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ESSENTIAL OIL COMPOSITION OF SOME *CENTAUREA* SP. (ASTERACEAE) FROM DIFFERENT ITALIAN ISLANDS

Abstract - *Essential oil composition of some Centaurea sp. (Asteraceae) from different Italian islands.* The volatile constituents of leaves and flower heads of several *Centaurea* species from different islands of Ligurian and Tyrrhenian Sea were investigated for the first time. *C. veneris* (Sommier) Bég. from Palmaria Island (Ligurian Sea), *C. gymnocarpa* Moris & De Not. from Capraia Island, *C. aetaliae* (Somm.) Bég. and *C. ilvensis* (Sommier) Arrigoni from Elba Island (Northern Tyrrhenian Sea); *C. aeolica* Lojac. subsp. *aeolica* from Lipari, Aeolian Islands, *C. busambarensis* Guss., *C. panormitana* Lojac. subsp. *ucraiae* (Lacaita) Greuter, *C. panormitana* Lojac. subsp. *umbrosa* (Fiori) Greuter, *C. panormitana* Lojac. subsp. *todaroi* (Lacaita) Greuter, *C. panormitana* Lojac. subsp. *seguenzae* (Lacaita) Greuter from different localities in Sicilia (southern Tyrrhenian Sea), were collected at flowering stage. All samples were extracted by steam distillation to obtain the volatile fraction with a yield ranging from 0.02-0.13% in leaves and 0.01-0.09% in flower heads (weight/fresh weight basis), respectively. The oils were then analyzed by GC/FID and GC/MS methods and more than 100 compounds belonging to several chemical classes were identified and quantified in all the samples. Sesquiterpenes represent the most abundant class of compounds (22.35-61.67% and 35.16-57.51% of the total volatiles in leaves and flower heads, respectively) of which germacrene D was the dominant constituent (10.03-42.65% and 7.33-30.32% in leaves and flower heads, respectively). Aldehydes, hydrocarbons, ketones, monoterpenes, alcohols, acids, esters and miscellaneous compounds were also identified and quantified in different amount in the volatile oils from all the examined plant organs. Other compounds were also detected and they appear to be species-specific, as their presence was only detected in either one or the other *Centaurea* species.

Key words - *Centaurea aeolica*; *C. aetaliae*; *C. busambarensis*; *C. ilvensis*; *C. gymnocarpa*; *C. panormitana* subsp. *ucraiae*, subsp. *umbrosa*, subsp. *todaroi* and subsp. *seguenzae*; *C. veneris*; Asteraceae; essential oil composition; GC/MS; Ligurian and Tyrrhenian sea Islands, Italy.

Riassunto - *Composizione di oli essenziali di alcune specie del genere Centaurea raccolte in diverse isole italiane.* Sono stati estratti per distillazione in corrente di vapore i costituenti volatili delle parti aeree (foglie e infiorescenze) di diverse specie appartenenti al genere *Centaurea*: *C. veneris* (Sommier) Bég., *C. gymnocarpa* Moris & De Not., *C. ilvensis* (Sommier) Arrigoni e *C. aetaliae* (Somm.) Bég. da diverse isole del Mar Ligure e del Mar Tirreno Settentrionale; *C. aeolica* Lojac. subsp. *aeolica* da Lipari, Isole Eolie; *C. busambarensis* Guss., *C. panormitana* Lojac. subsp. *ucraiae* (Lacaita) Greuter, *C. panormitana* Lojac.

subsp. *umbrosa* (Fiori) Greuter, *C. panormitana* Lojac. subsp. *todaroi* (Lacaita) Greuter, *C. panormitana* Lojac. subsp. *seguenzae* (Lacaita) Greuter da diverse località della Sicilia. La resa in olio essenziale è risultata essere compresa tra 0.02 e 0.13% per le foglie e tra 0.01 e 0.09% per le infiorescenze, calcolata sul peso fresco. Gli estratti sono stati quindi analizzati mediante GC/FID e GC/MS e più di 100 composti appartenenti a diverse classi chimiche sono stati identificati e quantificati. I sesquiterpeni rappresentano la classe più abbondante di composti (valutati rispettivamente 22.35-61.67% e 35.16-57.51% dei volatili totali in foglie ed infiorescenze), tra cui il germacrene D è risultato il composto dominante (valutato 10.03-42.65% dei volatili totali nelle foglie e 7.33-30.32% dei volatili totali nelle infiorescenze). In tutti i campioni analizzati sono stati identificati anche: aldeidi, idrocarburi, chetoni, monoterpeni, alcoli, acidi ed esteri in quantità minore. Da queste indagini sono stati inoltre evidenziati alcuni composti specie-specifici che sembra possano avere anche un significato chemotassonomico.

Parole chiave - *Centaurea aeolica*; *C. aetaliae*; *C. busambarensis*; *C. ilvensis*; *C. gymnocarpa*; *C. panormitana* subsp. *ucraiae*, subsp. *umbrosa*, subsp. *todaroi* e subsp. *seguenzae*; *C. veneris*; Asteraceae; composizione di oli essenziali; GC/MS; Isole dei Mari Ligure e Tirrenico, Italia.

INTRODUCTION

The genus *Centaurea* (Cardueae tribe, Asteraceae) is represented by a very large number of species (400-700) with predominantly Old World distribution (Hellwig, 2004; Bancheva & Greilhuber, 2006; Greuter, 2006-09; Greuter 2008; Hilpod *et al.*, 2011). Several papers on secondary metabolites of *Centaurea* species are available from literature (Baykan-Erel *et al.*, 2010), and a number of them are on volatile constituents (Rosselli *et al.*, 2009; Formisano *et al.*, 2010, 2011; Tava *et al.*, 2010; Viegi *et al.*, 2010, 2011, 2013; Esmaceli & Khodadadi, 2012; Jemia *et al.*, 2012; Politeo *et al.*, 2012; Demirtas & Sahin, 2012; Kilic, 2013). This taxon is very complex and could benefit from research using new cytological and chemical techniques.

