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RESEARCH ARTICLE

QUANTITATIVE ORGANIC COMPOSITION AND ANTIOXIDANT POTENTIAL OF THE ESSENTIAL OIL FROM *ORIGANUM SYRIACUM* L. (LAMIACEAE) ACCLIMATED IN CÔTE D'IVOIRE

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ABSTRACT

Objective: This study aims to establish the quantitative organic composition, and to appreciate the antioxidant potential of the essential oils (EOs) from the fresh and dried leafy stems of *Origanum syriacum* acclimated in Côte d'Ivoire.
Methods: The chemical composition determined by GC (Ir), GC-MS and 13C-NMR showed that the species belongs to the carvacrol family. The antiradical activity was evaluated with respect to the radical DPPH$^{\bullet}$, (2,2-diphenyl-1-picrylhydrazyl).
Results: In total 31 compounds have been identified accounting for 98.1-99.4% of the total composition. The major compound was carvacrol (61.5-69.8%) following by γ-terpinne (11.3-14.5%). In vitro antioxidant test showed that essential oil from *O. syriacum* had an antioxidant potential nevertheless lower than that of ascorbic acid (vitamin C).
Conclusion: This study highlights chemical composition of the essential oil from *O. syriacum* and an interesting antioxidant potential. These data would justify various traditional uses in therapeutic.

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INTRODUCTION

Origanum syriacum L. belonging to Lamiaceae family and native to eastern Mediterranean, southern Europe, and western Asia. It is a perennial species, reaching 0.8-1 m in height, and is sub-frutescent at the base. Stems and twigs are erect and covered with spreading hairs. The leaves are petiolate, often with very dense hairs, the blade (1-3 cm) is over, usually obtuse, sometimes subacute, margin entire or infrequently, wavy, pubescent, more or less greyish, and very prominent veins (Mouterde, 1983). In traditional medicine, *O. syriacum* is used in the treatment of respiratory, gastrointestinal and urinary diseases. In gastronomy, *O. syriacum* is employed as a spice and preservative (Daouk, 1995). The EO of wild and cultivated *O. syriacum* , from the countries of North Africa and Mediterranean, had been described in several studies. It turns out that the main constituent of this EO was either thymol or carvacrol. Indeed, carvacrol composition was detected in EO of plant from three populations of *O. syriacum* (69.8, 63.8 and 53.8%) out of 11 studied by Syrian researchers, and thymol chemotype was found in a population of the same species (57.6%) (Lucas, 2009). Also, a comparative study conducted by Lebanese researchers, has demonstrated that the carvacrol chemotype was specific to EO extracted from the cultivated leaves, when that from leaves of the wild species was predominated by thymol (Zein, 2011). In addition, the literature indicates that the chemical composition of EO from *O. syriacum* can be dominated on the one hand by carvacrol-thymol pair (Youssef, 2011 and Lucas, 2009), and on the other hand, by mixtures of one of the main constituents with other constituents (Baser, 2003 and Arnold, 2011). Regarding the pharmacological properties of EO of *O. syriacum* leaves, it is reported to be antimicrobial (Bektas, 2004), antioxidant (Safwan, 2012), antifungal (Lakis, 2012), antiprotozoal (Khalil, 2013), and additionally exhibits leukemic cytotoxicity (Basim, 2014). To our knowledge, investigations on EO of *O. syriacum* grown in tropical climate especially in West Africa are not referenced to date. Consequently, the present work whose objective was to determine the chemical composition and to appreciate the potential antioxidant character of EO

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from leafy stems of *O. syriacum* acclimated in Côte d'Ivoire, must contribute to fill that void.

MATERIALS AND METHODS

Plant material and hydrodistillation: Fresh leafy stems of *O. syriacum* were harvested on June 30, 2016 at a food crop site in the vicinity of Riviera M'Pouto in the District of Abidjan. Plant material was authenticated by Dr Malan Djah François (Laboratoire de Botanique, Université Nangui Abrogoua). Fresh organs were divided into 3 lots of equal mass (850 g) before being submitted to hydrodistillation for 3 h using a Clevenger-type apparatus. The leafy stems of lot N°I were used fresh, and those of lots N°II and N°III were used after drying respectively for 21 and 42 days in a well-ventilated enclosure. The oil samples were dried over anhydrous sodium sulfate and stored in refrigerator (4°C) in hermetically sealed brown glass vials before analysis.

