

## Essential Oil Composition of *Centaurea sempervirens* L. (Asteraceae)

Belbache Hanene<sup>1</sup>, Mechehoud Youcef<sup>1</sup>, Chalchat Jean-Claude<sup>2</sup>, Figueredo Gilles<sup>3</sup>, Chalard Pierre<sup>2</sup>, Benayache Samir<sup>1</sup>, Benayache Fadila<sup>1\*</sup>

<sup>1</sup>Unité de Recherche Valorisation des Ressources Naturelles, Molécules Bioactives et Analyses Physicochimiques et Biologiques. Université des Frères Mentouri, Constantine 1, Route d'Aïn El Bey, 25000, Constantine, Algérie.

<sup>2</sup>Laboratoire de Chimie des Hétérocycles et Glucides, Ecole Nationale Supérieure de Chimie de Clermont-Ferrand, Ensemble Scientifique des Céseaux, BP 187- 63174, Clermont-Ferrand, France.

<sup>3</sup>Laboratoire d'Analyses des Extraits Végétaux et des Arômes (LEXVA Analytique), 460 Rue du Montant, Beaumont, France.

Received: 20<sup>th</sup> Nov, 16; Revised: 5<sup>th</sup> Dec, 16; Accepted: 24<sup>th</sup> Dec, 16; Available Online: 15<sup>th</sup> January, 2017

### ABSTRACT

The essential oil of the aerial parts of *Centaurea sempervirens* L. (Asteraceae), synonym : *Cheirolophus sempervirens* (L.) Pomel, was obtained by steam distillation and analyzed by GC-FID and GC-MS. 30 components were identified corresponding to 78.5% of the total oil. Among the identified constituents, oxygenated compounds represented 33.4%, from which 21.2% were hydrocarbons, 10.7% were sesquiterpenes. The non oxygenated compounds were hydrocarbons (9.8%). Phthalates represented 35.3% of the total oil. The major components were 6,10,14-trimethylpentadecan-2-one (12.4%) and *epi*-torilenol (5.1%). This is the first report on the chemical composition of the essential oil of this species.

**Keywords:** *Centaurea sempervirens* L., *Cheirolophus sempervirens* (L.) Pomel, Asteraceae, Essential oil composition.

### INTRODUCTION

The genus *Centaurea* from the Asteraceae family is one of the most widely distributed plant genera in the world. This genus includes more than 500 species, 45 of which grow spontaneously in Algeria, with 7 species localized in the Sahara<sup>1,2</sup>. Many *Centaurea* species are used in the popular medicine of many countries<sup>3-8</sup>. In addition, various studies have shown medicinal properties of *Centaurea* species such as antimicrobial<sup>9</sup>, antibacterial<sup>10,11</sup>, hypoglycemic<sup>12</sup>, antifungal<sup>13-16</sup>, cytotoxic and phytotoxic<sup>17-19</sup>, analgesic<sup>20</sup>, anti-inflammatory and immunological<sup>21</sup>. *Centaurea* species are well known for their high structural diversity in major bioactive compounds, including triterpenes, flavonoids, lignans, fatty acids and sesquiterpene lactones<sup>22-29</sup>.

Studies conducted on the chemical composition of their essential oils showed the presence of fatty acids, hydrocarbon derivatives, phytol,  $\beta$ -eudesmol and caryophyllene oxide in the majority of them<sup>30-32</sup>. Within the context of the study of *Centaurea* species growing in Algeria<sup>33-36</sup>, we have taken interest in the study of *Centaurea sempervirens* and more precisely the chemical composition of its essential oil. Previous studies on this plant led to the isolation of sesquiterpene acids and sesquiterpene lactones<sup>37</sup>, flavonoids<sup>38</sup> and polyacetylenes<sup>39,40</sup>. To the best of our knowledge there is no previous report on the essential oil composition of this species.

### MATERIALS AND METHODS

#### Plant material

The aerial parts of *Centaurea sempervirens*, synonym : *Cheirolophus sempervirens* (L.) Pomel<sup>41</sup> were collected on June 2011 from Djebel El Wahch in the area of Constantine Northeast Algeria, and authenticated by professor Mohamed Kaabeche, Setif 1 university Algeria, according to Quezel and Santa (1963)<sup>1</sup>. A voucher specimen has been deposited in the Herbarium of the VARENBIOMOL research unit, University Frères Mentouri Constantine 1.