The present study extends our work on *Centaurea* spe-

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cies in Italy and its aim was to investigate the essential oil composition of seven species from different Italian islands.

Four species were from Ligurian and Northern Tyrrhenian Sea Islands:

C. veneris (Sommier) Bég. is an endemic perennial species that grows among the calcareous rocks of Portovenere and the nearby islands of Palmaria, Tino and Tinetto, in the Ligurian Sea. Chromosome number is $2n=18$ (Viegi *et al.*, 1972b, sub. *C. cineraria* L. var. *veneris* (Somm) Bég.). This taxon has not yet been assessed for the IUCN Red List, but it is in the Catalogue of Life (<http://www.catalogueoflife.org/>);

C. gymnocarpa Moris & De Not is an endemic perennial species that inhabits among the acid rocks (vulcanites) of Capraia island (Tuscan Archipelago), in the northern Tyrrhenian Sea. Its localities have been described as "Habitat 8220" in the EC Habitats Directive. Chromosome number is $2n=18$ (Guinochet & Foissac, 1962; Viegi & Cela Renzoni, 1976). The species is considered endangered (EN), according to I.U.C.N. Red List criteria (Conti *et al.*, 1997; Foggi, 2006);

C. aetaliae (Sommiér) Béguinot is an endemic perennial species that grows on siliceous rocks of Mt Volterraio on Elba island, in the northern Tyrrhenian Sea. Chromosome number is $2n=18$ (Viegi & Cela Renzoni, 1976; Signorini *et al.*, 2001, sub *C. apolepa* Moretti subsp. *aetaliae* (Sommier) Dostál). This taxon has not yet been assessed for the IUCN Red List, but it is in the Catalogue of Life (<http://www.catalogueoflife.org/>);

C. ilvensis (Sommiér) Arrigoni is an endemic perennial species that inhabits among the acid rocks (granites) of Mt Capanne on Elba island, in the northern Tyrrhenian Sea. Chromosome number is $2n=18$ (Viegi & Cela Renzoni, 1976, sub *C. dissecta* Ten. var. *ilvensis* Sommier). This species has not yet been assessed for the IUCN Red List, but it is in the Catalogue of Life (<http://www.catalogueoflife.org/>).

Three species were from Southern Tyrrhenian Sea Islands and are all endemics:

C. aeolica Lojac. grows on volcanic rocks at Lipari (Aeolian Islands). Chromosome number is $2n=18$ (Viegi *et al.*, 1972b, sub. *C. aeolica* Guss. in Lojac.). This taxon has not yet been assessed for the IUCN Red List, and also it is not in the Catalogue of Life;

C. busambarensis Guss. inhabits on calcareous rocks at Mt Busambra and Isnello (PA). Chromosome number is $2n=18$ (Tornadore *et al.*, 1974; Cela Renzoni & Viegi, 1982; Devesa *et al.*, 1988). This taxon has not yet been assessed for the IUCN Red List, but it is in the Catalogue of Life;

C. panormitana Lojac., of which four subspecies were collected in different sicilian localities. This taxon has

not yet been assessed for the IUCN Red List, and also it is not in the Catalogue of Life;

7a - *C. panormitana* Lojac. subsp. *ucraiae* (Lacaita) Greuter (= *C. ucraiae* Lacaita) grows on arid rocks in front of the sea;

7b - *C. panormitana* Lojac. subsp. *umbrosa* (Fiori) Greuter (= *C. umbrosa* Lacaita) inhabits on shady rocks. Chromosome number is $2n=18$ (Viegi *et al.*, 1972b; Cela Renzoni & Viegi, 1982, sub. *C. ucraiae* Lac. subsp. *umbrosa* (Lac.) Cela Renzoni et Viegi);

7c - *C. panormitana* Lojac. subsp. *todaroi* (Lacaita) Greuter (= *C. todaroi* Lacaita) grows on arid rocks. Chromosome number is $2n=18$ (Brullo & Pavone, 1978, sub. *C. todari* Lacaita; Cela Renzoni & Viegi, 1982, sub. *C. ucraiae* Lac. subsp. *todari* (Lac.) Cela Renzoni et Viegi);

7d - *C. panormitana* Lojac. subsp. *seguenzae* (Lacaita) Greuter (= *C. todari* Lacaita subsp. *seguenzae* (Lacaita) Giardina & Raimondo) grows on arid rocks in front of the sea. Chromosome number is $2n=18$ (Viegi *et al.*, 1972b, sub. *C. todari* Lacaita forma *seguenzae* Lacaita). The nomenclature follows Greuter (2006).

