Effect of the Altitude Geographic and Species Type on the Volatile Compounds of the Genus *Origanum* from Southern Perú

Huamán-Castilla, Nils L.; León, Nilton C.; Arroyo, Gregorio; Quintana, José
Escuela de Ingeniería Agroindustrial, Universidad Nacional de Moquegua, Prolongación calle Ancash s/n, Moquegua 18001, PERÚ

Maximiliano, Martínez-Cifuentes; Mariotti – Celis, María S.*+
Programa Institucional de Fomento a la Investigación, Desarrollo e Innovación, Universidad Tecnológica Metropolitana, Ignacio Valdivieso 2409, P.O. Box 9845, Santiago 8940577, CHILE

ABSTRACT: *Genus Origanum is an aromatic plant used in folk medicine and as a culinary herb, whose composition of volatile compounds is influenced by the type of species and geographical locations. This research evaluates the effect of altitude on the composition of volatile compounds and the contents of carvacrol and thymol of Origanum x majoricum* Cambess *and Origanum majorana L. Fifty samples of both species were recollected from different altitudes (2,500 – 3,500 MASL) of the Southern Perú and analyzed by solid-phase microextraction integrated to gas chromatography-mass spectrometry. The principal component analysis was used to differentiate the plants rich in thymol and carvacrol. Peruvian oregano presented 30 different volatile compounds between some monoterpenes (>30%) and some sesquiterpenes (>5%). The thymol and carvacrol contents of O. majorana and O. majoricum varied between 0.38 and 16.47% and 0.44 and 11.16%, respectively. Interestingly an inverse correlation of the concentration of thymol and carvacrol with their precursors (p-cymene and γ-terpinene) was also observed. Altitudes between 3,000 and 3,200 MASL favors the high proportions of volatile compounds. The data obtained contribute to planning programs for the selection of species and agricultural conditions that allow obtaining a better quality of oregano essential oil.*

KEYWORDS: *Origanum majoricum; Origanum majorana; Volatile compounds; Thymol and carvacrol.*

INTRODUCTION

Species of the genus *Origanum* (tribe Mentheae, Labiatae family), commonly named "oregano" are aromatic plants widely distributed in several countries of Latin American. Brazil, Mexico, and Perú present the highest production of this crop reaching 7,500; 6,000 and 4,000 tons/year; respectively [1]. Oregano

** To whom correspondence should be addressed.*
+ E-mail: mmariotti@utem.cl
1021-9986/2020/1/243-256 14/$/6.04

is used as a culinary ingredient worldwide due to its sensorial attributes and preservation effects [2] Additionally, due to its bioactive potential, pharmaceutical industry has shown interest in this vegetal raw material [3,4].

Genus *Origanum* is characterized by its high morphological and chemical variability which has given rise to a confusion in its correct taxonomic identification [5]. *Origanum x majoricum* Cambess and *Origanum majorana* L. are two species of the forty-nine species identified to date, whose essential oil content can vary between 6 and 10 mL per 100 g dry weight [6]. This essential oil is located in glandular trichomes of the leaf, which consists of various volatile compounds, alcohols, esters and aromatic substances [7]. The volatile compounds such as monoterpenes, sesquiterpenes, and oxygenated derivatives are responsible of the aroma and antioxidant properties of these species [8,9]. The species of *Origanum* are characterized by have a high concentration of hydrocarbon and oxygenated terpenes (> 50% peak area), followed by some sesquiterpenes (between 5 and 15% peak area) and a low proportion of phenolic terpenes (<5% peak area) [10]. Interesting, these species accumulates important concentrations of phenolic monoterpenes deriving from the cymyl-pathway (mainly carvacrol and thymol and their biosynthetic precursors ɣ-terpinene and *p*-cymene) [11]. These compounds are responsible of the characteristic aroma of oregano and additionally they have important antioxidant, antimicrobial, expectorant, antispasmodic and antibacterial effects [12, 13]. However, the content of the volatile compounds may vary according to the area, the climate and the harvest season [14]. Additionally, the flowering stage is characterized because during this period *Origanum* presents the highest concentration of volatile compounds [15].

Different methods can be used to obtain volatile compounds; these include steam distillation, Soxhlet extraction, simultaneous distillation-extraction, supercritical fluid extraction, and Solid-Phase Micro Extraction (SPME). In this sense, SPME allows absorbing volatile compounds without promoting the oxidation, hydrolysis, and isomerization of monoterpenes and sesquiterpenes due to the low extraction temperatures (~40 °C) [16, 17]. However, the use SPME could present some disadvantages such as the low affinity and adsorption to volatile compounds due to the limited presence of active sites in the fiber, decreasing their recovery [18]. Subsequently, all these compounds can be identified by gas chromatography and mass spectroscopy [19].

The principal components analysis (PCA) is a mathematical algorithm that reduces the dimensionality of the data at two principal components, along which the variation in the data is maximal [14]. This technique has been used to correlate volatile compounds (e.g., thymol and carvacrol) with the species of *Origanum*

Currently, there are not many reports about the effect of altitude and type of species on the content of volatile compounds present in the species of *Origanum*. This information is critical to develop feasible strategies which ensure the production of oregano with high contents of thymol and carvacrol. The present work identified the volatile compounds of two species (*Origanum xmajoricum* C. and *Origanum majorana* L.) by solid phase microextraction-gas chromatography-mass spectroscopy (SPME-GC-MS) and evaluated the existence of some correlation between the content of these compounds (thymol and carvacrol) and the geographical distribution (altitude).

EXPERIMENTAL SECTION

Samples

Fifty fresh samples (whole plant) of *Origanum x marjorana* C. and *Origanum majuricum* L. were collected during the flowering phase in March 2015, from different locations of the district of Torata, Region Moquegua from southern Perú, placed between 2500 and 3500 MASL (meters above sea level) (Table 1 and Fig. 1). After collection, all the samples were transported in microperforated paper and placed under shade (15 °C) for further chemical analysis.

Identification of volatile compounds by SPME and GC-MS

Volatile compounds of oregano samples were absorbed in a triple SPME fiber of CARBOXEN / DVB / PDMS (Sigma Aldrich, Steinheim, Germany) with a thickness of 50/30 μm. Before each analysis, the fibers were activated at 260 °C for 30 minutes, following the manufacturer's instructions. Then ~ 6 g of sample were placed in a glass chamber of 500 mL volume conditioned

Table 1: Zones where O. x majoricum and O. majorana L. were recollected.