#### Extraction of the essential oil

The aerial parts (400 g) of *Centaurea sempervirens* were subjected to steam distillation in a Kaiser Lang apparatus for three hours. The obtained essential oil was collected and dried over anhydrous sodium sulphate and kept at 4°C until analysis. The yield of the oil was calculated in relation of the dry weight of the plant.

#### GC-FID Analysis

The essential oil was analyzed on an Agilent gas chromatograph (GC-FID) Model 6890, equipped with a HP-5MS fused silica capillary column (5% -diphenyl-95% -dimethylpolysiloxane (25 m x 0.25 mm, film thickness 0.25  $\mu$ m), programmed from 50 °C (5 min) to 250 °C at 3 °/min and held for 10 min. Injector and flame ionization detector temperatures were 280 and 300 °C, respectively. The essential oil was diluted in acetone (3.5%, v/v) and injected in split mode (1/60), helium was used as a carrier gas (1.0 mL/min). Solutions of standard alkanes (C<sub>8</sub>-C<sub>20</sub>) were analyzed under the same conditions to calculate

Table 1: Composition of the essential oil of *Centaurea sempervirens* L. with retention times, retention indices and percentages.

Peak N°	RT	<sup>b</sup> RI	<sup>a</sup> Components	%
1.	6.169	983	Oct-1-en-3-ol	0.9
2.	6.829	1005	Hepta-2,4-dienal	0.4
3.	8.404	1100	Nonanal	0.8
4.	9.462	1165	Nonanol	1.0
5.	10.003	1201	Decanal	0.6
6.	10.895	1267	Nonanoic acid	1.9
7.	11.618	1315	Deca-2,4-dienal	0.4
8.	12.209	1367	Undecanol	1.7
9.	12.420	1383	$\beta$ -Damascenone	0.5
10.	12.502	1388	Dodecan-2-one	0.7
11.	13.832	1494	1-Pentadecene	1.0
12.	14.652	1563	<i>epi</i> -Torilenol	5.1
13.	14.781	1574	1,5-Epoxyalsvial-4(14)-ene	0.5
14.	14.879	1577	Spathulenol	1.4
15.	14.947	1582	Caryophyllene oxide	1.5
16.	15.015	1594	1-Hexadecene	0.3
17.	15.068	1597	Salvia-4(14)-en-1-one	0.4
18.	15.182	1609	$\beta$ -Oplophenone	0.4
19.	15.763	1661	$\beta$ -Eudesmol	1.4
20.	16.137	1694	1-Heptadecene	1.5
21.	16.387	1717	Pentadecanal	0.4
22.	17.696	1843	6,10,14-Trimethylpentadeca-2-one	12.4
23.	17.868	1860	Phthalate	2.3
24.	18.892	1963	Diisopentylphthalate	33.0
25.	20.202	2093	Phytol	1.0
26.	21.918	2299	Tricosane	1.6
27.	22.740	2399	Tetracosane	0.5
28.	23.529	2499	Pentacosane	1.8
29.	25.027	2700	Heptacosane	2.0
30.	26.426	2900	Nonacosane	1.1
Total identified				78.5
Grouped compounds				
hydrocarbons				9.8
Oxygenated hydrocarbons				21.2
Rose ketone				0.5
Oxygenated sesquiterpenes				10.7
Oxygenated diterpene				1.0
Phthalates				35.3

<sup>a</sup>Compounds are listed in order of their RI

<sup>b</sup>RI (retention index) measured relative to *n*-alkanes (C<sub>8</sub>-C<sub>20</sub>) using HP-5MS

retention indices (RI) with Van del Dool and Kratz equation.

#### GC-MS Analysis

Mass spectrometry was performed on an Agilent gas chromatograph-mass spectrometer (GC-MS) Model 7890/5975, equipped with HP-5MS capillary column (25 m x 0.25 mm, film thickness 0.25  $\mu$ m) programmed with the same conditions as for GC-FID. The mass spectrometer (MS) ionization was set in positive electron impact mode at 70 eV and electron multiplier was set at 2200 V. Ion source and MS quadrupole temperatures were 230 °C and 180 °C, respectively. Mass spectral data were acquired in the scan mode in the *m/z* range 33-450. The essential oil constituents were identified by matching their mass spectra

and retention indices (RI) with those of reference compounds from libraries such as Adams<sup>42</sup> and Mc Lafferty & Stauffer<sup>43</sup>. The proportions of the identified compounds were calculated by internal normalization.