C. gymnocarpa, *C. veneris*, *C. aeolica*, *C. busambarensis*, *C. panormitana* belong to the group known as the "cineraria" group (Viegi *et al.*, 1972a; Cela Renzoni & Viegi, 1982; Bancheva *et al.*, 2006; Hilpod *et al.*, 2011). This group probably was once a single species when the land masses were united, but as islands were formed, new species evolved on each island (I.U.C.N., 2014)

MATERIALS AND METHODS

Plant materials

The aerial parts (fresh and dry flower heads and leaves) of *C. veneris* from Palmaria, *C. gymnocarpa* from Capraia, as well as of *C. ilvensis* and *C. aetaliae* from Elba island (Mt Capanne and Mt Volterraio, respectively), as well as of Sicilian species, *C. aeolica* subsp. *aeolica* from Lipari (Aeolian Islands), *C. busambarensis* from Isnello (PA), *C. panormitana* subsp. *ucraiae* from Sferracavallo (PA), *C. panormitana* subsp. *umbrosa* from Mt Gallo and Mt Pellegrino, at Mondello (PA), *C. panormitana* subsp. *todaroi* from Bagheria (PA) (along motorway), *C. panormitana* subsp. *seguenzae* from Capo Tindari (ME) were collected during their flowering period (April-July) in 2006 and 2007. Voucher specimens of these plants are deposited in PI (Pisa University, Herbarium Horti Pisani). Numbers as follows: PI - New acquisitions - 9476 *Centaurea gymnocarpa*/ 5; PI - New acquisitions - 9476 *Centaurea veneris*/7; PI - New acquisitions - 9476 *Centaurea aetaliae*/ 3; PI - New acquisitions - 9476 *Centaurea ilvensis*/ 2; PI - New acquisitions -9476 *C. aeolica* subsp. *aeolica* /4; PI - New acquisitions - 9476 *C. busamba-*

rensis/6; PI - New acquisitions - 9476 *C. panormitana* subsp. *ucraiae* (sub. *C. ucraiae* subsp. *ucraiae*) from Sferracavallo/4; PI - New acquisitions - 9476 *C. panormitana* subsp. *umbrosa* (sub. *C. ucraiae* subsp. *umbrosa*) from Mt Gallo /4, PI - New acquisitions - 9476 *C. panormitana* subsp. *umbrosa* (sub. *C. ucraiae*) from Mondello (PA) /3; PI - New acquisitions - 9476 *C. panormitana* subsp. *todaroi* (sub. *C. todari*) from Bagheria (PA) /2; PI - New acquisitions - 9476 *C. panormitana* subsp. *seguenzae* (sub. *C. todari* subsp. *seguenzae*) /4.

For each population, a sample of 20 individuals was collected. Samples were stored in a corked glass bottle with 100 ml of CH₂Cl₂ as preservative and stored at 4°C until analysis.

Isolation of the essential oils

The tissue samples were steam distilled with odor-free water in a Clevenger-type apparatus to give 300 mL of distillate. This was saturated with NaCl, extracted with freshly distilled diethyl ether (3 × 100 mL), dried over anhydrous Na₂SO₄ and concentrated in a rotary evaporator to give a pale-yellow oil. The obtained essential oils were then analyzed by gaschromatographic methods.

Gas chromatography and gas chromatography-mass spectrometry

GC/FID analyses were carried out using a Perkin Elmer model 8500 GC equipped with a 30 m × 0.32 mm Elite-5MS capillary column (0.5 mm film thickness). Samples (0.5 mL) were injected in the split mode (1:30) with a column temperature program of 40°C for 5 min, then increased to 280°C at 4°C/min and finally held at this last temperature for 10 min. Injector and detector were set at 250°C and 300°C, respectively; the carrier gas was He with a head pressure of 12.0 psi. GC/MS analyses were carried out using a Perkin Elmer Clarus 500 GC equipped with a Clarus 500 mass spectrometer using the same capillary column and chromatographic conditions as for the GC-FID analyses. Mass spectra were acquired over 40–500 amu range at 1 scan/s with ionizing electron energy 70 eV, ion source 200°C. Transfer line was set at 300°C, carrier gas was He at 1.0 mL/min.

Identification and quantification of the essential oil components

The identification of the oil components was performed by their retention indices (RI), authentic reference compounds, peak matching library search, as well as published mass spectra (NIST/EPA/NIH, 2000; Joulain & König, 1998; Adams, 2007). Retention indices (RI) were calculated using n-alkane series (C6-C32) under the same GC conditions as for the samples. The relative amounts (RA) of individual components of the oil are expressed as percent peak area relative

to total peak area from the GC-FID analyses of the whole extracts. The amount of essential oil from leaves and flower heads of the examined *Centaurea* species ranged from 0.01 to 0.15% of fresh material.

RESULTS AND DISCUSSION

The volatile oil of the different *Centaurea* species under investigation contain several compounds belonging to different chemical classes, including sesquiterpenes, aldehydes, hydrocarbons, alcohols, monoterpenes, ketones, acids, esters and miscellaneous. The percentage composition of these classes of compounds in the volatile oils of leaves and flower heads of the *Centaurea* sp.pl. from different islands of Ligurian and Tyrrhenian Sea are listed in Table 1. As shown in this table, the most abundant classes of compounds were sesquiterpenes, ranging from 22.35 to 61.67% of the total volatiles in leaves and from 35.16 to 57.51% of the total oil in flower heads. The other classes of detected compounds were: aldehydes (6.48-15.42% in leaves, 6.47-24.47% in flower heads), hydrocarbons (1.67-18.26% in leaves, 8.09-24.29% in flower heads), alcohols (1.76-21.83% in leaves, 0.55-8.35% in flower heads), monoterpenes (1.24-2.74% in leaves, 0.59-2.45% in flower heads), ketones (0.34-11.85% in leaves, 0.71-3.51% in flower heads), acids (0.57-4.50% in leaves, 0.38-5.94% in flower heads), esters (0.75-4.67% in leaves, 0.05-1.45% in flower heads) and miscellaneous (0.09-7.72% in leaves, 0.24-1.21% in flower heads).