Sample N	Site collection	Code	Taxonomic description	Altitude (MASL)	Sample N°	Site collection	Code	Taxonomic description	Altitude (MASL)
1	Torata	T1	*O. x majoricum*	3403.8	26	San June	S10	*O. majorana* L.	3173
2	Torata	T2	*O. x majoricum*	3405.5	27	San June	S11	*O. x majoricum*	3144
3	Torata	T3	*O. majorana* L.	3403.6	28	San June	S12	*O. x majoricum*	3206
4	Torata	T4	*O. majorana* L	3406.7	29	San June	S13	*O. majorana* L.	3194
5	Torata	T5	*O. majorana* L.	3410.4	30	San June	S14	*O. majorana* L	3231
6	Torata	T6	*O. majorana* L	3385.5	31	San June	S15	*O. majorana* L.	3219
7	Torata	T7	*O. x majoricum*	3384.9	32	San June	S16	*O. majorana* L	3278
8	Torata	T8	*O. x majoricum*	3435.4	33	San June	S17	*O. x majoricum*	3342
9	Torata	T9	*O. x majoricum*	3463.7	34	San June	S18	*O. x majoricum*	3380
10	Torata	T10	*O. majorana* L.	3473	35	San June	S19	*O. x majoricum*	3371
11	Torata	T11	*O. majorana* L	3482	36	San June	S20	*O. x majoricum*	3376
12	Torata	T12	*O. majorana* L	3413.2	37	San June	S21	*O. x majoricum*	3402
13	Torata	T13	*O. majorana* L.	3428.3	38	San June	S22	*O. x majoricum*	3418
14	Torata	T14	*O. majorana* L	3402	39	San June	S23	*O. x majoricum*	3414
15	Doce quebradas	D1	*O. majorana* L	2173.7	40	San June	S24	*O. x majoricum*	3389
16	Doce quebradas	D2	*O. majorana* L.	2174.5	41	Otora	O1	*O. x majoricum*	3472.02
17	San June	S1	*O. majorana* L	3184.8	42	Otora	O2	*O. x majoricum*	3463.25
18	San June	S2	*O. majorana* L	3185.5	43	Otora	O3	*O. x majoricum*	3441.26
19	San June	S3	*O. majorana* L	3183.6	44	Otora	O4	*O. x majoricum*	3445.24
20	San June	S4	*O. x majoricum*	3176.7	45	Porobaya	P1	*O. x majoricum*	2612.2
21	San June	S5	*O. x majoricum*	3170.4	46	Porobaya	P2	*O. x majoricum*	2621.2
22	San June	S6	*O. x majoricum*	3205.5	47	Porobaya	P3	*O. x majoricum*	2763.8
23	San June	S7	*O. x majoricum*	3304.9	48	Porobaya	P4	*O. x majoricum*	2734.9
24	San June	S8	*O. majorana* L.	3135.4	49	Porobaya	P5	*O. x majoricum*	2735.7
25	San June	S9	*O. majorana* L	3063.7	50	Porobaya	P6	*O. x majoricum*	2612.2

***MASL: meters above sea level.*

with 3 entrances for the SPMEs. The chamber was equilibrated at 40 °C in order to saturate it with the volatile compounds. After that, the SPMEs were exposed to the chamber (~ 15 min) to capture the volatile compounds. The analysis was done directly on dry material without grinding and the SPME fiber captured the volatilized compounds in the headspace of the chamber. Subsequently, the fibers were heated at 260 °C for 5 min in the gas chromatograph injector to desorb the volatiles. The desorbed compounds were analyzed by GC-MS using the following method: i) Program temperature of the oven: 40 ° C for 2 minutes. 40 - 150 °C at 5 °C/min, 150 - 170 °C at 10 °C/min, 170 -250 °C at 30 °C/min and at 250° C for 2 minutes; ii) Flow: 0.9 mL / min; iii) Injector temperature: 260 °C; iv) Flow division mode: Splitless (without division); v) A column HP-5ms ((5% -phenyl) -methylpolysiloxane) of 30 m long with 0.25 mm internal diameter and 0.25 µm thick stationary phase; vi) Equipment and detector: Agilent 7890B chromatograph with MultiMode Injector (MMI)

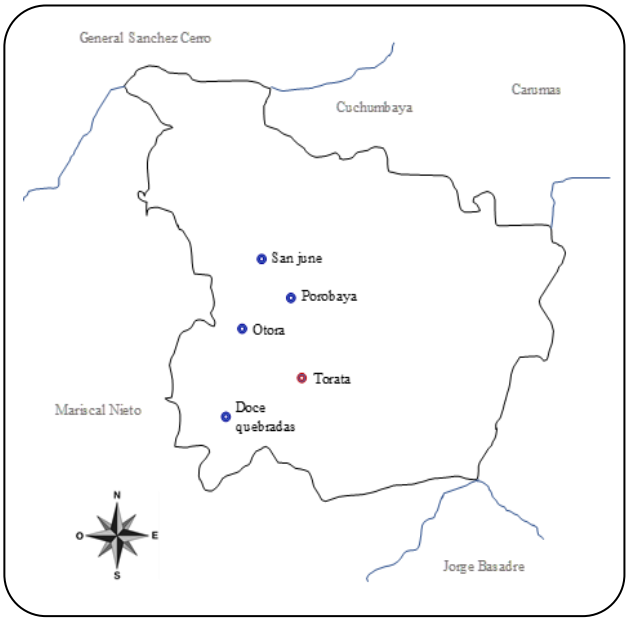

Fig. 1: Site collection of the district of Torata, Region Moquegua – Perú.

and selective quadrupole mass detector, Agilent 5977C with electron ionization at 70eV.

For the identification of volatile compounds, we used the NIST 2011 mass spectrums library, laboratory standards and the comparison of linear retention indexes. The purity was further evaluated using the AMDIS mass spectral deconvolution software version 2.68. The quantification of % areas under the peaks of the chromatogram was made with the help of the Chem Station program version F.01.00.1903 of Agilent.

For each compound obtained, the experimental retention index was calculated from a sample of calibration standards of linear alkanes (C8 to C20 - Sigma-Aldrich). Also, to verify the identity of the compounds, the retention times and fragmentation profiles were compared with the respective standards.

Statistical Analyses

Principal Component Analysis (PCA) was applied using the following variables such as *ɣ*-terpinene, *p*-cymene, thymol, and carvacrol to provide an overview of the ability to associate groups according to the volatile compounds of the different geographic zones of collection and spices of *Origanum*. Windows 4.0 statistical software (Statpoint Technologies, Inc., Virginia, USA) was used for this analysis

RESULTS AND DISCUSSION

Fifty individual plants (twenty-nine from *O. x majoricum* and twenty-one from *O. majorana* L.) were analyzed to identify their volatile compounds. Additionally, the effect of the altitude (msnm) on the thymol and carvacrol content of samples was determined by PCA.

Identification of volatile compounds in spices of oregano

Thirty different volatile compounds were identified in fifty samples of *O. majorana* L. and *O. x majoricum* collected from different sites of Moquegua Region (Table 1 and Fig. 1). Then, the compounds were classified according to their chemical structure such as monoterpenes hydrocarbons, oxygenated monoterpenes, sesquiterpenes hydrocarbons and phenolic monoterpenes (Table 2 and Supplementary material).

