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Asian Pacific Journal of Tropical Biomedicine

journal homepage: www.elsevier.com/locate/apjtb

Original article http://dx.doi.org/10.1016/j.apjtb.2015.06.007

Chemical constituents in the essential oil of the endemic plant *Cotula cinerea* (Del.) from the southwest of Algeria

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ARTICLE INFO

Article history:
Received 8 May 2015
Received in revised form 18 May, 2nd revised form 5 Jun 2015
Accepted 19 Jun 2015
Available online 12 Aug 2015

Keywords:
Essential oil
Gas chromatography-mass spectrometry analysis
Cotula cinerea
Asteraceae
Sahara plants

ABSTRACT

Objective: To extract and identify the main constituents of the essential oil of *Cotula cinerea* (Del.) (Asteraceae family) from southwest of Algeria.
Methods: The essential oils obtained by hydrodistillation, from the aerial parts of the endemic plant *Cotula cinerea* which was collected in the region of Sahara from southwest of Algeria, were analyzed by gas chromatography-mass spectrometry.
Results: A total of 33 compounds were identified representing 98.66% of the oil. The main compounds were (E)-citral (24.01%), limonene epoxide cis- (18.26%), thymol methyl ether (15.04%), carvacrol (15.03%), trans-carveol (13.79%), carvone (3.06%) and trans-piperitol (2.54%).
Conclusions: The main constituents in essential oil of the aerial part of the plant from southwest of Algeria were different from that collected from southeast of Algeria or in Morocco.

1. Introduction

Because of their therapeutic properties, medicinal plants used in traditional medicine are one of the most interesting fields for the discovery of new drugs for the treatment of discharged diseases [1,2].

No one can deny or retract that the beneficial effects of Saharan plants guide us to explore the essential oils secreted from endemic plants that were found in the Béchar region (southwest of Algeria). *Cotula cinerea* (Del.) (*C. cinerea*) from Asteraceaes (Compositae) family [3], is one of these endemic species and also the object of our study (Figure 1).

The genus *Cotula* comprising 80 species is widespread in the southern hemisphere and is represented by three species in Algeria among which is our plant *C. cinerea* [4].

The species *C. cinerea*, with synonym of *Brocchia cinerea* [5], is a small woolly whitish plant, with thick leaf divided in their upper part to a 3–5 obtuse teeth, stems are 10–40 cm, slept then raised; capitulum from 6 to 10 mm in diameter, woolly involucres with a tubular flower, and brown buds which would become golden yellow [2–6].

Figure 1. Photograph of *C. cinerea* taken at the time of collection.

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Peer review under responsibility of Hainan Medical University.

The phytochemical screening of *C. cinerea* revealed the presence of flavonoids, tannins, alkaloids, saponins, terpenoids, steroids, and cardenolides [2]. These compounds are known to exhibit a wide range of biological effects including antibacterial and antioxidant activities. These data could justify the traditional use of this plant [7].

This plant is usually known as "Guertofa" among the local people in Northern Sahara and is commonly used in traditional medicine in the southwest of Algeria, for the treatment of several diseases like colic, cough, diarrhea, headache, and digestive disorders. All parts of the plant are used in different forms (maceration, decoction, infusion or inhalation), according to the treated diseases [2].

The aim of this work is to extract and identify the main constituents of the essential oil obtained from the aerial part (leaves and stem) of *C. cinerea*, collected in the Southwestern Algeria.

2. Materials and methods

2.1. Plant materials

The aerial parts (leaves and stems) of *C. cinerea*, were collected in February and March, 2011, in the mountains and desert of Lahmar City (30 km far away from Béchar State), in southwest of Algeria and southeast of Morocco. The plants were identified and a voucher specimen was deposited at the herbarium of the Valorization of Resource and Food Security in Semiarid Areas Laboratory, South West of Algeria, University of Béchar.

2.2. Extraction of essential oil

The aerial parts (1 kg) were washed, sorted and dried for one month at room temperature in the shade and then were finely pulverized by using a mill blade. Clevenger-type apparatus was used for extraction, and hydrodistillation was performed for 6 h. The essential oil was dried with magnesium sulfate, then weighed and stored under refrigeration at 4–6 °C in dark glass tubes for further use.