Concerning the single oil constituent, the sesquiterpene germacrene D is detected in all the analysed samples and represents one of the most abundant constituents. This compound is also reported as a constituent of the volatile fraction of other *Centaurea* sp.pl. from different countries (Binder *et al.*, 1990; Senatore *et al.*, 2003; Beck *et al.*, 2008; Rosselli *et al.*, 2009; Formisano *et al.*, 2010, 2011; Tava *et al.*, 2010; Viegli *et al.*, 2010; 2011; 2013; Esmaeili *et al.*, 2012; Demirtas *et al.*, 2012; Jamia *et al.*, 2012; Politeo *et al.*, 2012; Kilic, 2013).

Moreover, differences in the quantitative composition of the volatile fraction of the investigated *Centaurea* sp.pl. from Italian islands can be observed. In particular, the five most abundant detected compounds - listed in terms of percentage amount - in the seven *Centaurea* species were:

from Ligurian and Northern Tyrrhenian Sea Islands: *C. veneris*: germacrene D (42.65% leaves - 30.32% flower heads); *E*-caryophyllene (5.73% leaves, 5.80% flower heads); phenylacetaldehyde (4.52% leaves, 3.23% flower heads); bicyclogermacrene (3.28% leaves, 2.70% flower heads); α -ylangene (1.34% leaves, 4.31% flower heads);

Table 1 - Percentage composition of the most abundant chemical classes of compounds detected in *Centaurea* species from Ligurian (Palmaria), Northern Tyrrhenian Sea Islands (Capraia and Elba) and from Southern Tyrrhenian Sea Islands (Lipari, Aeolian Island and Sicily).