O. majorana L. presented between 0.09 and 18.08% of monoterpenes hydrocarbons, with majority presence of α-terpinene (>5%) and p-cymene (>4%). Oxygenated monoterpenes were determined in a range between 0 and 20.33% with predominance of linalool (>15%). Phenolic monoterpenes content varied between 0.01 and 16.47 % presenting an inversely proportional relationship between thymol and carvacrol. Contrary, the content of sesquiterpenes hydrocarbons was relatively low between 0.02 and 8.07%.

On the other hand, *O. x majoricum* samples contained lower amounts of monoterpenes hydrocarbons (<12.9%) and phenolic monoterpenes (<11.5%) compared to *O. majorana* L. samples (~21%), while the content of sesquiterpenes hydrocarbons was similar in both species (Table 2 and Supplementary material).

The variation between volatile compounds observed in *O. majorana* L and *O. x majoricum* could be associated to the different metabolic pathways of the cymyl-compounds (p-cymene, ɣ-terpinene, thymol methyl ether, carvacrol methyl ether, thymoquinone, thymol and carvacrol), sabinyl-compounds (sabinene, trans-/cis-sabinene hydrate) and linalool/acyclic linalyl acetate [11]. These metabolic pathways are known as mevalonic acid (MVA) and methylerythritol phosphate (MEP). The MVA pathway gives rise to volatile sesquiterpenes, while the MEP pathway provides precursors to volatile hemiterpenes, monoterpenes and diterpenes [20]. Additionally, the composition of volatile compounds

Table 2: Volatile compounds identified in some samples of Origanum, whose percentage of phenolic monoterpenes is > 10%

Compounds	Tr	LRI	% peak area							
			O. majorana L.				*O. x majoricum*			
			T10	S3	S13	S15	S18	S23	S7	P6
Monoterpenes hydrocarbons										
α-thujene	7.78	920	1.37	1.02	0.86	0.65	0.91	1.03	1.08	1.27
α -pinene	7.96	926	0.20	0.20	0.19	0.17	0.22	0.22	0.20	0.28
sabinene	9.18	968	2.93	2.11	1.30	1.17	1.80	2.01	2.10	2.56
β -myrcene	9.76	989	1.70	1.51	1.04	0.80	1.52	1.38	1.03	1.26
α -phellandrene	10.12	1001	1.00	1.00	0.84	0.76	0.62	0.77	0.45	1.12
α -terpinene	10.52	1014	4.50	4.77	3.31	2.76	3.78	4.13	2.66	4.39
p-cymene	10.79	1022	9.24	3.84	2.06	3.08	3.92	2.24	1.39	4.93
(*Z*)-β-ocimene	11.22	1036	1.59	2.03	1.55	1.74	2.61	2.39	1.18	0.36
(*E*)- β-ocimene	11.53	1047	0.49	0.65	0.67	0.63	2.19	1.73	0.55	0.28
γ -terpinene	11.87	1058	9.06	8.95	6.01	0.36	0.70	0.63	5.18	6.54
limonene	10.90	1026	4.34	3.28	1.98	5.37	8.42	7.37	1.55	2.98
terpinolene	12.73	1085	1.69	1.81	1.44	1.28	1.49	1.68	1.04	1.89
Oxygenated monoterpenes										
1-octen-3-ol	9.41	977	0.34	0.42	0.26	0.18	0.34	0.46	0.37	0.33
cis-sabinene hydrate	12.09	1064	1.94	1.86	1.42	1.78	2.53	1.99	1.58	1.86
trans-sabinene hydrate	13.11	1097	-	-	-	-	-	-	-	-
linalool	13.14	1100	17.89	16.62	10.70	11.37	18.32	18.45	20.43	15.58
terpinen-4-ol	15.47	1179	4.51	5.85	5.80	7.04	6.68	3.69	2.81	9.03
α -terpineol	15.84	1191	2.42	2.80	2.80	2.72	3.05	2.42	3.02	--
thymol methyl ether	17.09	1236	3.23	3.76	4.60	4.48	2.99	4.21	4.22	2.96
carvacrol methyl ether	17.34	1246	4.66	5.04	5.94	6.11	5.62	6.12	6.45	5.31
Linalyl acetate	17.69	1259	6.86	8.16	10.93	1.83	0.87	0.56	11.75	7.59
thymoquinone	17.51	1251	0.64	0.66	1.14	6.21	9.81	10.11	0.46	0.54
Phenolic monoterpenes										
thymol	18.72	1295	10.11	10.06	13.54	16.47	9.60	10.00	11.16	11.52
carvacrol	18.96	1303	1.18	0.38	0.55	0.83	0.47	0.47	0.44	1.11
Sesquiterpenes hydrocarbons										
caryophyllene	22.14	1422	4.31	6.05	8.07	7.42	4.71	7.66	8.52	5.97
Aromadendrene	22.61	1441	0.46	0.81	2.25	1.22	0.77	1.16	1.80	0.77
humulene	22.98	1455.2	0.59	0.90	1.59	1.48	0.72	1.12	1.32	0.48
alloaromadendrene	23.15	1462.4	1.95	0.11	0.26	0.21	0.11	0.12	0.15	0.22
α -muroleno	24.06	14.98.9	0.21	3.95	7.34	6.63	3.96	5.21	6.35	2.96
β-bisabolene	24.31	1510.3	0.00	0.15	0.34	4.26	0.10	0.16	0.14	0.16

The values (relative peak area percent) represent averages of three determinations. LRI: Linear retention index relative. Tr: retention time (min) relative to standard mixture of n-alkanes.

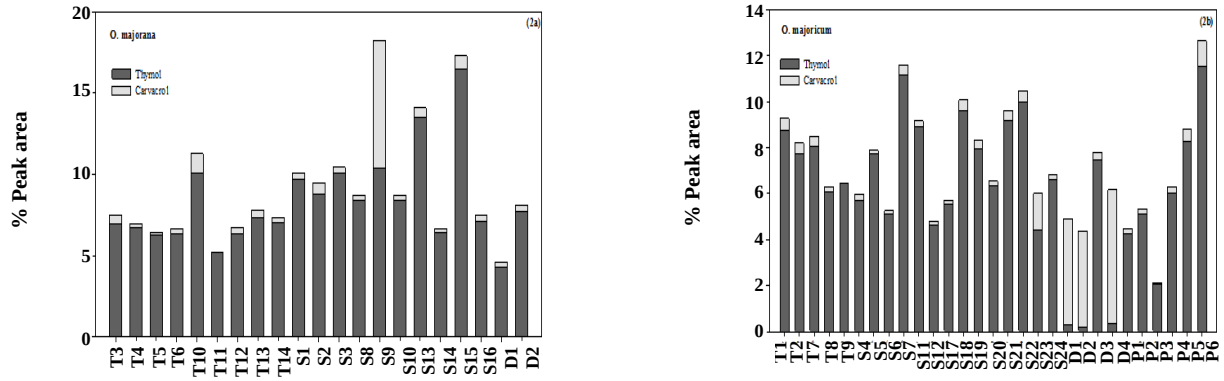

Fig. 2: Thymol and carvacrol content in the samples recollected.

present in the essential oil of can vary depending on the species type, agricultural practices, soil type, climatic conditions and harvest time.

