1382	1547	*β*-Cubebene	R_i, MS	–	–	–	t	t	–	–
1411	1568	*α*-Cedrene	R_i, MS	–	1.5	–	–	–	–	–
1415	1612	*β*-Caryophyllene	R_i, MS, CoGC	0.7	–	–	t	1.1	**30.8**	**2.6**
1435	1573	*trans*-*α*-Bergamotene	R_i, MS	–	t	–	–	–	–	–
1437	1628	Aromadendrene	R_i, MS	–	–	–	1.3	–	**3.3**	–
1455	1689	*α*-Humulene	R_i, MS	–	–	–	t	0.3	**2.8**	–
1463	1661	allo-Aromadendrene	R_i, MS	–	–	–	–	–	0.3	0.2
1477	1726	D-Germacrene	R_i, MS	0.6	–	t	0.3	–	–	–
1487	1679	*α*-Amorphene	Ri, MS	–	–	–	–	–	0.1	–
1491	1756	Bicyclogermacrene	R_i, MS	–	–	–	t	0.2	–	–
1508	1741	*β*-Bisabolene	R_i, MS	–	–	–	–	–	–	0.5
1515	1776	*δ*-Cadinene	R_i, MS	0.3	0.6	t	t		0.1	0.4
1577	2008	Caryophyllene oxide	R_i, MS, CoGC	–	t	–	–	–	–	–
1580	2152	Spathulenol	R_i, MS, CoGC	–	–	–	0.3	–	–	–
1604	2160	Cedrol	R_i, MS, CoGC	**8.1**	**8.7**	t	–	–	–	–
Monoterpene hydrocarbons				85.2	84.1	97.3	29.9	1.9	7.9	16.6
Oxygenated monoterpenes				4.3	2.7	0.8	66.1	86.9	50.2	70
Sesquiterpene hydrocarbons				1.5	2.1	0	1.8	1.6	39.9	4.6
Oxygenated sesquiterpenes				8.1	8.8	0	0.3	–	–	–
Total identified				96.9	97.7	98.1	98.1	90.5	98	91.2

Notes: Bold numbers indicate the percentages higher than 2% to show main components. [a]Retention index on a HP-5MS column; [b]Retention index on an Innovax column; [c]R_i, retention index identical to bibliography; MS, identification based on comparison of mass spectra; Co-GC, retention time identical to authentic compounds; t, trace, <0.05.

Table 2. *In vitro* antiradical capacity of essential oils measured by the 2,2′-azino-*bis*(3-ethylbenzothiazoline-6-sulfonic acid) ($ABTS^{\cdot+}$) radical cation and 2,2-diphenyl-picryl hydrazyl (DPPH·) radical assays.

Essential oil	ABTS (μmol eq Trolox/mL)	DPPH, RSA[1] (%)
Juniperus excelsa (twigs)	0.54 ± 0.01a[2]	8.7 ± 0.45a
Juniperus excelsa (leaves)	0.34 ± 0.01b	4.0 ± 0.29b
Juniperus excelsa (berries)	0.11 ± 0.01c	1.3 ± 0.09c
Thymbra spicata (leaves)	286 ± 22.9d	90.9 ± 6.27d
Mentha spicata subsp *condensate* (flowering tops)	7.35 ± 0.03e	48.7 ± 2.9e
Salvia fruticosa (leaves)	4.27 ± 0.12f	33.2 ± 1.66f
Coridothymus capitatus (flowering tops)	874.6 ± 69.9g	92.7 ± 5.42d

Notes: [1]Radical-scavenging activity. [2]Results are mean ± standard deviation of three independent analyses; different letters indicate means significantly different at $p<0.05$ (Fisher's least significant difference test).

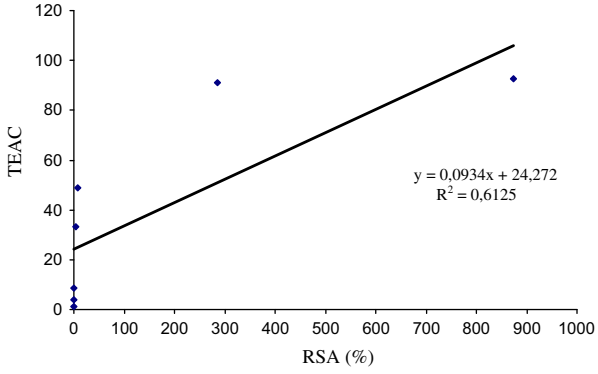

Figure 1. Correlation analysis based on simple linear regression at the 95% confidence level between antiradical capacity of essential oils determined by $ABTS^{\cdot+}$ [2,2′-azino-*bis*(3-ethylbenzothiazoline-6-sulfonic acid)] (TEAC, mmol eq trolox/L wine) and DPPH· (2,2-diphenyl-picrylhydrazyl) (RSA, radical scavenging activity %) radical-scavenging assays.

herb of economic importance endemic of the eastern Mediterranean. Our results are in agreement with other studies reporting 1,8-cineole as the major component of its essential oil (29). In our experimental conditions, the antiradical capacity of *M. spicata* subsp *condensta* and *S. fruticosa* oils was significantly higher compared with *J. excelsa* samples. The antioxidant activity of essential oil of *M. spicata* from Pakistan was reported, even if, to the best of our knowledge (30), no information on *M. spicata* subsp *condensata* is available. Previously, some authors reported that *S. fruticosa* essential oil exhibited high antioxidant power, compared with commercial antioxidants, and inhibitory activity on lipid oxidation (31). Variations in the yield and composition of *S. fruticosa* oil were also recorded according the plant development stage (32). In *J. excelsa* essential oils obtained from different plant parts, α-pinene was the most abundant constituent, as previously reported in Turkish samples (33). Moreover, oil from berries was richer in α-pinene than the leaf oil, in accordance with data on two different *J. excelsa* subspecies grown in Iran (34). Interestingly, in *J. excelsa* subsp *polycarpos*, the antioxidant activity of berry oil was lower than the leaf oil, in agreement with our results (34).

Conclusion

The antioxidant potential of Lebanese species belonging to the Lamiaceae family was documented and confirmed (35). Oxygenated monoterpenes, i.e. carvacrol, 1,8-cineole and pulegone, represented the dominant fraction of essential oils obtained from these plants (*C. capitatus*, *T. spicata*, *M. spicata* subsp *condensata* and *S. fruticosa*). Therefore, carvacrol and other oxygenated monoterpenes may be responsible for the antiradical capacity measured in oils from Lamiaceae plants (36). Conversely, monoterpene hydrocarbons were the most abundant components of essential oils from *J. excelsa* (Cupressaceae), and α-pinene seems to be less relevant in terms of antiradical capacity of *J. excelsa* oils.

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