Analytical GC: Analyses were carried out with a Clarus 500 Perkin Elmer (Perkin Elmer, Courtaboeuf, France) apparatus equipped with two flame ionization detectors (FID), and two fused-silica capillary columns (50 m x 0.22 mm i.d., film thickness 0.25 µm), BP-1 (polymethylsiloxane) and BP-20 (polyethylene glycol). The oven temperature was programmed from 60°C to 220°C at 2°C/min. and then held isothermal at 220°C for 20 min; injector temperature: 250°C; detector temperature: 250°C; carrier gas, helium (0.8 mL/min); split: 1/60; injected volume: 0.5 µL. The relative contents of the oil constituents were expressed as percentage obtained by peak-area normalization without using correction factors.

GC-MS Analysis: The essential oils were analyzed with a Perkin-Elmer TurboMass detector (quadrupole), directly coupled to a Perkin-Elmer Autosystem equipped with a fused-silica capillary column (50 m × 0.22 mm i.d., film thickness 0.25 µm), BP-1 (dimethylpolysiloxane). Carrier gas, helium at 0.8 mL/min; split: 1/60; injection volume: 0.5 µL; injector temperature: 250°C; oven temperature programmed from 60°C to 220°C at 2°C/min. and then held isothermal (20 min); Ion source temperature, 250°C; energy ionization, 70 eV; electron ionization mass spectra were acquired over the mass range 40-400 Da.

NMR Analysis: All ^{13}C NMR spectra were recorded on a Bruker AVANCE (Bruker, Wissembourg, France) 400 Fourier Transform spectrometer operating at 100.623 MHz for ^{13}C, equipped with a 5 mm probe, in $CDCl_3$, with all chemical shifts referred to internal tetramethylsilane (TMS). ^{13}C NMR spectra were recorded with the following parameters: pulse width (PW), 4 µs (flip angle 45°); acquisition time, 2.7 seconds for 128 K data table with a spectral width (SW) of 24,000 Hz (240 ppm); digital resolution, 0.183 Hz/pt. The number of accumulated scans was 3000 for the oil samples (about 50 mg of essential oil in 0.5 mL of $CDCl_3$).

Identification of individual components: component identification was based on: (a) comparison of their GC retention indices (RI) on polar and apolar columns determined relative to the retention times of a series of *n*-alkanes (C7-C28) with linear interpolation (Target Compounds software from Perkin Elmer) with those of authentic compounds or literature data (König, 2001), (b) on computer matching with a laboratory-made and commercial mass spectral libraries and comparison of spectra with literature data (McLafferty, 1994 and Adams, 2001), (c) on comparison of the signals in the ^{13}C-NMR spectra of essential oils with those of reference spectra compiled in the laboratory spectral library with the help of laboratory-developed software (Tomi, 1995 and Rezzi, 2002). In the investigated samples, individual components were identified by ^{13}C-NMR at contents as low as 0.4 - 0.5%.

***In vitro* radical scavening test:** The antioxidant potential with respect to $DPPH^{\bullet}$, was measured using a spectrophotometer (AL 800 / Spectro Direct) according to the method described by Blois (Blois, 1958). $DPPH^{\bullet}$ was solubilized in absolute ethanol (EtOH) to obtain a concentration solution of 0.03 mg / mL. Different ranges of concentrations (0.56, 0.28, 0.14, 0.07, 0.035 and 0.0175 mg / mL) of the EO were prepared in the same solvent. In dry sterile tubes, were added 2.5 mL of prepared EO analyte, and 1 mL of ethanolic solution of $DPPH^{\bullet}$. After 30 min of incubation, protected from light, the absorbance was read at 517 nm. The reference positive control is vitamin C, prepared under the same conditions as the study analytes. The percentage inhibition (I) of the $DPPH^{\bullet}$ is calculated according to the formula:

$$I = \frac{(Ab - As)}{Ab \times 100}$$

I: percentage inhibition (%);
Ab: absorbance of $DPPH^{\bullet}$ (nm);
As: absorbance of the sample (nm).

To overcome the influence of concentration, the effective half-concentration (EC50) of the antioxidant (%) was determined using the Graph pad prism software (Sharififar, 2007).

RESULTS AND DISCUSSION

Yield of extraction and Chemical composition

Yield of extraction: Hydrodistillation of plant material *O. syriacum* acclimated in Côte d'Ivoire gave an EO too fluid, pale yellow, with a highly pungent aroma. The following yields (*w/w* calculated on fresh weight basis) was obtained: 0.49 (fresh leaves, j-0), 0.55 (dried leaves, j-21) and 0.62% (dried leaves, j-42) (Figure 1). With regard to extraction yields, we deduced that obtaining EO from dried leafy stems was significantly advantageous than that provided with the same fresh organs.