## RESULTS AND DISCUSSION

The steam distillation of the essential oil of *Centaurea sempervirens* gave a viscous liquid with a green color and a strong odor. The yield of essential oil was 0.2% (w/w) in relation to the dry weight of the plant. The analysis and identification of the compounds of the essential oil was performed using the GC-MS. The general chemical profile of the essential oil, the percentage content and retention indices of the constituents are summarized in Table 1. This investigation allowed the identification of 30 constituents

corresponding to 78.5% of the total oil. Among the identified constituents, oxygenated compounds represented 33.4%, from which 21.2% were hydrocarbons, 10.7% were sesquiterpenes. The non oxygenated compounds were hydrocarbon derivatives (9.8%). Phthalates represented 35.3% of the total oil. The major components were diisopentylphthalate (33.0%); 6,10,14-trimethylpentadecan-2-one (12.4%) and *epi*-torilenol (5.1%). The presence of 6,10,14-trimethylpentadecan-2-one as main component of *C. sempervirens* agreed with the reported results on *C. grisebachii* subsp. *grisebachii*<sup>44</sup> while the presence of the phthalate esters could be explained in most cases by a probable pollution of the harvest area of studied plants<sup>45</sup>.

To the best of our knowledge this is the first report on essential oil composition of *Centaurea sempervirens* L.

## CONCLUSION

We report for the first time the essential oil composition of *Centaurea sempervirens* collected from the area of Constantine in the Northeast of Algeria. Analysis by GC-FID and GC-MS allowed the identification of 6,10,14-trimethyl-pentadeca-2-one (12.4%) and *epi*-torilenol (5.1%) as major components. In addition, the presence in significant amount of hydrocarbons (9.8%), oxygenated hydrocarbons (21.2%) and oxygenated sesquiterpenes (10.7%) was in accordance with the chemical composition of other *Centaurea* species.

## ACKNOWLEDGEMENTS

We are grateful to professor Mohamed Kaabeche (Setif 1 university, Algeria) for the identification of the plant material and MESRS (DGRSDT) for financial support.