T10, S3, S13 and S15 for *O. majorana* L. and S18, S23, S7 and P6 for *O. x majoricum* presented the highest content (>10%) of phenolic monoterpenes (Table 2). These compounds play a key role in the aroma and bioactive properties of the essential oil of oregano [17,21]. Nevertheless, it is important to mention that phenolic monoterpenes such as thymol and carvacrol are also considered oxygenated monoterpenes due to the presence of oxygen in their chemical structure which could explain their antioxidant properties [22].

Analysis of the thymol and carvacrol content

The samples of *O. majorana* L. presented relative higher concentrations of thymol (from 4.28 to 16.47%) compared to *O. x majoricum* samples (from 2.05 to 11.52 %). The content of carvacrol was low in both species (< 7% and < 4% respectively), however only the samples de *O. x majoricum* (O1, O2 and O4) showed high proportions of carvacrol (from 4.21 to 7.91%) with respect to thymol (Fig. 2a & 2b). Because of the high amount of thymol presents in both species, these can be considered an attractive raw material for food, pharmaceutical and cosmetic industries [23].

The inverse relation found between the contents of thymol and carvacrol could be associated to the fact that these volatile compounds present different precursors (*p*-cymene and α-terpinene for thymol and carvacrol, respectively) [24,25]. The low concentrations of carvacrol detected both in *O. x majoricum* and *O. majorana* L. samples can be associated with a decreasing in the α-terpinene content (Table 2) cause by a phenomenon of oxidation and hydroxylation of *p*-cymene [26]. Samples of *O. x majoricum* and *O. majorana* L. showed high proportions of p-cymene and ɣ-terpinene (> 15%). The low temperatures in fall would reduce the hydrolase enzyme activity resulting in an accumulation of p-cymene and ɣ -terpinene [27].

PCA 1 and PCA 2 analyses of O. *majorana* L. explained the 75.96% of the variability of the results with 50.14 and 25.82% respectively. PC1 and PC2 allowed to separate the plants rich in thymol and carvacrol, respectively. Only S9 showed high concentrations in both compounds. With respect to *O. x majoricum*, the PCA 1 and PCA 2 explained the 82.14% of the variability of the results with 62.02 and 20.10% respectively. Only O4, O2 and O1 samples were rich in carvacrol. Additionally, both for O. *majorana* L. and *O. x majoricum;* PCA clearly distinguished the inverse correlation between thymol and carvacrol and their respective precursors (Fig. 3a & 3b).

Effect of the altitude on thymol and carvacrol content

Depending on the geographic altitude differences in the content of thymol and carvacrol in *O. x majoricum* and *O. majorana* L. were observed (Fig. 4a & 4b). Altitudes between 3,000 and 3,200 MASL promoted important concentrations of thymol and carvacrol between 16.4% and 7.9 %, respectively. On the other hand, altitudes above 3200 MASL reduced their presences. This behavior was reported in other studies which found the positive effect of altitude on the content of thymol and carvacrol in oregano grown between 100 and 2,500 MASL [27,28].

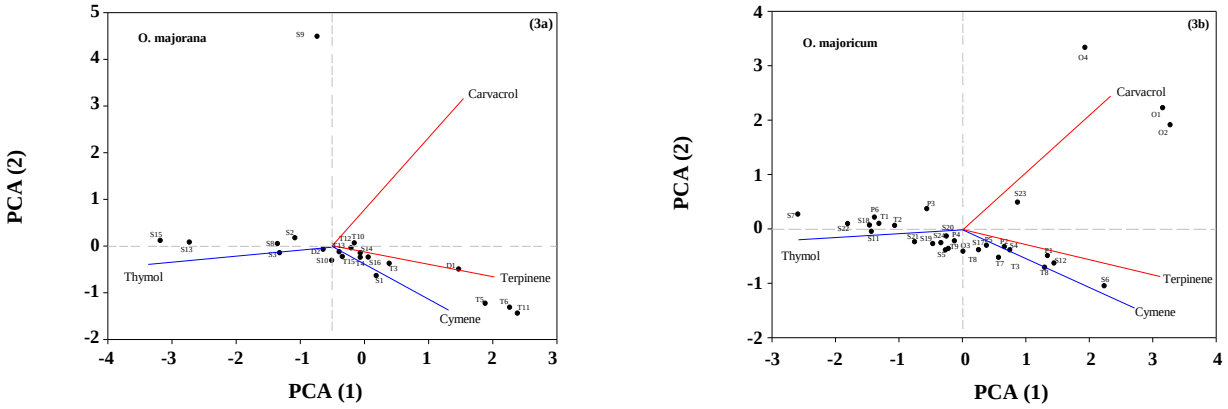

Fig. 3: Principal components analysis (PCA) that groups the zones according to their thymol and carvacrol content.

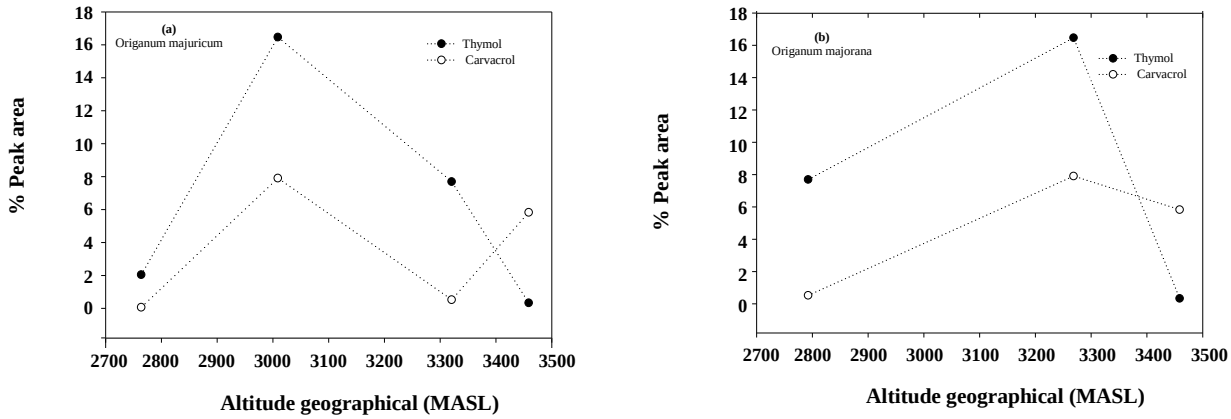

Fig. 4: Effect of geographic altitude on thymol and carvacrol content.