Chemical composition: The combination of analytical techniques GC (in combination with retention indices on two columns of different polarity), GC-MS and ^{13}C-NMR applied to the three essential oils samples of *O. syriacum* led to the identification of 31 components accounting for 98.1-99.4% of the global composition (Table 1). EOs were characterized by the predominance of monoterpenes whose most important constituent was carvacrol (61.5-69.8%), an antibacterial compound. This analysis clearly demonstrated the carvacrol chemotype of EO of *O. syriacum* acclimated in Côte d'Ivoire. Beside this monoterpenoid phenol, γ-terpinene (11.3-14.5%) was present with appreciable amount and *p*-cymène (4.3-6.8%) with no negligible content. Other compounds coexisted at relatively lower levels, such as myrcene (2.9%). Among these, β-bisabolene (0.4-1.1%) and (E)-β-caryophyllene (0.3-0.5%) were the only identified sesquiterpenes. This carvacrol-dominated composition was similar to that of EO of individual plants from some Syrian populations of *O. syriacum* (Lucas, 2009) and plants cultivated in Lebanon (Zein, 2011).

Table 1. Constituents of EOs of *O. syriacum*

	Constituents	RIa	RIp	Relative proportion (%)		
				j-0	j-21	j-42
1	α-thujene	925	1015	1.8	1.04	1.9
2	α-pinene	933	1013	0.7	0.6	0.9
3	camphene	946	1062	0.1	0.1	0.1
4	oct-1-en-3-ol	963	1444	0.6	0.7	0.9
5	octan-3-one	966	1252	0.1	0.1	0.1
6	sabinene	967	1120	0.2	0.1	0.1
7	β-pinene	973	1109	0.2	0.2	0.2
8	octan-3-ol	981	1388	0.6	0.6	0.8
9	myrcene	983	1158	2.4	2.2	2.9
10	α-phellandrene	999	1162	0.3	0.3	0.4
11	Δ-3-carene	1007	1145	0.1	0.1	0.1
12	α-terpinene	1011	1177	2.2	2.1	2.8
13	*p*-cymene	1014	1268	4.3	4.8	6.8
14	β-phellandrene*	1023	1208	0.3	0.2	0.3
15	limonene*	1023	1198	0.3	0.2	0.3
16	(Z)-β-ocimene	1027	1229	0.1	0.1	0.1
17	(E)- β-ocimene	1038	1246	0.1	0.1	0.1
18	γ-terpinene	1051	1242	13.0	11.3	14.5
19	*trans* sabinenehydrate	1055	1460	0.3	0.8	1.1
20	terpinolene	1080	1279	0.1	0.1	0.1
21	*cis*sabinene hydrate	1084	1544	-	-	0.5
22	linalol	1085	1542	0.4	0.4	-
23	borneol	1151	1694	0.2	0.2	0.2
24	terpinen-4-ol	1163	1597	0.8	0.4	0.6
25	α-terpineol	1174	1691	0.3	0.2	0.2
26	carvone	1218	1730	0.1	0.1	0.1
27	carvacrolmethyloxyde	1226	1588	0.2	-	0.2
28	thymol	1270	2175	0.3	0.4	0.3
29	carvacrol	1283	2205	67.8	69.8	61.5
30	(E)-β-caryophyllene	1418	1588	0.4	0.5	0.3
31	β-bisabolene	1501	1717	1.1	0.4	0.5
	Hydrogenated monoterpenes			26.4	24.3	33.2
	Oxygenated monoterpenes			71.5	72.9	64.9
	Sesquiterpenes			1.5	0.9	0.8
	Total :			99.4	98.1	98.9

[a]) Order of elution and percentages of individual components are given on apolar column (BP-1), except those with an asterisk (percentage on polar column (BP-20) [b]) RIa retention indices determined on the apolar column *(BP-1)*. [c]) RIp retention indices determined on the polar column *(BP-20)*.[d]) (j-0): fresh leafy stems; [f]) (j-21): leafy stems dried for 21 days; [g]) (j-42): leafy stems dried for 42 days.

