Composition and antioxidant activity of the essential oil of *Origanum glandulosum* Desf. from Algeria

Semra I¹, Benmerache A¹, Chibani S¹, Kabouche A¹, Abuhamdah S² and Kabouche Z¹*

¹University of Constantine 1, Department of Chemistry, LOST, 25000 Constantine, Algeria
²Natural Products Laboratory Room 253, chemistry department, university of Jordan, Amman 11942, Jordan

**ABSTRACT**

The chemical composition of the hydrodistilled essential oil of *Origanum glandulosum* Desf., collected from Zighoud-Youcef (Eastern Algerian), was analyzed by GC and GC/MS. 18 Components representing 93.65% of the essential oil were detected with thymol (70.59%), tert-butylbenzene (5.76%), carvacrol (5.44%) and γ-terpinene (5.04%) as the major components. The antioxidant activity of the essential oil was investigated by the use of β-carotene bleaching method.

**Keywords**: *Origanum glandulosum*, Antioxidant activity, Linoleic acid, β-carotene.

**INTRODUCTION**

*Origanum glandulosum* Desf. Syn: *Origanum vulgare* subsp. *glandulosum* [1], belongs to the Lamiaceae family from which a great number of species are concentrated in the Mediterranean area. *Origanum glandulosum* “zaatar, zitra” is endemic to North Africa [2], it’s used in Algerian culinary preparations and in local folk medicine against cough flu, respiratory diseases and some gastrointestinal disorders [3-7]. In continuation of our works on Lamiaceae [8-32], we report here the composition and the antioxidant activity of the essential oil of *O. glandulosum* Desf., collected from Zighoud -Youcef (Eastern Algerian).

**MATERIALS AND METHODS**

**Plant Material**

Fresh aerial parts of *Origanum glandulosum* Desf., growing at Zighoud-Youcef (Eastern Algerian) were collected in May 2012. A voucher specimen was deposited at the herbarium of the University of Constantine 1, Algeria (LOST Og/05/12).

**Extraction of the essential oil**

Hydrodistillation of 100 g of fresh aerial parts of *O. glandulosum*, for 3 h using a Clevenger-type apparatus, yielded 2.2% (w/w) of a yellowish good smell oil.

**Gas chromatography**: GC analysis was performed on a Shimadzu GC17A gas chromatograph equipped with a split/splitless injector (250°C) and a flame ionization detector (250°C). Retention times for comparison with authentic compounds were measured using a cross-linked DB5-MS column (40 m × 0.18 mm, film thickness 0.18 μm). The oven temperature was programmed as isothermal at 60°C for 5 min, then raised to 275°C at 5°C/min and held at this temperature for 5 min. Helium was used as the carrier gas at a rate of 1 ml/min. Relative percentage amounts were calculated from peak area without the use of correction factors.
Gas Chromatography-Mass spectrometry: Gas chromatography-mass spectrometry: GC-MS was performed using a Shimadzu QP5050 mass selective detector using a cross-linked DB5-MS column (40 m × 0.18mm, film thickness 0.18 μm). The oven temperature was programmed as isothermal at 60°C for 5 min, then raised to 275°C at 5°C/min and held at this temperature for 5 min. Helium was used as the carrier gas at a rate of 1 ml/min. 0.1 μl oil was introduced directly into the source of the MS via a transfer line (280°C) with a split ratio of 1:50 and a linear velocity of 30.0 cm/sec. Ionization was obtained by electron impact (70 eV, source temperature 200°C, resolution 1000).

Components identification
Identification of the components was based on their retention indices (determined with reference to a homologous series of normal alkanes), and by comparison of their mass spectral fragmentation patterns with those reported in the literature [33, 34] and with authentic compounds for the major components.

Antioxidant activity
The antioxidant activity of the essential oils was evaluated by β-carotene–linoleic acid model system [35]. 0.5 mg of β-carotene in 1 mL of chloroform was added to 25 μL of linoleic acid and 200 mg of Tween 40 emulsifier mixture. After chloroform was evaporated under vacuum, 100 mL of distilled water saturated with oxygen were added by vigorous shaking. Four thousand microliters of this mixture were transferred into test tubes containing different concentrations of the sample. As soon as the emulsion was added to each tube, the zero time absorbance was measured at 470 nm using a spectrophotometer. The emulsion system was incubated for 2 h at 50°C. A blank, devoid of β-carotene, was prepared for background subtraction. Vitamin E was used as a standard. The bleaching rate (R) of β-carotene was calculated according to the following equation:

\[ R = \ln(a/b)/t \]

where ln is the natural log, a is the absorbance at time 0, b is the absorbance at time t (120 min). The antioxidant activity (AA) was calculated in terms of percent inhibition relative to the control, using the following equation:

\[ AA = \left(\frac{R_{\text{control}} - R_{\text{sample}}}{R_{\text{control}}}\right) \times 100 \]