## REFERENCES

1. Quezel P, Santa S. Nouvelle Flore de l'Algérie et des Régions Désertiques Méridionales. CNRS Paris, edn. Vol. II, 1963. p. 1016-1032.
2. Ozenda P. Flore du Sahara Septentrional et Central. CNRS. 1958. p. 450.
3. Vázquez FM, Suarez MA, Pérez, A. Medicinal plants used in the Barros Area, Badajoz Province (Spain). J. Ethnopharmacol. 1997; 55(2), 81–85.
4. Arif R, Küpeli E, Ergun F. The biological activity of *Centaurea* L. species. GU J. Sci. 2004; 17(4),149-164.
5. Baytop T. Türkiye'de bitkiler ile tedavi (Geçmişte ve bugün). Istanbul: Nobel Tıp Kitabevleri; 1999; p.316.
6. Alapetite GP. Flore de la Tunisie. Imprimerie Officielle de la RépubliqueTunisienne: Tunis 1981, p. 1060.
7. Fakhfakh JA, Damak M. Sesquioneolignans from the flowers of *Centaurea furfuracea* Coss. et Dur. (Asteraceae). Nat. Prod.Res. 2007; 21(12),1037–1041.
8. Aclinou P, Boukerb A, Bouquant J, Massiot G, Le Men-Olivier, L. Plantes des Aures: Constituants des racines de *Centaurea incana*. Plant. Med. Phytother. 1982; 16, 303–309.
9. Karioti A, Skaltsa H, Lazari D, Sokovic M, Garcia B, Harvala C. Secondary Metabolites from *Centaurea deusta* with Antimicrobial Activity. Z. Naturforsch., C. 2002; 57c, 75-80.
10. Yesilada E, Gürbüz I, Shibata H. Screening of Turkish anti-ulcerogenic folk remedies for anti-*Helicobacter pylori* activity. J. Ethnopharmacol. 1999; 66(3), 289-293.
11. Ciric A, Karioti A, Glamoclija J, Sokovic M, Skaltsa H. Antimicrobial activity of secondary metabolites isolated from *Centaurea spruneri* Boiss. & Heldr. J. Serb. Chem. Soc. 2011; 76, 27-34.
12. Chucla MT, Lamela M, Gato A, Cadavid I. *Centaurea corcubionensis*: a study of its hypoglycemic activity in rats. Planta Med. 1988; 54, 107–109.
13. Skaltsa H, Lazari D, Garcia B, Pedro JR, Sokovic M, Constantinidis T. Sesquiterpene lactones from *Centaurea achaia*, a greek endemic species. Antifungal activity. Z. Naturforsch. 2000; 55c, 534-539.
14. Vajs V, Todorović N, Ristić M, Tešević V, Todorović B, Janačković P, Marin P, Milosavljević S. Guaianolides from *Centaurea nicolai*: antifungal activity. Phytochemistry. 1999; 52, 383-386.
15. Barrero AF, Oltra JE, Alvarez M, Raslan DS, Saúde DA, Akssira M. New sources and antifungal activity of sesquiterpene lactones. Fitoterapia. 2000; 71, 60.
16. Koukoulitsa C, Geromichalos GD, Skaltsa H. VolSurf analysis of pharmacokinetic properties for several antifungal sesquiterpene lactones isolated from Greek *Centaurea* sp. J. Comput. Aided Mol. Des. 2005; 19(8), 617–623.
17. Koukoulitsa E, Skaltsa H, Karioti A, Demetzos C, Dimas K. Bioactive sesquiterpene lactones from *Centaurea* species and their cytotoxic/cytostatic activity against human cell lines *in vitro*. Planta Med. 2002; 68(7), 649-652.
18. Tukov FF, Anand S, Gadepalli RSVS, Gunatilaka AAL, Mathews JC, Rimoldi JM. Inactivation of the Cytotoxic Activity of Repin, a Sesquiterpene Lactone from *Centaurea repens*. Chem. Res. Toxicol. 2004; 17(9), 1170-1176.
19. Medjroubi K, Benayache F, Bermejo J. Sesquiterpene lactones from *Centaurea musimomum*. Antiplasmodial and cytotoxic activities. Fitoterapia 2005; 76(7-8),744-746.
20. Djeddi S, Argyropoulou C, Chatter R. Analgesic properties of secondary metabolites from Algerian *Centaurea pullata* and Greek *C. grisebachii* ssp. *grisebachii*. J. Appl. Sci. Res. 2012; 8(6),2876-2880.
21. Garbacki N, Gloaguen V, Damas J, Bodart P, Tits M, Angenot, L. Antiinflammatory and immunological effects of *Centaurea cyanus* flower-heads. J. Ethnopharmacol. 1999; 68(1-3), 235–241.
22. Seghiri R, Boumaza O, Mekkiou R, Benayache S, Mosset P, Quintana J, Estévez F, León F, Bermejo J, Benayache F. A flavonoid with cytotoxic activity and other constituents from *Centaurea africana*. Phytochem. Lett. 2009; 2(3), 114-118.
23. Kolli EH, León F, Benayache F, Estévez S, Quintana J, Estévez F, Brouard I, Bermejo J, Benayache S. Cytotoxic sesquiterpene lactones and other