Kf*	Compounds ^b	Palmaria Island <i>C. venetis</i>		Capraia Island <i>C. gymnocarpa</i>		Elba Island <i>C. aetaliae</i>		Lipari (Aeolian Island) <i>C. aeolica</i>		Isolino (Sicily) <i>C. busambarensis</i>		Steraçavallo (Sicily) <i>C. panormitana</i> subsp. <i>ucifera</i>		Mt Gallo (Sicily) <i>C. panormitana</i> subsp. <i>umbrosa</i>		Mt Pellegrino (Mondello) (Sicily) <i>C. panormitana</i> subsp. <i>umbrosa</i>		Bagheria (Sicily) <i>C. panormitana</i> subsp. <i>todari</i>		Capo Tindari (Sicily) <i>C. panormitana</i> subsp. <i>segurginae</i>		
		leaves	flower heads	leaves	flower heads	leaves	flower heads	leaves	flower heads	leaves	flower heads	leaves	flower heads	leaves	flower heads	leaves	flower heads	leaves	flower heads	leaves	flower heads	
	Alcohols																					
	679 1-penten-3-ol	0.12	0.07	0.33	0.05	0.56	0.735	0.68	0.18	0.88	0.51	0.81	0.05	0.34	0.06	0.53	0.34	0.38	0.09	0.21	0.13	
	723 3-methyl-3-buten-1-ol	0.15	0.22	0.23	0.05	0.47	0.05	0.06	0.15	0.16	0.09	0.22	0.22	0.45	0.23	0.35	0.34	0.36	0.16	0.18	0.08	
	733 3-methyl-1-butanol	0.06	0.06	0.06	0.22	0.04	0.22	0.24	0.06	0.47	0.06	0.06	0.06	0.38	0.24	0.32	0.23	0.24	0.06	1.88	0.23	
	737 2-methyl-1-butanol	0.05	0.45	0.05	0.07	0.04	0.07	0.14	0.08	0.56	0.46	0.73	1.87	0.19	0.36	0.32	0.21	0.36	1.13	0.21	1.29	
	761 3,4-dimethyl pentanol							0.28	0.09	0.33	0.07	0.11	0.09	0.18	0.20	0.35	0.30	0.07	0.04	0.26	0.26	
	766 pentanol	0.02	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	
	768 cis-2-penten-1-ol							0.13	0.11	0.41	0.13	0.17	0.08	0.03	0.15	0.24	0.13	0.02	0.13	0.21	0.21	
	769 3-methyl-2-buten-1-ol							0.13	0.11	0.41	0.13	0.17	0.08	0.03	0.15	0.24	0.13	0.02	0.13	0.21	0.21	
	855 cis-3-hexen-1-ol	1.13	0.12	4.57	4.75	0.01	0.01	0.15	0.07	0.15	0.09	0.12	0.08	0.09	0.14	0.10	0.26	0.09	0.04	0.45	0.21	
	865 trans-2-hexen-1-ol			0.05	0.10	0.05	0.10	0.15	0.07	0.15	0.09	0.12	0.08	0.09	0.14	0.10	0.26	0.09	0.04	0.45	0.21	
	880 3-ol							0.04	0.04	0.03	0.03	0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.04	
	1035 benzyl alcohol	0.11	0.01	0.05	0.66	0.03	0.37	0.20	0.29	0.35	0.35	3.90	0.43	1.43	0.40	1.32	0.32	0.64	0.64	2.73	0.47	
	1070 octanol			0.14	0.05	0.16	0.14	0.16	0.16	0.16	0.16	1.78	0.41	1.02	0.28	0.94	0.24	2.94	0.70	1.22	0.44	
	1111 2-phenyl ethanol							0.01	0.01	3.01	0.29	tr	tr	tr	tr	0.03	1.49	tr	tr	0.07	0.07	
	1473 dodecanol							0.70	0.31	2.39	0.29	tr	tr	tr	tr	0.08	0.11	tr	tr	0.07	0.07	
	1678 tetradecanol																					
	Aldehydes																					
	650 3-methylbutanal	8.29	10.22	6.47	7.30	6.48	7.30	8.24	11.99	11.99	15.42	23.83	15.42	10.19	22.54	8.75	9.17	8.65	18.46	10.04	24.47	
	657 2-methylbutanal	0.44	0.28	0.41	0.56	0.41	0.56	0.36	0.88	0.51	0.51	5.79	2.23	0.41	0.04	1.50	0.97	0.24	0.38	0.82	4.34	
	753 2-hexenal	0.50	1.42	0.01	0.07	0.01	0.07	0.36	1.81	1.77	2.23	0.04	0.04	0.27	0.07	0.28	1.50	0.36	0.19	0.36	4.69	
	802 hexanal	0.23	1.87	0.33	0.79	0.21	0.30	0.12	0.09	2.46	tr	tr	tr	3.92	0.21	1.78	1.05	4.28	0.34	0.09	0.60	
	831 fufural																					
	853 trans-2-hexenal	0.63	0.18	0.34	tr	0.76	0.31	0.21	0.21	0.03	0.20	0.05	0.63	0.02	0.27	1.07	0.79	1.95	0.22	1.31	0.04	
	888 2-ethenyl-2-butenal	0.26	0.24	0.28	tr	0.15	0.07	0.71	0.08	0.64	0.04	0.00	0.83	0.20	0.37	0.13	0.02	0.13	0.02	0.01	0.88	
	902 heptanal	0.03	0.19	0.06	0.15	0.07	0.22	0.04	0.08	0.08	0.24	0.04	0.08	0.09	0.21	0.03	0.25	0.04	0.02	0.16	0.37	