CONCLUSIONS

Species of *O. x majoricum* and *O. majorana* L. showed a wide diversity in the composition of volatiles, with important amount of thymol (>11 % and > 16% respectively) and low concentrations of carvacrol (from 0 to 4.62 %and from 0 to 7.9 %, respectively). The variability in the composition was probably caused by differences in the geographical area of collection (altitude) as well as the spice types. Additionally, samples collected at altitudes between 3,000 and 3,200 MASL showed the highest content of thymol and carvacrol (between 16.4% and 7.9 %, respectively). Thus, the microclimatic variation might act as an important force in the selection of specialized genotypes to obtain a good quality essential oil.

Conflict of interest

The authors declare that there are not conflicts of interest in this research work.

Acknowledgements

The authors appreciate the financial support of the Universidad Nacional de Moquegua - Perú.

Received : Jan. 31, 2018 ; Accepted : Nov. 5, 2018

REFERENCES

[1] Chishti S., Kaloo Z.A., Sultan, P., Medicinal Importance of Genus Origanum : A Review. *J. Pharmacogn. Phyther.*, **5** (10): 170–177 (2013).

[2] Black C., Haughey S.A., Chevalier O.P., Galvin P., Elliott C., Comprehensive Strategy to Detect the Fraudulent Adulteration of Herbs, *Food Chem.*, **210**: 551–557 (2016).

[3] Sellami I.H., Maamouri E., Chahed T., Wannes W.A., Kchouk M.E., Marzouk B., Effect of Growth Stage on the Content and Composition of the Essential Oil and Phenolic Fraction of Sweet Marjoram (Origanum Majorana L.). *Ind. Crops Prod.*, **30** (3): 395–402 (2009).

[4] Tabanca N., Özek T., Baser K.H.C., Tümen G., Comparison of the Essential Oils of Origanum Majorana L. and Origanum x Majoricum Cambess. *J. Essent. Oil Res.*, **16** (3): 248–252 (2004**)**.

[5] Bernath J., "Some Scientific and Practical Aspects of Production and Utilisation of Oregano in Central Europe", *Oregano Proceedings of the IPGRI International Workshop on Oregano*, May 8-12, CIHEAM, Valenzano, Bari, Italy (1996).

[6] Moncada J., Tamayo J.A., Cardona C.A., Techno-Economic and Environmental Assessment of Essential Oil Extraction from Oregano (Origanum Vulgare) and Rosemary (Rosmarinus Officinalis) in Colombia. *J. Clean. Prod.*, **112**: 172–181 (2016).

[7] Baranska Q.M., Chemotaxonomy of Aromatic Plants of the Genus Origanum via Vibrational Spectroscopy, *Anal. Bional. Chem.*, **381**: 1241–1247 (2005).

[8] Chalchat J.C., Pasquier B., Morphological and Chemical Studies of Origanum Clones: Origanum Vulgare L. Ssp. Vulgare. *J. Essent. Oil Res.*, **10** (September): 119–125 (1998).

[9] Mockute D., Bernotiene G., Judzentiene A., The Essential Oil of Origanum Vulgare L. Ssp. Vulgare Growing Wild in Vilnius District (Lithuania). *Phytochemistry*, **57** (1): 65–69 (2001**)**.

[10] Fornari T., Vicente G., Vázquez E., García-Risco M.R., Reglero G., Isolation of Essential Oil from Different Plants and Herbs by Supercritical Fluid Extraction. *J. Chromatogr. A*, **1250**: 34–48 (2012).

[11] Lukas B., Schmiderer C., Novak J., Essential Oil Diversity of European Origanum Vulgare L. (Lamiaceae). *Phytochemistry*, **119**: 32–40 (2015).

[12] Fachini-Queiroz F.C., Kummer R., Estevão-Silva C.F., Carvalho M.D.D.B., Cunha J.M., Grespan R., Bersani-Amado C.A., Cuman R.K.N., Effects of Thymol and Carvacrol, Constituents of Thymus Vulgaris L. Essential Oil, on the Inflammatory Response. *Evidence-based Complement. Altern. Med.*, **2012**: 1 - 10 (2012).

[13] Suntres Z.E., Coccimiglio J., Alipour M., The Bioactivity and Toxicological Actions of Carvacrol. *Crit. Rev. Food Sci. Nutr.*, **55** (3): 304–318 (2015).

[14] Gong H.Y., Liu W.H.,LV G.Y., Zhou X., Analysis of Essential Oils of Origanum Vulgare from Six Production Areas of China and Pakistan. *Brazilian J. Pharmacogn.*, **24** (1): 25–32 (2014**)**.

[15] Kizil S., Ipek A., Arslan N., Khawar K.M., Some Agronomical Characteristics and Essential Oil Content of Oregano (Origanum Onites l.) as Influenced by Planting Densities, *J. Essent. Oil-Bearing Plants*, **12** (2): 172–180 (2009**)**.

[16] Diaz-Maroto M.C., Perez-Coello M.S., Cabezudo M.D., Headspace Solid-Phase Microextraction Analysis of Volatile Components of Spices, *Chromatographia*, **55** (11–12): 723–728 (2002).

[17] Asadollahi-Baboli M., Aghakhani A., Headspace Adsorptive Microextraction Analysis of Oregano Fragrance Using Polyaniline-Nylon-6 Nanocomposite, GC-MS, and Multivariate Curve Resolution, *Int. J. Food Prop.*, **18** (7): 1613–1623 (2015).

[18] Murray R.A., Limitations to the Use of Solid-Phase Microextraction for Quantitation of Mixtures of Volatile Organic Sulfur Compounds. *Anal. Chem.*, **73** (7): 1646–1649 (2001).

[19] Fan Y., Li Z., Xue Y., Hou H., Xue C., Identification of Volatile Compounds in Antarctic Krill (Euphausia Superba) Using Headspace Solid-Phase Microextraction and GC-MS. *Int. J. Food Prop.*, **20** (sup1): S820–S829 (2017).

[20] Dudareva N., Klempien A., Muhlemann J.K., Kaplan I., Biosynthesis, Function and Metabolic Engineering of Plant Volatile Organic Compounds. *New Phytol.*, **198** (1): 16–32 (2013).

[21] Crocoll C., Asbach J., Novak J., Gershenzon J., Degenhardt J., Terpene Synthases of Oregano (Origanum Vulgare L.) and Their Roles in the Pathway and Regulation of Terpene Biosynthesis. *Plant Mol. Biol.*, **73** (6): 587–603 (2010).

[22] Bina F., Rahimi R., Sweet Marjoram: A Review of Ethnopharmacology, Phytochemistry, and Biological Activities. *J. Evidence-Based Complement. Altern. Med.*, **22** (1): 175–185 (2017).

[23] Rajput J.D., Bagul S.D., Pete U.D., Zade C.M., Padhye S.B., Bendre R.S., Perspectives on Medicinal Properties of Natural Phenolic Monoterpenoids and Their Hybrids. *Mol. Divers.*, **22** (1): 225–245 (2018).

[24] Napoli E.M., Curcuruto G., Ruberto G., Screening of the Essential Oil Composition of Wild Sicilian Rosemary, *Biochem. Syst. Ecol.*, **38** (4): 659–670 (2010).