- constituents from *Centaurea omphalotricha*. J. Braz. Chem. Soc. 2012; 23(5), 977-983.
24. López-Rodríguez M, García V P, Zater H, Benayache S, Benayache F. Cynaratriol, a sesquiterpene lactone from *Centaurea musimomum*. Acta Cryst. 2009; E65, o1867-o1868.
  25. Shoeb M, MacManus SM, Jaspars M, Kong-Thoo-Lin P, Nahar L, Celik S, Sarker SD. Bioactivity of two Turkish endemic *Centaurea* species, and their major constituents. Braz. J. Pharmacog. 2007; 17(2), 155-159.
  26. Shoeb M, MacManus SM, Kumarasamy Y, Jaspars M, Nahar L, Thoo-Lin PK, Nazemiyeh H, Sarker SD. Americanin, a bioactive dibenzylbutyrolactone lignan, from the seeds of *Centaurea americana*. Phytochemistry 2006; 67(21), 2370-2375.
  27. Demir S, Karaalp C, Bedir E. Unusual sesquiterpenes from *Centaurea athoa* DC. Phytochem. Lett. 2016; 15, 245-250.
  28. Aktumsek A, Zengin G, Guler GO, Cakmak YS, Duran A. Assessment of the antioxidant potential and fatty acid composition of four *Centaurea* L. taxa from Turkey. Food Chem. 2013; 141(1), 91-97.
  29. Milošević Ifantis T, Solujić S, Pavlović-Muratspahić D, Skaltsa H. Secondary metabolites from the aerial parts of *Centaurea pannonica* (Heuff.) Simonk. from Serbia and their chemotaxonomic importance. Phytochemistry 2013; 94, 159-170.
  30. Senatore F, Formisano C, Raio A, Bellone G, Bruno M. Volatile components from flower-heads of *Centaurea nicaeensis* All., *C. parlatoris* Helder and *C. solstitialis* L. ssp. *schowii* (DC.) Dostál growing wild in southern Italy and their biological activity. Nat. Prod. Res. 2008; 22(10), 825-832.
  31. Polatoglu K, Sen A, Bulut G, Bitis L, Gören N. Essential Oil Composition of *Centaurea stenolepis* Kerner. from Turkey. J. Essent. oil bear. Pl. 2014; 17(6), 1268-1278.
  32. Ben Jemia M, Senatore F, Bruno M, Bancheva S. Components from the Essential oil of *Centaurea aeolica* Guss. and *C. diluta* Aiton from Sicily, Italy. Rec. Nat. Prod. 2015; 9(4), 580-585.
  33. Azzouzi D, Mekkiou R, Chalard P, Chalchat JC, Boumaza O, Seghiri R, Benayache F, Benayache S. Essential Oil Composition of *Centaurea choulettiana* Pomel (Asteraceae) from Algeria. International Journal of Pharmacognosy and Phytochemical Research 2016; 8(9), 1545-1548.
  34. Zater H, Huet J, Fontaine V, Benayache S, Stévigny C, Duez P, Benayache F. Chemical constituents, Cytotoxic, antifungal and Antimicrobial properties of *Centaurea diluta* Ait. subsp. *algeriensis* (Coss. & Dur.). Maire. Asian Pac. J. Trop. Med. 2016, 9(6), 554-561.
  35. Bicha S, Chalard P, Hammoud L, León F, Brouard I, Garcia VP, Lobstein A, Bentamene A, Benayache S, Bermejo J, Benayache F. Maroccanin: a new  $\gamma$ -lactone and Other constituents from *Centaurea maroccana* Ball. (Asteraceae). Rec. Nat. Prod. 2013; 7(2), 114-118.
  36. Boudjerda A, Zater H, Benayache S, Chalchat JC, González-Platas J, León F, Brouard I, Bermejo J, Benayache F. A new guaianolide and other constituents from *Achillea ligustica* Biochem. Syst. Ecol. 2008; 36 (5-6), 461- 466.
  37. Marco. J A, Sanz-Cervera J F, Garcia-Lliso V, Susanna A, Garcia-Jacas N. Sesquiterpene lactones, lignans and aromatic esters from *Cheirolophus* species. Phytochemistry 1994; 37(4), 1101-1107.
  38. Plouvier V. Structure of flavone glycosides by nuclear magnetic resonance. Compounds of the genera, *Centaurea*, *Kerria*, *Rhus*, and *Scabiosa*. Comptes Rendus des Séances de l'académie des sciences, Serie D: Sciences naturelles 1970; 270(22), 2710-2713.
  39. Bohlmann F, Postulka S, Ruhnke J. Polyacetylene compounds. XXIV. Polyynes of the species *Centaurea*. Chem. Ber. 1958; 91, 642-656.
  40. Bohlmann F, Burkhardt T, Zdero C. Naturally occurring acetylenes. Academic Press, London. 1973.
  41. The Plant List (2010). Version 1. Published on the Internet; <http://www.theplantlist.org/> (accessed 1st January).
  42. Adams RP. Identification of essential oil components by gas chromatography/mass spectroscopy. Allured Publishing Co. Carol Stream, Illinois. 1995.
  43. Mc Lafferty FW, Stauffer DB. The Wiley/NBS registry of mass spectral data. 5<sup>th</sup> Edition, J. Wiley and Sons, New York. 1991.
  44. Djeddi S, Sokovic M, Skaltsa H. Analysis of the essential oils of some *Centaurea* species (Asteraceae) growing wild in Algeria and Greece and investigation of their antimicrobial activities. J. Essent. Oil Bear. Pl. 2011; 14(6), 658-666.
  45. Manayi A, Kurepaz-mahmoodabadi M, Gohari AR, Ajani Y, Saeidnia S. Presence of phthalate derivatives in the essential oils of a medicinal plant *Achillea tenuifolia*. DARU 2014; 22(1), 78-83.