	907 3-methylpropional	0.06	0.06	0.08	0.28	0.04	0.24	0.06	0.06	0.06	0.06	0.06	0.06	0.06	0.06	0.06	0.06	0.06	0.06	0.06	0.06	
	957 trans-2-heptenal	0.11	0.17	0.16	0.28	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	
	961 benzaldehyde	0.08	0.25	0.14	0.18	0.17	0.11	0.33	0.31	0.27	0.31	0.40	0.23	0.36	0.21	0.14	0.57	0.43	1.71	0.37	0.89	
	1002 octanal	0.03	0.25	0.25	0.13	tr	tr	0.07	0.07	0.33	0.12	0.22	0.23	0.54	0.17	0.75	0.54	0.03	0.58	0.14	0.42	
	1008 trans-trans-2,4-heptadienal																					
	1164 phenylacetaldehyde	4.52	3.23	3.15	3.62	3.01	3.62	2.34	3.29	3.76	2.70	7.01	2.70	2.08	12.73	1.29	2.02	6.15	2.02	2.30	5.02	
	1206 decanal	0.42	0.23	1.51	0.57	0.54	0.57	0.81	1.38	1.38	1.38	1.38	1.38	1.38	1.38	1.38	1.38	1.38	1.38	1.38	1.38	
	1262 trans-2-decenal	0.86	1.13	3.65	0.48	0.57	0.48	1.58	1.68	0.52	1.92	0.71	1.92	2.78	1.62	1.86	1.62	1.06	0.07	2.32	1.84	
	1307 undecanal																					
	1364 trans-2-undecenal	0.03	0.10	0.05	0.07	tr	tr	0.03	0.09	0.06	0.06	0.30	0.13	0.67	0.16	0.27	0.01	0.15	0.19	0.57	0.24	
	1408 dodecanal																					
	1714 hexadecanal																					
	Monoterpenes																					
	932 α -pinene	1.78	1.19	1.17	2.21	2.20	2.21	1.92	2.74	0.79	1.24	2.45	1.24	2.30	2.06	1.88	1.88	1.47	1.16	1.73	1.07	
	976 β -pinene	0.02	0.03	0.03	0.08	0.08	0.08	0.02	0.96	0.36	0.11	0.68	0.11	0.68	0.68	0.29	0.31	0.02	0.06	0.64	0.62	
	989 β -myrcene	0.05	0.87	0.08	0.26	tr	tr	0.13	0.09	0.04	0.06	0.33	0.10	0.21	0.04	0.04	0.06	0.19	0.19	0.09	0.12	
	1031 limonene																					
	1035 (Z)- β -ocimene																					
	1047 (E)- β -ocimene	1.67	0.25	0.16	0.59	1.04	0.59	1.12	0.07	0.07	0.06	0.03	0.22	0.36	0.03	0.33	0.12	0.16	0.16	0.03	0.08	
	1082 lincol oxide (furanoid)																					
	1099 linalool	0.03	tr	0.70	0.44	0.14	0.14	0.17	0.23	0.09	0.20	0.65	0.63	0.86	0.16	1.04	0.27	0.16	0.47	0.19	0.08	
	1195 p-menth-1-en-8-ol																					
	1196 α -terpineol	0.01	tr	0.20	tr	tr	tr	0.04	0.09	0.04	0.14	0.04	0.14	0.04	0.12	0.06	0.12	0.02	0.04	0.10	0.03	
	Sesquiterpenes																					
	1337 α -elemene	61.67	53.52	34.36	49.04	49.85	49.04	43.30	30.97	48.74	57.51	47.93	22.35	32.38	39.17	55.44	44.19	35.31	48.05	49.46	35.16	
	1349 α -cubebene	0.24	0.46	0.06	0.03	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	
	1359 α -longiphenene																					
	1373 α -ylangene	1.34	4.31	0.09	0.19	0.47	0.19	0.14	0.20	0.02	0.30	tr	0.05	0.15	tr	0.03	0.15	tr	tr	0.04	tr	
	1379 α -copaene	0.69	1.46	0.64	0.50	0.53	0.50	2.54	0.99	1.69	1.69	0.62	0.88	0.57	0.35	0.11	0.15	0.05	0.24	0.00	0.27	
	1391 β -elemene							1.68	0.26	0.29	1.02	0.26	0.59	2.44	0.26	0.33	0.82	1.00	0.66	1.00	0.47	
	1423 (E)-caryophyllene	5.73	5.80	4.88	8.96	8.96	9.79	14.86	5.71	17.79	1.94	8.14	2.67	7.57	0.03	4.93	14.45	5.03	11.33	4.52	9.97	
	1435 (E)- α -bergamotene	0.37	2.04	5.96	tr	tr	tr	0.39	0.23	1.41	0.00	0.61	0.00	0.13	0.03	0.30	tr	0.35	0.91	0.06	tr	
	1463 (E)- β -farnesene	0.53	0.58	0.04	0.89	0.89	0.89	0.11	0.16	0.46	0.24	0.12	0.12	0.49	0.09	0.18	0.11	0.16	0.10	0.33	0.22	
	1466 α -humulene	1.18	1.43	0.33	0.21	0.20	0.21	0.00	0.00	0.11	0.46	0.17	0.17	0.12	0.12	0.16	0.16	0.16	0.16	0.16	0.16	
	1478 γ -murolene																					
	1485 germacrene D	42.65	30.32	13.84	21.83	16.92	21.83	7.33	10.03	17.62	11.11	27.87	16.30	20.96	16.30	38.52	16.83	20.59	23.73	33.01	15.21	
	1490 (E)- α -farnesene							0.47	0.43	0.33	1.24	0.36	0.31	0.81	0.81	0.87	1.07					

