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Short communication

Essential oil diversity of *Origanum vulgare* L. populations from Southern Italy

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ABSTRACT

Essential oils (EOs) belonging to 25 wild populations of *Origanum vulgare* L. samples, growing wild in different locations of Calabria Region (Southern Italy), were analyzed using gas chromatography–mass spectrometry. The quantitative and qualitative data showed EO concentrations ranging from 0.96 to 5.10% and 37 compounds detected, representing more than 80% of the total composition of the oils. By applying hierarchical cluster analysis on the basis of the EO constituents, two main groups and three subgroups were found, reflecting the variation in the chemical composition of EOs from wild oregano populations. The first group consisted of acyclic (linalool/linalyl acetate) chemotypes with a predominant presence of linalyl acetate; the second was characterized by chemotypes rich in cymyl-compounds, mainly carvacrol, thymol and γ-terpinene.

The data obtained contribute to broaden the inventory of wild oregano populations from Calabria to plan programs for the selection of chemotypes with new and specific uses.

1. Introduction

Origanum vulgare L. is the most variable species of the *Origanum* genus, characterized by a great morphological and chemistry diversity (Tucker & Maciarello, 1994). The taxonomic revision by Ietswaart (1980) divided *Origanum vulgare* L. into six subspecies: *gracile* (Kock) Ietswaart, *glandulosum* (Desfontaines) Ietswaart, *hirtum* (Link) Ietswaart, *vulgare* L., *virens* (Hoffmannsegg et Link) Ietswaart and *viride* (Boissier) Hayek, depending on the indumentum differences, number of sessile glands and size/color of bracts and flowers (Kokkini, 1996). However, in spite of huge efforts to better classify *Origanum*, the literature confirms the large variation within species including the morphological diversity (Chalchat & Pasquier, 1999; D'Antuono, Galletti, & Bocchini, 2000), in addition to the quantitative and the qualitative differences in the compositions of the EOs produced (De Mastro, Ruta, & Marzi, 2004; Lukas, Schmiderer, & Novak, 2015).

Three subspecies, *O. vulgare* L. subsp. *glandulosum* (Desfontaines) Ietswaart, *O. vulgare* L. subsp. *hirtum* (Link) Ietswaart and *O. vulgare* L. subsp. *gracile* (Koch) Ietswaart, from the islands and southern mainland, are rich in EOs, whereas the other two, *O. vulgare* L. subsp. *virens* (Hoffmannsegg et Link) Ietswaart, *O. vulgare* L. subsp. *vulgare* L. and *O. vulgare* L. subsp. *viride* (Boissier) Hayek, which are distributed in the northern part of Europe, are considered to be poor in volatiles (Kokkini, 1996). The subspecies with an EO content >2%, are characterized by a more active 'cymyl'-pathway, which is involved in the biosynthesis of carvacrol, thymol (Mancini et al., 2014) and their biosynthetic precursors γ-terpinene and *p*-cymene (Bonfanti et al., 2012; Mechergui et al., 2010). The acyclic (linalool, linalyl acetate, β-ocimene, myrcene) (Shafiee-Hajiabad, Novak, & Honermeier, 2016), bicyclic (sabinene, cis-/*trans*-sabinene hydrate) (Crocoll, Asbach, Novak, Gershenzon, & Degenhardt, 2010), and/or bornane type compounds (camphor, borneol, bornyl acetate) (Lukas et al., 2015) monoterpenoids pathways are usually the feature of essential oil-poor subspecies with higher amounts of sesquiterpenes (β-caryophyllene, germacrene D, bicyclogermacrene, α- and γ-muurolene, β-caryophyllene oxide) (Skoula & Harborne, 2002).

The variability in EO composition and yield can result from geographical location of the collection site (De Martino, De Feo, Formisano, Mignola, & Senatore, 2009), different climatic and

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edaphic conditions (Prakash, Kanyal, Chandra, & Pant, 2013), and other environmental variables (Kumar, Shukla, Yadav, & Bagchi, 2007). Further, the quantitative and qualitative variations in EO are dependent on the plant origin (Figuérédo, Cabassu, Chalchat, & Pasquier, 2006), the harvesting season (Nurzyńska-Wierdak, 2009) and, of course, on intrinsic factors, such as sexual polymorphism or genetic mechanism, particularly the phenolic components (Vokou, Kokkini, & Bessiere, 1993).

Although variability in EO profiles, from wild *O. vulgare*, is intensively explored in different regions of Italy (De Martino et al., 2009; De Mastro et al., 2004; D'Antuono et al., 2000; Leto, Tuttolomondo, La Bella, & Licata, 2013; Napoli, Curcuruto, & Ruberto, 2009; Russo, Galletti, Bocchini, & Carnacini, 1998), more attention is needed regarding the genetic and chemical diversity of this important plant, considering the commercial perspective. Meeting the demand of the worldwide oregano market with low growing costs is of great interest to increase the profitability of small producers, adding value to oregano production. Obviously, the exploitation of wild local populations may represent the first action in achieving high quality products that are required by the final user (industry and/or consumer) (De Mastro, Fracchiolla, Verdini, & Montemurro, 2006).

The aim of the present investigation was to determine the chemical variability in 25 oregano populations originating from Calabria Region (Italy), extend the knowledge on their EOs and characterize a valuable source of EO, in order to provide a solid basis for better conservation and exploitation of this genetic resource.