2.3. Analysis by gas chromatography–mass spectrometry (GC–MS)

Essential oils were analyzed by GC–MS (a Perkin Elmer Clarus 600 GC–MS system coupled with a Perkin Elmer Clarus 600C MS). Helium (1 mL/min) was used as a carrier gas. The initial temperature was 60 °C (2 min), then was increased to 200 °C at rate of 5 °C/min, and remained at 200 °C for 5 min, and then was continued to increase to 250 °C at rate of 10 °C/min, with a final stage of 10 min at 250 °C. The essential oils were placed in a glass capillary column (60 m × 0.25 mm × 0.25 μm) filled with Rtx-5MS, and the oven temperature was programmed from 50 °C to 250 °C at a 5 °C/min dynamic rate, and remained for 15 min at 250 °C.

2.4. Identification of essential oil composition

Identification of components was performed by comparing their relative retention index (RI) determined with the reference of a homologous series of *n*-alkanes (C_8 to C_{24}) [8–13]. The fragmentation patterns of the mass spectra were compared with the WILEY and NIST 05 libraries. The linear temperature-programmed RIs of all the constituents were calculated based on the GC through the interpolation between bracketing *n*-alkanes as follow:

$$RI = 100 \times \left[\frac{t_{R(i)} + t_{R(Z)}}{t_{R(Z+1)} + t_{R(Z)}} Z \right]$$

where Z was the number of carbon atoms in the smaller *n*-alkane, and $t_{R(i)}$, $t_{R(Z)}$ and $t_{R(Z+1)}$ were the retention time of the desired compound, the smaller *n*-alkane and the larger *n*-alkane, respectively [14].

3. Results

The yield of the greenest essential oil extracted by hydrodistillation from aerial parts of *C. cinerea* was 0.282% (v/w). The results of GC–MS analysis showed the identification of 33

Table 1
Chemical composition of *C. cinerea* essential oils.

No.	RT	Name of compounds	Formula	Area (%)	KI
1	10.93	Unknown	–	0.02	912.07
2	14.28	α-fenchene	$C_{10}H_{16}$	0.26	958.09
3	17.54	α-phellandrene	$C_{10}H_{16}$	0.08	1 002.44
4	19.43	Limonene	$C_{10}H_{16}$	0.98	1 024.47
5	20.52	(E)-β-ocimene	$C_{10}H_{16}$	0.41	1 037.17
6	21.14	(Z)-β-ocimene	$C_{10}H_{16}$	0.14	1 044.40
7	21.86	Cis-dihydromultifidene	$C_{11}H_{18}$	0.25	1 052.79
8	22.45	(Z)-sabinyl acetate	$C_{12}H_{18}O_2$	1.18	1 059.66
9	23.80	Terpinolene	$C_{10}H_{16}$	0.05	1 075.39
10	24.10	Linalool oxide II (pyran)	$C_{10}H_{18}O_2$	0.13	1 078.89
11	26.39	Linalool	$C_{10}H_{18}O$	0.61	1 105.53
12	27.62	Limonene epoxide cis-	$C_{10}H_{16}O$	18.26	1 119.73
13	35.48	Trans-piperitol	$C_{10}H_{18}O$	2.54	1 210.91
14	35.83	Thymol methyl ether	$C_{11}H_{16}O$	15.04	1 215.10
15	36.64	Trans-carveol	$C_{10}H_{16}O$	13.79	1 224.82
16	36.80	Carvone	$C_{10}H_{14}O$	3.06	1 226.73
17	37.94	(E)-citral	$C_{10}H_{16}O$	24.01	1 240.40
18	40.88	Carvacrol	$C_{10}H_{14}O$	15.03	1 275.65
19	42.75	Bornyl acetate	$C_{12}H_{20}O_2$	1.15	1 298.08
20	47.59	Geranyl acetate	$C_{12}H_{20}O_2$	0.60	1 359.61
21	50.01	β-Elemene	$C_{15}H_{24}$	0.15	1 390.44
22	50.78	Bornyl isobutyrate	$C_{14}H_{24}O_2$	tr	1 400.26
23	52.10	Trans-caryophyllene	$C_{15}H_{24}$	0.16	1 417.86
24	53.28	Caryophyllene B	$C_{15}H_{24}$	0.04	1 433.60
25	56.07	Neryl isobutyrate	$C_{14}H_{24}O_2$	0.05	1 470.80
26	57.58	Bicyclogermacrene	$C_{15}H_{24}$	0.44	1 490.94
27	59.91	(Z)-nerolidol	$C_{15}H_{26}O$	0.10	1 522.57
28	61.86	Nerolidol trans-	$C_{15}H_{26}O$	0.21	1 549.22
29	62.28	Germacrene B	$C_{15}H_{24}$	0.06	1 554.96
30	64.55	Scapanol	$C_{15}H_{26}O$	0.12	1 586.00
31	65.41	Geranyl isopentanoate	$C_{15}H_{26}O_2$	0.11	1 597.75
32	67.90	Unknown	–	0.27	1 636.54
33	70.01	α-Bisabolol	$C_{15}H_{26}O$	0.06	1 669.70
34	71.85	Unknown	–	0.28	1 698.61
35	73.07	Farnesol (isomer 2)	$C_{15}H_{26}O$	0.03	1 717.87
36	74.53	Bisabolol oxide A	$C_{15}H_{26}O_2$	0.05	1 740.93
Grouped compounds					
Monoterpene hydrocarbons				2.17	
Oxygenated monoterpenes				95.40	
Sesquiterpene hydrocarbons				0.41	
Oxygenated sesquiterpenes				0.68	