1516 - cadinene	-	0.30	-	0.11	-	0.20	0.48	0.41	0.33	0.55	0.20	0.16	0.51	0.07	0.16	tr	0.32
1521 - β -cedrene	-	0.58	-	0.44	-	0.35	1.05	0.44	0.80	0.86	0.40	0.32	1.70	0.19	0.53	tr	0.87
1559 (E)-nerolidol	0.58	2.31	-	0.51	-	0.44	0.36	0.05	0.28	tr	0.07	0.06	tr	0.05	0.06	0.09	0.16
1564 germacrene B	-	-	0.58	-	0.44	0.35	0.48	0.26	0.05	0.77	0.21	0.37	0.31	-	0.06	tr	0.09
1581 spathulenol	-	-	-	-	-	-	0.19	0.26	tr	0.46	0.11	0.16	tr	0.23	tr	0.41	0.23
1587 caryophyllene oxide	-	-	-	-	-	-	0.46	4.33	0.73	1.71	1.01	0.88	1.39	1.69	2.89	1.32	1.83
1600 guaiol	-	-	-	-	-	-	0.08	0.15	0.14	0.13	0.10	0.18	0.12	0.25	0.20	0.43	0.43
1642 caryophylla-(14)(18)(15)-dien-5-ol	0.87	2.44	-	0.83	-	1.90	0.56	0.25	0.32	0.22	0.07	0.26	0.17	0.23	0.12	0.06	0.17
1647 epi- α -muroliol	-	-	-	-	-	-	0.22	0.16	0.22	0.14	0.54	0.86	0.13	0.20	0.39	0.02	0.10
1687 epi- α -muroliol	0.38	0.16	0.41	0.77	0.10	0.42	0.45	0.33	0.25	0.42	0.03	0.66	0.13	0.20	0.19	0.57	0.07
1689 gamma-4(15),5,10(14)trien-1-ol	2.36	0.97	10.09	3.32	4.61	0.95	1.12	0.95	0.77	1.01	0.59	0.61	1.19	0.59	0.91	1.18	0.75
1689 gamma-4(15),5,10(14)trien-1-ol	2.36	0.97	10.09	3.32	4.61	0.95	1.12	0.97	0.79	1.01	0.49	1.73	0.36	0.68	1.47	1.08	0.57
Esters																	
651 methyl propanoate	-	1.61	0.23	1.97	0.05	1.60	4.67	1.28	0.42	2.89	1.45	3.02	0.61	2.66	0.83	1.01	1.17
814 butyl acetate	-	-	-	-	-	-	0.25	0.05	0.02	tr	0.01	tr	tr	tr	tr	tr	tr
1005 cis-3-hexenyl acetate	0.50	0.01	1.01	0.05	1.06	3.84	-	tr	0.02	tr	0.04	tr	tr	tr	tr	0.03	0.96
1185 cis-3-hexenyl benzoate	0.04	-	-	0.13	-	0.36	0.09	-	0.05	2.12	0.01	2.44	0.05	1.09	0.03	0.11	0.01
1091 3-methyl-2-buten-1-ol acetate	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
1193 methyl salicylate	1.07	0.22	0.83	tr	0.18	0.74	-	0.37	0.03	0.43	0.05	0.38	-	0.94	0.02	0.31	-
Ketones																	
837 4-methyl-4-hydroxy-2-pentanone	2.39	1.61	1.69	0.71	0.94	0.47	10.73	3.30	4.95	9.35	2.12	3.13	1.16	10.87	1.36	4.34	3.81
957 tetrahydro-2H-furan-2-one	-	-	-	-	-	0.13	0.30	0.81	1.26	2.68	0.70	1.58	0.55	0.05	0.35	0.82	2.38
969 4-methyl-5H-furan-2-one	-	-	-	-	-	-	-	0.43	1.46	0.20	0.20	0.23	0.03	0.40	0.24	0.18	0.17
985 6-methyl-5-hepten-2-one	0.03	tr	0.03	tr	-	-	-	-	-	0.07	0.24	0.08	0.22	0.07	0.16	0.05	0.50
1008 4,5-dimethyl-4-hepten-3-one	2.31	1.29	1.33	0.58	-	-	7.49	0.05	1.94	tr	8.93	0.06	0.26	7.12	-	2.02	tr
1029 4-methyl-4-hepten-3-one	0.05	0.32	0.33	0.13	-	-	0.91	0.09	1.73	tr	1.51	1.62	tr	0.02	2.97	0.82	tr
1447 6,10-dimethyl-5,9-undecadien-2-one	-	-	-	-	-	-	0.14	tr	0.06	0.41	0.12	0.05	0.12	0.12	0.12	0.21	0.01
1841 6,10,14-trimethyl pentadecan-2-one	-	-	-	-	-	0.21	0.17	0.42	0.16	0.27	0.46	0.09	0.03	0.11	0.38	0.24	0.45
Hydrocarbons																	
797 4,5-dimethyl-1-hexene	5.87	10.26	10.86	14.70	1.87	4.76	18.26	10.23	10.46	12.95	11.05	11.65	11.65	10.12	8.09	6.83	20.81
800 3-methyl-2-hexene	0.11	0.06	0.22	-	-	-	1.40	0.05	1.05	0.13	0.79	0.26	0.26	0.99	1.14	1.36	0.18
1100 undecane	-	-	-	-	-	0.20	0.15	-	-	-	-	-	-	-	-	-	-
1291 1-tridecene	-	-	-	-	-	0.20	0.15	-	-	-	-	-	-	-	-	-	-
1300 tridecane	0.06	0.20	0.14	0.22	0.36	0.04	tr	0.01	0.01	0.33	0.21	0.59	0.27	0.16	0.98	tr	0.06
1492 1-pentadecane	2.08	0.67	7.57	6.87	6.86	3.96	18.19	3.06	18.19	0.76	1.46	0.78	9.17	1.52	3.91	0.39	3.96
1693 1-heptadecane	-	-	-	-	-	-	-	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr
2300 triecane	0.04	4.40	0.41	3.36	0.01	-	tr	tr	2.94	0.21	3.02	0.50	2.68	1.97	0.29	1.06	4.67
2300 pentacosane	0.09	1.39	0.25	1.30	0.09	0.10	0.26	0.35	0.35	1.53	0.76	0.88	0.34	0.49	0.26	0.39	2.03
2500 heptacosane	2.97	1.96	0.90	1.60	0.12	0.20	1.66	0.20	1.96	2.01	2.17	2.17	0.88	0.48	0.66	0.44	5.06
2900 heneicosane	0.02	0.03	1.33	1.15	0.05	0.05	0.97	0.14	1.16	2.16	3.75	3.75	0.88	2.77	2.03	2.03	2.03
3100 entriacontane	0.02	0.03	1.33	1.15	0.05	0.05	0.97	0.14	1.25	0.02	0.15	0.88	0.05	0.95	0.12	0.64	1.23
Acids																	
846 isovaleric acid	1.50	0.82	2.70	1.33	3.99	2.98	1.57	0.71	2.07	1.67	3.09	1.95	0.38	0.57	0.93	0.88	4.73
990 hexenoic acid	0.03	0.34	0.05	0.12	-	-	0.16	tr	tr	0.18	0.14	0.11	tr	tr	0.19	0.05	0.35
1761 tetradecanoic acid	-	-	-	-	-	-	tr	tr	-	-	-	-	-	-	-	-	-
1964 hexadecanoic acid	0.09	0.48	0.88	1.21	2.12	0.21	1.30	0.33	1.89	1.00	2.08	0.89	0.05	0.01	0.02	0.02	-
2129 linoleic acid	0.48	tr	0.92	tr	0.51	0.37	0.35	0.22	0.18	0.48	0.81	0.95	0.02	0.04	0.13	0.26	4.36
2135 linoleic	0.23	tr	0.34	tr	0.38	0.42	-	-	tr	0.01	0.06	tr	tr	0.02	tr	tr	tr
Miscellaneous																	
921 diethylsulfide	0.09	0.38	0.19	1.21	0.30	0.63	1.12	0.24	1.61	0.89	0.34	1.56	0.16	0.96	0.31	3.00	0.74
1217 2,3-dihydrozofurane	0.09	0.38	0.19	tr	-	-	-	-	-	-	-	-	-	-	-	-	-
1310 2-methoxy-4-vinyl phenol	-	-	-	-	0.15	0.35	0.67	0.14	0.46	0.55	0.18	0.91	0.12	0.63	0.18	0.96	0.21
1352 eugenol	-	-	-	-	0.01	0.07	0.21	0.02	tr	tr	0.02	0.15	0.04	0.06	0.01	0.03	tr
Total identified	84.96	79.24	65.76	62.72	72.07	79.41	77.80	93.83	86.84	91.10	88.04	91.10	88.04	92.44	85.79	86.40	97.01
Unidentified	15.04	20.76	34.24	37.28	27.93	20.69	22.20	6.17	11.16	18.92	13.15	8.38	26.72	7.56	14.21	13.60	2.99