2. Material and methods

2.1. Plant material

Samples of wild populations from *Origanum vulgare* L. were randomly collected, during the full flowering stage (end of June/middle of July), from 25 sites located in Calabria Region (Italy), at an altitude range of 347–1000 m above sea level (Table 1). The 25 accessions were identified and maintained *ex situ* (field collections) at the teaching and experimental farm "E. Pantanelli" of the University of Bari "A. Moro", Policoro (MT).

All plants were cut 10 cm above ground to ensure regrowth and were then dried in a forced convection oven (38 °C) until constant weight was achieved.

2.2. Essential oil extraction

Leaves and inflorescences from each sample (100 g) were submitted to hydrodistillation (Clevenger apparatus, 4 h). Then, the EO was dried over anhydrous sodium sulfate and stored in a dark glass vial at 4 °C for further analysis. The oil percentage was expressed as v/w based on dry matter of the initial material.

2.3. Composition of EO

The EOs were analyzed using an HP 6890 gas chromatograph coupled to an HP 5972 MSD and fitted with a capillary column HP-5MS (30 m × 0.25 mm × 0.25 μm film thickness). Analytical conditions were: helium as carrier gas (flow rate,1.1 mL min^{-1}), injector temperature 250 °C, detector temperature 300 °C, split mode (1:50), temperature program: 60–110 °C (2 °C min^{-1}), 110–220 °C (10 °C min^{-1}).

The components were identified based on their linear retention index relative to C8-C32 *n*-alkanes, comparison with data reported in the literature (John Wiley & Sons) and, whenever possible, co-injections with authentic samples. Pure standards were purchased from Sigma–Aldrich Co. S.r.l., Milan, Italy.

2.4. Statistical analysis

The percentage concentrations of the components in EOs were used as matrix elements to perform hierarchical cluster analysis. All PC analyses were carried out using SAS software (SAS Institute Inc., Cary, NC).

3. Results and discussion

The EO yields of 25 samples of *O. vulgare* collected from different sites of Calabria Region (Fig. 1) are reported in Table 1. The content of EO ranged from 0.96 to 5.10%, where the highest yield was recorded for Or4 population, followed by Or22 (4.30%) and Or12

Table 1
Collection sites and essential oil yield of investigated wild populations of *Origanum vulgare* L.

Accession/Population		Collection site		Altitude (m a.s.l.)	Latitude N	Longitude E	Oil yield% (v/w)
Or1	Scaramella	CZ	Gizzeria	630	38°59′11.89″	16°12′48.98″	2.23 ± 0.08
Or2	Gabella	CZ	Lamezia Terme	532	38°58′58.29″	16°16′24.76″	2.61 ± 0.03
Or3	Santa Maria	CZ	Lamezia Terme	839	38°59′32.52″	16°22′03.45″	3.42 ± 0.12
Or4	Crozzano	CZ	Lamezia Terme	576	38°59′22.14″	16°16′57.86″	5.10 ± 0.09
Or5	Campo di Maggio	CZ	Martirano Lombardo	590	39°04′24.98″	16°13′41.60″	2.95 ± 0.07
Or6	C.da Caria	CZ	Conflenti	853	39°04′17.76″	16°17′59.51″	2.30 ± 0.24
Or7	Pietra del Corvo	CZ	Martirano Lombardo	971	39°03′43.35″	16°13′39.63″	1.92 ± 0.11
Or8	Suvereto	CZ	Montepaone	367	38°43′22.13″	16°29′43.04″	1.75 ± 0.12
Or9	Cipino	CZ	Sersale	820	39°00′50.73″	16°42′43.84″	2.18 ± 0.08
Or10	Cumuna	CZ	Cropani	347	38°58′09.18″	16°46′58.59″	2.65 ± 0.21
Or11	Castania	CZ	Sersale	740	39°00′47.62″	16°44′13.48″	3.75 ± 0.04
Or12	Monte Sottano	CS	Rocca Imperiale	403	40°06′33.18″	16°34′41.78″	4.05 ± 0.05
Or13	Monte Soprano	CS	Canna	536	40°05′49.07″	16°29′49.41″	2.87 ± 0.17
Or14	Catosa	CZ	Sersale	740	39°00′28.01″	16°43′41.47″	2.41 ± 0.23
Or15	Serra Longa	CS	Rose	973	39°25′55.75″	16°22′09.75″	3.58 ± 0.11
Or16	Monte Raga	CZ	Sersale	500	39°00′33.92″	16°45′04.20″	2.30 ± 0.24
Or17	Torre d'Oranges	CS	Aprigliano	700	39°14′43.41″	16°20′17.64″	2.65 ± 0.21
Or18	Piano Stella	CS	Spezzano della Sila	800	39°18′00.53″	16°20′31.65″	0.96 ± 0.09
Or19	Vinacce	CS	Cerchiara di Calabria	650	39°51′36.38″	16°22′50.63″	2.12 ± 0.07
Or20	Granpollina	CS	San Lorenzo Bellizzi	830	39°53′15.18″	16°19′43.08″	1.02 ± 0.16
Or21	C.da Melizia	CS	Cerchiara di Calabria	650	39°51′31.37″	16°23′03.40″	1.88 ± 0.10
Or22	Feudo	CZ	Chiaravalle Centrale	418	38°48′25.26″	16°30′33.06″	4.30 ± 0.18
Or23	Colle Santa Maria	CZ	Fossato Serralta	904	38°59′41.70″	16°34′59.17″	2.58 ± 0.13
Or24	Quattromura	VV	Vazzano	722	38°37′53.95″	16°14′20.80″	3.75 ± 0.04
Or25	Sant'Elia	RC	Palmi	583	38°20′45.06″	15°50′27.06″	1.50 ± 0.27