RI: Retention indices relative to C_8–C_{24} *n*-alkanes on the Rtx-5MS column; RT: Retention time; KI: Kovats index; tr: Trace ≤ 0.01%.

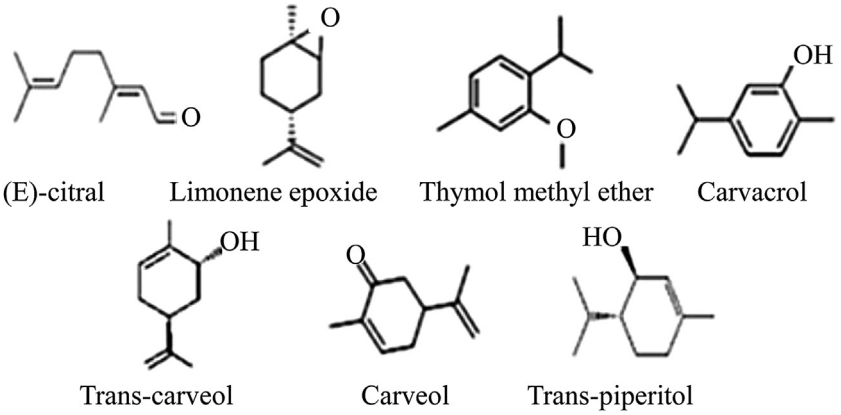

Figure 2. The major compounds of essential oil composition.

constituents representing 98.66% of the total oil composition. As shown in Table 1, the major compounds were (E)-citral (24.01%), limonene epoxide cis- (18.26%), thymol methyl ether (15.04%), carvacrol (15.03%), trans-carveol (13.79%), carvone (3.06%) and trans-piperitol (2.54%) (Figure 2). The oil composition was dominated by the presence of oxygenated monoterpenes (95.40%) followed by monoterpene hydrocarbons (2.17%), oxygenated sesquiterpenes (0.68%) and sesquiterpene hydrocarbons (0.41%).