[25] Nurzyńska-Wierdak R., Herb Yield and Chemical Composition of Common Oregano (Origanum Vulgare L.) Essential Oil According to the Plant's Developmental Stage. *Herba Pol.*, **55** (3): 55–62 (2009).

[26] Crocoll C., "Biosynthesis of the Phenolic Monoterpenes , Thymol and Carvacrol , by Terpene Synthases and Cytochrome P450s in Oregano and Thyme", Dissertation (Thesis), Facultad de Biología y Farmacia Universidad Friedrich Schiller Jena, Alemania (2011).

[27] Vokou D., Kokkini S., Bessiere J.-M., Geographic Variation of Greek Oregano (Origanum Vulgare Ssp. Hirtum) Essential Oils. *Biochem. Syst. Ecol.*, **21** (2): 287–295 (1993).

[28] Zouari S., Ayadi I., Fakhfakh N., Jdir H., Aloui L., Kossentini M., Rebai A., Zouari N., Essential Oil Variation in Wild Populations of Artemisia Saharae (Asteraceae) from Tunisia: Chemical Composition, Antibacterial and Antioxidant Properties. *Bot. Stud.*, **55** (1): 1–10 (2014).

Supplementary material 1A: Compounds identified in the samples of Origanum

Compounds	Tr	LRI	% peak area									
			T1	T2	T3	T4	T5	T6	T7	T8	T9	T10
Monoterpenes hydrocarbons												
α-thujene	7.78	920	1.16	1.23	1.10	1.06	1.23	1.65	2.02	1.61	2.22	1.37
α-pinene	7.96	926	0.17	0.21	0.16	0.19	0.17	0.19	0.20	0.24	0.46	0.20
Sabinene	9.18	968	2.31	2.47	1.93	2.56	2.34	2.76	3.79	3.50	4.56	2.93
β-myrcene	9.76	989	1.41	1.33	1.77	1.84	1.62	1.59	1.91	1.62	1.42	1.70
α –phellandrene	10.12	1001	1.11	1.46	2.13	1.50	3.81	3.34	1.18	1.47	0.98	1.00
α –terpinene	10.52	1014	4.69	5.24	7.40	5.40	11.79	12.86	6.30	7.36	5.51	4.50
p-cymene	10.79	1022	2.70	2.31	5.80	5.99	7.07	7.56	9.03	9.46	5.06	9.24
Limonene	10.90	1026	2.91	3.40	4.54	4.42	7.28	6.89	5.05	5.05	4.24	4.34
(*Z*)- β – ocimene	11.22	1036	2.03	1.74	0.92	1.70	1.85	0.69	1.08	1.04	1.30	1.59
(*E*)- β –ocimene	11.53	1047	0.66	0.59	0.51	0.52	0.57	0.33	0.40	0.36	0.33	0.49
ɣ-terpinene	11.87	1058	8.11	8.34	10.85	9.43	16.64	17.66	12.57	12.57	10.67	9.06
Terpinolene	12.73	1085	1.87	1.96	3.32	1.95	3.57	3.98	1.98	2.25	1.73	1.69
Oxygenated monoterpenes												
1-octen-3-ol	9.41	977	0.63	0.56	0.32	0.24	0.20	0.00	0.28	0.28	0.45	0.34
(*Z*)-sabinene hydrate	12.09	1064	2.12	1.62	1.34	1.72	1.05	1.40	1.86	2.33	2.07	1.94
(*E*)-sabinene hydrate	13.11	1097	0.00	0.00	7.43	13.80	6.32	6.46	15.70	16.91	0.00	0.00
Linalool	13.14	1100	10.44	15.63	1.16	1.14	1.59	1.00	0.73	0.66	21.63	17.89
terpinen-4-ol	15.47	1179	7.81	8.20	13.60	8.60	6.74	8.60	4.51	5.99	3.74	4.51
α –terpineol	15.84	1191	3.64	3.87	3.48	3.34	1.90	1.92	2.24	1.99	2.40	2.42
thymol methyl ether	17.09	1236	3.57	3.04	2.90	4.05	2.80	1.87	3.13	2.97	3.01	3.23
carvacrol methyl ether	17.34	1246	6.28	5.27	5.41	5.00	2.78	2.78	3.99	3.53	3.37	4.66
Thymoquinone	17.51	1251	0.52	0.37	0.16	0.38	0.19	0.11	0.39	0.34	0.00	0.64
Linalyl acetate	17.69	1259	10.41	8.87	5.20	6.85	3.97	3.31	5.75	5.89	0.67	6.86
Phenolic monoterpenes												
Thymol	18.72	1295	8.73	7.71	6.94	6.68	6.23	6.32	8.05	6.05	6.45	10.11
Carvacrol	18.96	1303	0.54	0.48	0.53	0.28	0.19	0.35	0.43	0.24	0.00	1.18
Sesquiterpenes hydrocarbons												
Caryophyllene	22.14	1422	6.94	6.20	5.77	5.26	4.11	3.11	4.41	3.26	9.19	4.31
Aromadendrene	22.61	1441	1.17	1.10	0.59	0.54	0.57	0.31	0.45	0.30	0.35	0.46
α-humulene	22.98	1455.16	1.04	0.85	0.74	0.68	0.56	0.37	0.56	0.42	3.96	0.59
Alloaromadendrene	23.15	1462.36	0.13	0.09	0.10	0.09	0.11	0.06	0.08	0.06	0.78	1.95
α –muroleno	24.06	1498.88	4.32	3.05	1.27	1.76	0.84	0.51	1.06	0.93	0.51	0.21
β –bisabolene	24.31	1510.3	0.22	0.18	0.18	0.23	0.15	0.07	0.25	0.10	2.19	0.00

The values (relative peak area percent) represent averages of three determinations. LRI: Linear retention index relative; Tr: retention time (min) relative to standard mixture of n-alkanes.