Table 2. EC_{50} values of studied EO and vitamin C

sample	Vitamin C	j-0	j-21	j-42
EC_{50} (mg/ml)	0.026	0.268	0.222	0.176

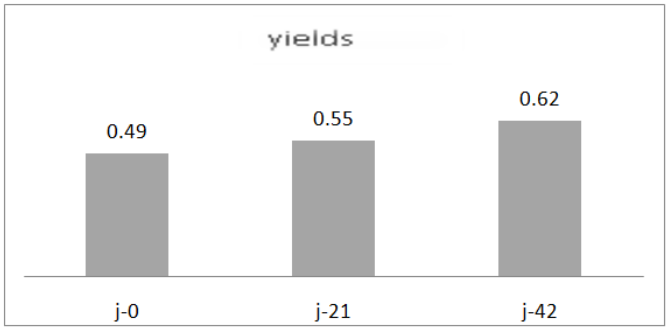

Figure 1. Extraction yields of EO from leafy stems fresh (j-0); dried for 21 (j-21) and 42 days (j-42)

This similarity in the composition of EO from various geographic areas suggests that soil and climate conditions do not have much influence on the chemical composition of EO of *O. syriacum*. In addition, drying had no influence on the chemical composition of EO of this plant which remained globally homogeneous.

Antioxidant potential of Eos

The antioxidant potential of an EO or any compound can be attributed, among other things, to its reducing power, to its ability to trap free radicals (Loizzo, 2009). Figure 2 shows the results of the in vitro antioxidant test of EO of *O. syriacum* against $DPPH^{\bullet}$, compared to vitamin C. From these results, it emerges that the inhibitory potential of the EO extracted against $DPPH^{\bullet}$ depended on the concentration. It increased gradually with increasing concentration of EO, same with vitamin C. However, the inhibitory capacity of EO were lower than that of vitamin C for all concentrations used. For a better intelligibility of the assessment of the inhibitory capacity of the tested EO, the EC50 were determined graphically (Table 2). EC50 of fresh leafy stems (0.268 mg / ml) was higher than that of dried organs (0.222 and 0.176 mg/ml). Indeed, more value of the EC50 is little more activity is significant. Thus, the antioxidant efficacy was relatively more consistent in the EO of dried organs. Globally, besides coexistence of monoterpenoids, antioxidant potential exhibited in EO of *O. syriacum* appears to be related to the predominant presence of carvacrol (Table 1).

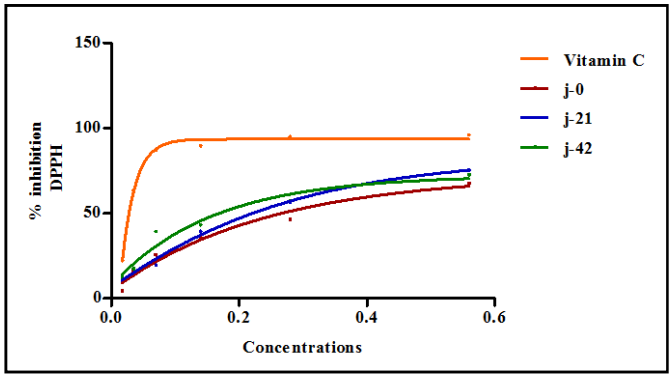

Figure 2. In vitro antioxidant test of EO from *O. syriacum* and vitamin C against DPPH •

This monoterpene phenol is a well-known antioxidant compound. In addition, EO was remarkably less effective than vitamin C, whose EC50 was 0.026 mg /ml. It is noted that the antioxidant activities of EOs of *O. syriacum* from Lebanon and Turkey have been reported (Loizzo, 2009 and Alma, 2003).

Conclusion

The GC(RI), GC-MS and ^{13}C-NMR analysis showed that the EO of *O. syriacum* acclimated in tropical zone, especially in Côte d'Ivoire, was qualitatively similar to the EO of species cultivated in the Mediterranean regions, and was dominated by carvacrol. Drying of leafy stems had no significant influence on organic composition of its oil, only the yield was improved. The antioxidant test against DPPH, established that this essential oil exhibited a significant antioxidant potential, but was still relatively low compared to that of vitamin C. The ability of the EO from *O. syriacum* , on the one hand, to clearly inhibit the oxidizing power of DPPH$^{\bullet}$, and on the other hand, its carvacrol chemotype, indicates that this species acclimated in Côte d'Ivoire, is a relevant nutritional candidate to integrate into Ivorian dietary habits as prevention against pathologies related to antioxidant stress.

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