*KI retention index determined on Elite-5 column using the homologous series of n-hydrocarbons. *Compounds grouped according to chemical class and listed in order of elution from an Elite-5 column.

C. gymnocarpa: germacrene D (13.84% leaves, 20.53% flower heads); 1-pentadecene (7.57% leaves, 6.87% flower heads); germacra-4(15),5,10(14)trien-1-ol (6.85% leaves, 0.09% flower heads); *E*-caryophyllene (4.88% leaves, 4.49% flower heads); phenylacetaldehyde (3.15% leaves, 2.85% flower heads);

C. aetaliae (leaves only): germacrene D (16.92%); germacra-4(15),5,10(14) trien-1-ol (10.09%); *E*-caryophyllene (8.96%); *cis*-3-hexen-1-ol (4.57%); phenylacetaldehyde (3.10%);

C. ilvensis (leaves only): germacrene D (21.83%); *E*-caryophyllene (9.79%); germacra-4(15),5,10(14) trien-1-ol (5.32%); *cis*-3-hexen-1-ol (4.75%); phenylacetaldehyde (3.82%).

From Southern Thyrrenian Sea Islands:

C. aeolica: germacrene D (10.03% leaves, 7.33% flower heads); *E*-caryophyllene (5.71% leaves, 14.86% flower heads); 4,5-di-methyl-4-hexen-3-one (7.49% leaves, 0.05% flower heads); *cis*-3-hexen-1-ol (4.38% leaves, 1.10% flower heads); germacra-4(15),5,10(14) trien-1-ol (4.61% leaves, 0.45% flower heads);

C. busambarensis: germacrene D (31.56% leaves, 17.62% flower heads); *E*-caryophyllene (5.22% leaves, 17.79% flower heads); *cis*-3-hexen-1-ol (6.22% leaves, 0.15% flower heads); phenylacetaldehyde (3.29% leaves, 3.76% flower heads); 1-pentadecene (3.06% leaves, 18.19% flower heads); benzyl alcohol (2.95% leaves, 0.35% flower heads);

7a. *C. panormitana* subsp. *ucraiae*: germacrene D (11.11% leaves, 27.87% flower heads); 4,5-di-methyl-4-hexen-3-one (8.93% leaves); phenylacetaldehyde (2.79% leaves, 7.91% flower heads); eugenol (7.17% leaves, 0.24% flower heads); *E*-caryophyllene (1.94% leaves, 8.14% flower heads);

7b. *C. panormitana* subsp. *umbrosa* (from Mt Gallo): germacrene D (16.30% leaves, 20.96% flower heads); *cis*-3-hexen-1-ol (5.56% leaves, 0.03% flower heads); *E*-caryophyllene (2.67% leaves, 7.57% flower heads); phenylacetaldehyde (2.16% leaves, 12.73% flower heads); 1-pentadecene (0.87% leaves, 1.95% flower heads);

7b. *C. panormitana* subsp. *umbrosa* (from Mt Pellegrino): germacrene D (38.52% leaves, 16.83% flower heads); *cis*-3-hexen-1-ol (1.23% leaves, 0.23% flower heads); *E*-caryophyllene (4.93% leaves, 14.45% flower heads); phenylacetaldehyde (1.29% leaves, 1.40% flower heads); 1-pentadecene (0.78% leaves, 9.17% flower heads);

7c. *C. panormitana* subsp. *todari*: germacrene D (20.59% leaves, 23.73% flower heads); *cis*-3-hexen-1-ol (9.13% leaves, 0.50% flower heads); *E*-caryophyllene (5.03% leaves, 11.33% flower heads); phenylacetaldehyde (2.02% leaves, 8.15% flower heads); 4,5-di-methyl-4-hexen-3-one (7.49% leaves);

7d. *C. panormitana* subsp. *sequenzae*: germacrene D (33.01% leaves, 15.21% flower heads); *E*-caryophyllene

(4.52% leaves, 9.97% flower heads); *cis*-3-hexen-1-ol (3.0% leaves, 0.2% flower heads); phenylacetaldehyde (2.30% leaves, 5.02% flower heads); 1-pentadecene (0.39% leaves, 3.96% flower heads).

Based on these results, a different trend can be observed for the *Centaurea* species from Northern and Southern Thyrrenian Islands under evaluation. As a general trend, the group of sesquiterpenes, that are the most abundant and representative constituents of the volatile oil and include germacrene D, *E*-caryophyllene, germacra-4(15),5,10(14)trien-1-ol, bicyclogermacrene and α -ylangene, were detected in lower amount in Sicilian species compared to Ligurian and Thyrrenian species, while the other classes of constituents, namely aldehydes, hydrocarbons, ketons, monoterpenes, alcohols, esters, acids, and miscellaneous, were detected in higher amount in Sicilian species than in Ligurian and Thyrrenian *Centaurea* plant extracts (see Table 1).

These differences, in most of the examined species, could be due to the local, ecological, climatic conditions of their habitat (Kilic, 2013). Additional researches are necessary to explain some debatable results.

The compounds identified in the volatile oils of the studied species only differed in quantitative composition, whereas a series of unidentified compounds seemed more species-specific, as beforehand observed in other Italian *Centaurea* species (Tava *et al.*, 2010; Viegi *et al.*, 2010, 2013).

As previously reported, these substances seem to belong to the class of polyunsaturated linear-chain compounds and their occurrence is not unusual, already having been reported in the Asteraceae family, including the genus *Centaurea* (Bohlmann *et al.*, 1966; Anderson *et al.*, 1977; Binder *et al.*, 1990). Further investigations are needed to identify these compounds, to compare their presence in other *Centaurea* volatile oils and to evaluate their role for a taxonomical point of view.

ACKNOWLEDGMENTS

The authors are grateful to Dr. Mirko Boracchia and Mr. Antonello Marchese for their assistance in the collection of plants on Palmaraia, Capraia and Elba Islands.

Financial support by M.I.U.R.-ex 60% and CRA is gratefully acknowledged.

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(ms. pres. il 21 luglio 2014; ult. bozze il 16 aprile 2015)