Supplementary material 1A (continue): Compounds identified in the samples of Origanum

Compounds	Tr	LRI	% peak area									
			T11	T12	T13	T14	S1	S2	S3	S4	S5	S6
Monoterpenes hydrocarbons												
α-thujene	7.78	920	0.44	1.75	1.95	2.25	2.20	1.53	1.02	1.28	1.77	0.91
α-pinene	7.96	926	0.09	0.23	0.27	0.36	0.30	0.29	0.20	0.25	0.22	0.18
Sabinene	9.18	968	0.86	2.74	3.15	3.81	3.85	3.18	2.11	2.39	3.53	2.13
β-myrcene	9.76	989	2.11	1.73	1.15	1.48	1.85	1.73	1.51	1.97	1.10	1.68
α –phellandrene	10.12	1001	5.73	1.25	1.13	1.32	1.82	0.81	1.00	1.28	0.81	1.65
α –terpinene	10.52	1014	12.68	4.78	5.67	5.43	6.84	3.79	4.77	5.50	5.55	8.96
p-cymene	10.79	1022	7.51	5.67	4.60	4.99	8.05	4.85	3.84	8.82	5.87	11.15
Limonene	10.90	1026	7.80	3.70	3.26	4.10	4.94	3.01	3.28	4.43	3.17	5.10
(*Z*)- β – ocimene	11.22	1036	1.60	1.72	0.45	0.93	1.16	1.67	2.03	1.84	1.50	2.01
(*E*)- β –ocimene	11.53	1047	0.69	0.66	0.29	0.46	0.50	0.53	0.65	0.62	0.37	0.40
γ-terpinene	11.87	1058	18.08	8.07	8.74	8.47	12.05	8.90	8.95	12.40	10.40	14.26
Terpinolene	12.73	1085	4.62	1.99	2.08	1.99	2.54	1.30	1.81	1.88	1.92	3.15
Oxygenated monoterpenes												
1-octen-3-ol	9.41	977	0.20	0.53	0.00	0.31	0.33	0.26	0.42	0.00	0.37	0.35
(*Z*)-sabinene hydrate	12.09	1064	0.50	1.91	2.27	1.68	1.84	2.15	1.86	2.39	2.91	2.33
(*E*)-sabinene hydrate	13.11	1097	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	6.96
Linalool	13.14	1100	1.53	17.35	19.10	16.50	15.64	20.67	16.62	13.56	21.82	1.73
terpinen-4-ol	15.47	1179	5.65	6.16	6.03	5.82	3.77	4.58	5.85	8.06	6.26	10.85
α –terpineol	15.84	1191	0.91	2.78	2.59	2.98	2.29	2.20	2.80	2.02	2.22	1.78
thymol methyl ether	17.09	1236	5.56	4.12	3.32	4.23	2.69	3.30	3.76	2.85	2.60	2.91
carvacrol methyl ether	17.34	1246	7.04	6.12	4.85	5.13	4.14	5.16	5.04	4.98	3.13	4.08
Thymoquinone	17.51	1251	0.00	0.37	0.40	0.37	0.44	0.57	0.66	0.82	0.66	0.74
Linalyl acetate	17.69	1259	1.86	8.93	8.41	9.01	5.62	9.14	8.16	5.94	6.51	2.17
Phenolic monoterpenes												
Thymol	18.72	1295	5.18	6.33	7.32	7.05	9.69	8.80	10.06	5.72	7.72	5.12
Carvacrol	18.96	1303	0.00	0.41	0.46	0.25	0.42	0.67	0.38	0.25	0.15	0.13
Sesquiterpenes hydrocarbons												
Caryophyllene	22.14	1422	6.37	5.55	6.86	5.78	3.75	5.25	6.05	5.52	4.53	4.31
Aromadendrene	22.61	1441	0.66	0.92	1.05	1.08	0.56	0.88	0.81	0.61	0.77	0.31
α-humulene	22.98	1455.16	0.57	0.68	0.91	0.78	0.53	0.74	0.90	0.67	0.64	0.54
Alloaromadendrene	23.15	1462.36	0.11	0.10	0.08	0.13	0.08	0.10	0.11	0.11	0.12	0.12
α –muroleno	24.06	1498.88	0.84	2.37	2.38	2.22	1.59	3.11	3.95	2.18	2.37	0.90
β –bisabolene	24.31	1510.3	0.17	0.15	0.11	0.14	0.14	0.18	0.15	0.12	0.11	0.58

The values (relative peak area percent) represent averages of three determinations.
LRI: Linear retention index relative; Tr: retention time (min) relative to standard mixture of n-alkanes.

Supplementary material 1A (continue): Compounds identified in the samples of Origanum

Compounds	Tr	LRI	% peak area									
			S7	S8	S9	S10	S11	S12	S13	S14	S15	S16
Monoterpenes hydrocarbons												
α-thujene	7.78	920	1.08	1.66	1.41	0.93	1.35	2.10	0.86	1.10	0.65	1.03
α-pinene	7.96	926	0.20	0.19	0.26	0.25	0.30	0.32	0.19	0.21	0.17	0.15
Sabinene	9.18	968	2.10	2.67	2.70	1.85	2.46	3.55	1.30	2.29	1.17	2.08
β-myrcene	9.76	989	1.03	1.62	1.20	1.35	1.19	1.44	1.04	1.35	0.80	1.08
α –phellandrene	10.12	1001	0.45	1.11	0.83	1.33	0.86	1.86	0.84	0.78	0.76	1.01
α –terpinene	10.52	1014	2.66	4.36	4.79	6.19	4.31	7.25	3.31	3.96	2.76	5.03
p-cymene	10.79	1022	1.39	2.67	2.83	4.44	2.91	9.02	2.06	7.52	3.08	7.35
Limonene	10.90	1026	1.55	2.95	2.54	3.63	2.69	5.31	1.98	2.98	1.74	3.57
(*Z*)- β – ocimene	11.22	1036	1.18	1.83	1.64	1.62	1.68	1.81	1.55	0.86	0.63	0.87
(*E*)- β –ocimene	11.53	1047	0.55	0.91	0.61	0.58	0.62	0.64	0.67	0.41	0.36	0.37
ɣ-terpinene	11.87	1058	5.18	7.39	9.04	11.01	8.24	11.76	6.01	8.53	5.37	9.17
Terpinolene	12.73	1085	1.04	1.84	1.69	2.18	1.61	2.84	1.44	1.51	1.28	2.11
Oxygenated monoterpenes												
1-octen-3-ol	9.41	977	0.37	0.36	0.31	0.24	0.36	0.51	0.26	0.26	0.18	0.18
(*Z*)-sabinene hydrate	12.09	1064	1.58	1.57	2.11	2.21	2.08	2.85	1.42	2.30	1.78	2.47
(*E*)-sabinene hydrate	13.11	1097	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Linalool	13.14	1100	20.43	14.93	20.33	14.68	20.13	13.97	10.70	19.81	11.37	14.31
terpinen-4-ol	15.47	1179	2.81	4.24	0.05	7.82	3.63	8.65	5.80	5.83	7.04	9.48
α –terpineol	15.84	1191	3.02	2.78	4.31	2.94	2.49	1.85	2.80	2.62	2.72	2.94
thymol methyl ether	17.09	1236	4.22	4.69	2.57	3.04	4.13	2.96	4.60	3.09	4.48	3.16
carvacrol methyl ether	17.34	1246	6.45	6.91	3.33	4.46	5.22	4.14	5.94	4.99	6.11	5.25
Thymoquinone	17.51	1251	0.46	0.49	4.90	0.76	0.55	0.68	1.14	0.69	1.83	0.80
Linalyl acetate	17.69	1259	11.75	9.55	0.47	8.44	9.20	3.37	10.93	9.97	6.21	6.20
Phenolic monoterpenes												
Thymol	18.72	1295	11.16	8.36	10.35	8.37	8.91	4.62	13.54	6.43	16.47	7.11
Carvacrol	18.96	1303	0.44	0.35	7.91	0.35	0.25	0.18	0.55	0.23	0.83	0.37
Sesquiterpenes hydrocarbons												
Caryophyllene	22.14	1422	8.52	7.94	0.26	4.60	6.78	4.14	8.07	6.85	7.42	8.55
Aromadendrene	22.61	1441	1.80	1.71	6.21	0.76	1.24	0.52	2.25	0.49	1.22	0.52
α-humulene	22.98	1455.16	1.32	1.17	1.13	0.68	1.03	0.52	1.59	0.86	1.48	1.16
Alloaromadendrene	23.15	1462.36	0.15	0.15	0.93	0.09	0.12	0.08	0.26	0.11	0.21	0.14
α –muroleno	24.06	14.98.88	6.35	4.71	0.11	2.79	4.98	1.14	7.34	2.85	6.63	1.99
β –bisabolene	24.31	1510.3	0.14	0.19	4.17	0.17	0.13	0.73	0.34	0.21	4.26	0.21