RESULTS AND DISCUSSION

Chemical composition of the essential oil
The hydrodistilled essential oil of *O. glandulosum* Desf, was GC and GC/MS analyzed. 18 compounds, representing 93.65 % of the essential oil, were characterized with thymol (70.59%), tert-butylbenzene (5.76%), carvacrol (5.44%) and γ-terpinene (5.04%) as the major components. This oil is thymol chemotype, it’s nearly similar to the essential oils of the species collected from Setif Eastern Algerian), mainly characterized by thymol (72.4%), γ-terpinene (7.8%), p-cymene (5.4%) [24], and that collected from India which was found to contain thymol (61.8%), p-cymene (8.6%) and 3-octanone (7.6%) as major compounds [36]. Common main components of *Origanum* essential oils, thymol (45.4%, 51.1%, 34.2%, 38.8%, 23.9-31.6%), γ-terpinene (20%, 14.5%, 13.4%, 5.1%, 11.5-16.5%), carvacrol, (3.6%, 6.8%, 30.5%, 32.9%, 2.9-8%) and p-cymene (7.5%, 6.6%, 17.7%, 7.9%, 16.8-17.2%) were mainly found in essential oils of the Algerian species collected from Jijel (Eastern Algerian) [24], Constantine (Eastern Algerian) [24], Tlemcen (Western Algerian) [37], Setif (Eastern Algerian) [38] and Blida (Central Algerian) [39]. However, tert-butylbenzene is reported here for the first time from *Origanum glandulosum* essential oil.

Antioxidant activity
Total antioxidant activity by β-carotene bleaching method assays of *Origanum glandulosum*, collected from Zigoud-Youcef, was carried out. The activity was increased as dose dependent. The essential oil exhibited a high activity, the best inhibition (76.47 %) was measured at 4 mg/ml (Figure 1).
Table 1: Chemical composition, Retention indices and percentage composition of the essential oil of _Origanum glandulosum_ collected from Zighoud-Youcef

<table>
<thead>
<tr>
<th>Pic</th>
<th>Compounds</th>
<th>RI</th>
<th>(%)</th>
</tr>
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<tbody>
<tr>
<td>1</td>
<td>α-Thujene</td>
<td>930</td>
<td>0.77</td>
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<tr>
<td>2</td>
<td>α-Pinene</td>
<td>939</td>
<td>0.39</td>
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<tr>
<td>3</td>
<td>l-Octen-3-ol</td>
<td>979</td>
<td>0.34</td>
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<tr>
<td>4</td>
<td>Tert-Butylbenzene</td>
<td>990</td>
<td>5.76</td>
</tr>
<tr>
<td>5</td>
<td>3-Octanol</td>
<td>996</td>
<td>0.22</td>
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<tr>
<td>6</td>
<td>γ-Terpinene</td>
<td>1060</td>
<td>5.04</td>
</tr>
<tr>
<td>7</td>
<td>Sabine hydrate</td>
<td>1076</td>
<td>1.37</td>
</tr>
<tr>
<td>8</td>
<td>Borneol</td>
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</tr>
<tr>
<td>9</td>
<td>Terpinen-4-ol</td>
<td>1177</td>
<td>0.51</td>
</tr>
<tr>
<td>10</td>
<td>α-Terpineol</td>
<td>1189</td>
<td>0.36</td>
</tr>
<tr>
<td>11</td>
<td>Thymol</td>
<td>1290</td>
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</tr>
<tr>
<td>12</td>
<td>Carvacrol</td>
<td>1299</td>
<td>5.44</td>
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<tr>
<td>13</td>
<td>Caryophyllene</td>
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<td>1.10</td>
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<td>14</td>
<td>α-Humulene</td>
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<td>Veridiflorene</td>
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<td>16</td>
<td>Spathulenol</td>
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<td>Caryophyllene Oxide</td>
<td>1583</td>
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<td>18</td>
<td>Allosormadendrene</td>
<td>1641</td>
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</tr>
<tr>
<td></td>
<td>Identified compounds</td>
<td>Total</td>
<td>93.65</td>
</tr>
</tbody>
</table>

Ref: Compounds listed in order of their RI

RI (retention index) measured relative to n-alkanes (C₆-C₂₄) using DB-5MS column

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**CONCLUSION**

The hydrodistilled essential oil of _O. glandulosum_ Desf., collected from Zighoud-Youcef (Eastern Algerian) was characterized by thymol (70.59%), tert-butylbenzene (5.76%), carvacrol (5.44%) and γ-terpinene (5.04%) as the major components. This oil is thymol chemotype and is a little different from other reported Algerian _O. glandulosum_ essential oils. Tert-butylbenzene is reported here for the first time from _O. glandulosum_ essential oil.
This difference may be due to the soil, climate, altitude... The essential oil exhibited a high antioxidant activity by the β-carotene bleaching method.

Acknowledgments
We are grateful to the ANDRS (Algeria) and MESRS-DG/RSDT (Algeria) for financial support and to Mr. Abaza (Amman University, Jordan) for technical help.

REFERENCES