4. Discussion

The essential oils from the aerial part of the species *C. cinerea* have been studied and described in the literature by several groups because of their interesting biological properties and varying bioactive compounds. Studies from El Bouzidi *et al.* and Kasrati *et al.* described trans-thujone (41.4%), cis-verbenyl acetate (24.7%), 1,8-cineole (8.2%) and camphor (5.5%) as the major components of the plants collected in Morocco [15,16], while Atef *et al.* described 3-carène (30.99%), thujone (21.73%), santolina triene (18.58%) and camphor (6.21%) from the oil obtained from a specimen collected in the Oued Souf Sahara in southeast of Algeria [17]. These essential oils have very different compositions from the essential oil extracted from the aerial parts of *C. cinerea* presented in this study, with the major compounds of (E)-citral (24.01%), limonene epoxide cis- (18.26%), thymol methyl ether (15.04%), carvacrol (15.03%), trans-carveol (13.79%), carvone (3.06%) and trans-piperitol (2.54%). These qualitative and quantitative differences in the chemical composition of essential oils could be attributed to several factors such as geographical location, the climatic effects, harvest season, nature of the soil, age of the plant parts (young or adult), the state of used plant materials (dried or fresh), the part of the plant used, time of collection and chemotype [18–20]. The oxygenated monoterpenes were found to be the major group in essential oil of *C. cinerea*, constituting 95.40%, and reported to be responsible for the antimicrobial activity of several essential oils [21,22]. This activity can justify the use of this plant in traditional medicine by the local people of Northern Sahara.

In conclusion, the essential oil of the aerial parts of *C. cinerea* was analyzed by GC–MS. (E)-citral, limonene epoxide cis-, thymol methyl ether, carvacrol, trans-carveol, carvone and trans-piperitol were the major constituents of the oil comprising 24.01%, 18.26%, 15.04%, 15.03%, 13.79%, 3.06% and 2.54%, respectively, of the total compounds present.

The main constituents of the aerial part essential oil of the plant from southwest of Algeria were different from that collected from southeast of Algeria or in Morocco. These data present the existence of different chemotypes for this species [23].

Conflict of interest statement

We declare that we have no conflict of interest.

References

[1] Hamada D, Ladjel S. Chemical composition, *in-vitro* anti-microbial and antioxidant activities of the methanolic extract of *Anvillea radiata* Asteraceae. *Res J Pharm Biol Chem Sci* 2015; **6**(2): 1367-73.
[2] Djellouli M, Moussaoui A, Benmehdi H, Ziane L, Belabbes A, Badraoui M, et al. Ethnopharmacological study and phytochemical screening of three plants (Asteraceae family) from the region of south west Algeria. *Asian J Nat Appl Sci* 2013; **2**(2): 59-65.
[3] Talibi I, Amkraz N, Askarne L, Msanda F, Saadi B, Boudyach EH, et al. Antibacterial activity of Moroccan plants extracts against *Clavibacter michiganensis* subsp. michiganensis, the causal agent of tomatoes' bacterial canker. *J Med Plants Res* 2011; **5**(17): 4332-8.
[4] Dendougui H, Seghir S, Jay M, Benayache F, Benayache SB. Flavonoids from *Cotula cinerea* Del. *Int J Med Arom Plants* 2012; **2**(4): 589-95.
[5] Markouk M, Redwane A, Lazrek HB, Jana M, Benjama A. Antibacterial activity of *Cotula cinerea* extracts. *Fitoterapia* 1999; **70**(3): 314-6.
[6] Ozenda P. [*Flora of the Sahara*]. Paris: French National Center for Scientific Research; 1983, p. 270. French.
[7] Metrouh-Amir H, Duarte CMM, Maiza F. Solvent effect on total phenolic contents, antioxidant, and antibacterial activities of *Matricaria pubescens*. *Ind Crops Prod* 2015; **67**: 249-56.
[8] Comai S, Dall'Acqua S, Grillo A, Castagliuolo I, Gurung K, Innocenti G. Essential oil of *Lindera neesiana* fruit: chemical analysis and its potential use in topical applications. *Fitoterapia* 2010; **81**(1): 11-6.
[9] Davies NW. Gas chromatographic retention indices of monoterpenes and sesquiterpenes on methyl silicon and Carbowax 20M phases. *J Chromatogr A* 1990; **503**: 1-24.
[10] Paolini J, Costa J, Bernardini AF. Analysis of the essential oil from aerial parts of *Eupatorium cannabinum* subsp. corsicum (L.) by gas chromatography with electron impact and chemical ionization mass spectrometry. *J Chromatogr A* 2005; **1076**(1–2): 170-8.
[11] De Falco E, Mancini E, Roscigno G, Mignola E, Taglialatela-Scafati O, Senatore F. Chemical composition and biological activity of essential oils of *Origanum vulgare* L. subsp. vulgare L. under different growth conditions. *Molecules* 2013; **18**(12): 14948-60.
[12] Lone SH, Bhat KA, Bhat HM, Majeed R, Anand R, Hamid A, et al. Essential oil composition of *Senecio graciliflorus* DC: comparative

analysis of different parts and evaluation of antioxidant and cytotoxic activities. *Phytomedicine* 2014; **21**(6): 919-25.

[13] Maggi F, Bramucci M, Cecchini C, Coman MM, Cresci A, Cristalli G, et al. Composition and biological activity of essential oil of *Achillea ligustica* All. (Asteraceae) naturalized in central Italy: ideal candidate for anti-cariogenic formulations. *Fitoterapia* 2009; **80**(6): 313-9.

[14] Jalali-Heravi M, Moazeni-Pourasil RS, Sereshti H. Elimination of chromatographic and mass spectrometric problems in GC–MS analysis of lavender essential oil by multivariate curve resolution techniques: improving the peak purity assessment by variable size moving window-evolving factor analysis. *J Chromatogr B Analyt Technol Biomed Life Sci* 2015; **983–984**: 83-9.

[15] El Bouzidi L, Abbad A, Fattarsi K, Hassani L, Leach D, Markouk M, et al. Chemical composition and anticandidal properties of the essential oil isolated from aerial parts of *Cotula cinerea*: a rare and threatened medicinal plant in Morocco. *Nat Prod Commun* 2011; **6**(10): 1491-4.

[16] Kasrati A, Alaoui Jamali C, Bekkouche K, Wohlmuth H, Leach D, Abbad A. Comparative evaluation of antioxidant and insecticidal properties of essential oils from five Moroccan aromatic herbs. *J Food Sci Technol* 2015; **52**(4): 2312-9.

[17] Atef C, Boualem M, Cherif MM, Youcef H, Azzedine C. Chemical composition and antimicrobial activity of essential oils in xerophytic plant *Cotula cinerea* Del (Asteraceae) during two stages of development: flowering and fruiting. *J Appl Pharm Sci* 2015; **5**(3): 29-34.

[18] Ben Marzoug HN, Romdhane M, Lebrihi A, Mathieu F, Couderc F, Abderraba M, et al. *Eucalyptus oleosa* essential oils: chemical composition and antimicrobial and antioxidant activities of the oils from different plant parts (stems, leaves, flowers and fruits). *Molecules* 2011; **16**(2): 1695-709.

[19] Pirbalouti AG, Ghahfarokhi BB, Ghahfarokhi SAM, Malekpoor F. Chemical composition of essential oil from the aerial parts and underground parts of Iranian valerian collected from different natural habitats. *Ind Crops Prod* 2015; **63**: 147-51.

[20] El-Akhal F, El Ouali Lalami A, Ez Zoubi Y, Greche H, Guemmouh R. Chemical composition and larvicidal activity of essential oil of *Origanum majorana* (Lamiaceae) cultivated in Morocco against *Culex pipiens* (Diptera: Culicidae). *Asian Pac J Trop Biomed* 2014; **4**(9): 746-50.

[21] Mahboubi M, Kazempour N. Chemical composition and antimicrobial activity of *Satureja hortensis* and *Trachyspermum copticum* essential oil. *Iran J Microbiol* 2011; **3**(4): 194-200.

[22] Carson CF, Riley TV. Antimicrobial activity of the major components of the essential oil of *Melaleuca alternifolia*. *J Appl Bacteriol* 1995; **78**(3): 264-9.

[23] Tadrent W, Kabouche A, Touzani R, Kabouche Z. Chemotypes investigation of essential oils of “Guertoufa” herbs. *J Mater Environ Sci* 2014; **5**(4): 1200-5.