The values (relative peak area percent) represent averages of three determinations.
LRI: Linear retention index relative; Tr::retention time (min) relative to standard mixture of n-alkanes.

Supplementary material 1A (continue): Compounds identified in the samples of Origanum

Compounds	Tr	LRI	% peak area									
			S17	S18	S19	S20	S21	S22	S23	S24	D1	D2
Monoterpenes hydrocarbons												
α-thujene	7.78	920	1.87	0.91	1.52	0.95	0.95	0.75	1.03	1.44	1.89	1.05
α-pinene	7.96	926	0.32	0.22	0.36	0.32	0.32	0.26	0.22	0.24	0.38	0.24
Sabinene	9.18	968	3.01	1.80	2.27	2.21	2.21	1.29	2.01	2.50	3.70	2.37
β-myrcene	9.76	989	1.51	1.52	1.86	1.32	1.32	1.36	1.38	1.14	1.77	1.61
α –phellandrene	10.12	1001	1.68	0.62	2.04	0.92	0.92	1.27	0.77	1.59	1.58	1.12
α –terpinene	10.52	1014	6.90	3.78	6.21	4.55	4.55	5.76	4.13	6.81	7.01	4.85
p-cymene	10.79	1022	4.72	3.92	4.24	5.85	5.85	4.50	2.24	5.92	9.05	4.79
Limonene	10.9	1026	4.38	2.61	4.56	3.32	3.32	3.46	2.39	3.94	5.42	3.63
(*Z*)- β – ocimene	11.22	1036	1.75	2.19	1.85	1.26	1.26	1.57	1.73	1.29	2.09	1.09
(*E*)- β –ocimene	11.53	1047	0.76	0.70	0.81	0.46	0.46	0.68	0.63	0.52	0.39	0.44
ɣ-terpinene	11.87	1058	10.66	8.42	10.48	9.39	9.39	9.89	7.37	10.69	10.84	8.43
Terpinolene	12.73	1085	2.65	1.49	2.26	1.64	1.64	2.27	1.68	2.64	2.37	1.90
Oxygenated monoterpenes												
1-octen-3-ol	9.41	977	0.34	0.34	0.27	0.27	0.27	0.32	0.46	0.26	0.97	0.26
(*Z*)-sabinene hydrate	12.09	1064	2.51	2.53	2.05	2.68	2.68	2.24	1.99	2.20	2.27	1.70
(*E*)-sabinene hydrate	13.11	1097	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	16.01
Linalool	13.14	1100	15.27	18.32	10.91	20.17	20.17	12.75	18.45	14.97	14.07	1.23
terpinen-4-ol	15.47	1179	7.12	6.68	9.43	6.30	6.30	9.07	3.69	9.21	8.80	6.77
α –terpineol	15.84	1191	2.56	3.05	3.28	2.19	2.19	2.20	2.42	2.98	3.70	3.32
thymol methyl ether	17.09	1236	3.42	2.99	3.12	3.59	3.59	3.69	4.21	3.08	3.34	3.78
carvacrol methyl ether	17.34	1246	4.12	5.62	4.81	5.15	5.15	5.45	6.12	4.29	3.73	5.27
Thymoquinone	17.51	1251	0.56	0.87	0.69	0.49	0.49	1.03	0.56	0.46	0.16	0.31
Linalyl acetate	17.69	1259	6.98	9.81	7.27	9.07	9.07	8.87	10.11	6.89	2.65	9.05
Phenolic monoterpenes												
Thymol	18.72	1295	5.54	9.60	7.96	6.33	6.33	9.18	10.00	4.40	4.28	7.70
Carvacrol	18.96	1303	0.17	0.47	0.36	0.22	0.22	0.44	0.47	1.60	0.32	0.41
Sesquiterpenes hydrocarbons												
Caryophyllene	22.14	1422	5.43	4.71	5.04	5.90	5.90	5.22	7.66	5.23	4.87	6.63
Aromadendrene	22.61	1441	1.06	0.77	1.19	0.46	0.46	0.69	1.16	0.64	0.48	0.68
α-humulene	22.98	1455.16	0.79	0.72	0.70	0.79	0.79	0.69	1.12	0.70	0.55	0.95
Alloaromadendrene	23.15	1462.36	0.11	0.11	0.13	0.10	0.10	0.11	0.12	0.10	0.00	0.09
α –muroleno	24.06	14.98.88	2.25	3.96	1.91	2.75	2.75	2.72	5.21	1.71	1.35	3.20
β –bisabolene	24.31	1510.3	0.13	0.10	0.15	0.16	0.16	0.14	0.16	0.20	0.09	0.21

The values (relative peak area percent) represent averages of three determinations.
LRI: Linear retention index relative; Tr: retention time (min) relative to standard mixture of n-alkanes.

Supplementary material 1A (continue): Compounds identified in the samples of Origanum

Compounds	Tr	LRI	% peak area									
			O1	O2	O3	O4	P1	P2	P3	P4	P5	P6
Monoterpenes hydrocarbons												
α-thujene	7.78	920	2.41	2.29	1.23	1.24	1.54	1.83	0.76	1.57	1.49	1.27
α-pinene	7.96	926	0.34	0.39	0.17	0.20	0.30	0.37	0.22	0.23	0.31	0.28
Sabinene	9.18	968	3.82	4.27	2.14	2.72	3.57	3.98	1.72	2.74	2.83	2.56
β-myrcene	9.76	989	1.31	1.43	1.48	1.54	1.55	1.78	0.81	1.43	1.29	1.26
α –phellandrene	10.12	1001	1.59	1.28	2.23	1.74	1.19	1.19	0.57	1.57	0.87	1.12
α –terpinene	10.52	1014	6.95	7.39	7.04	6.84	6.38	5.80	3.57	6.43	4.75	4.39
p-cymene	10.79	1022	11.15	11.29	5.03	4.96	9.27	7.51	1.81	3.86	10.12	4.93
Limonene	10.9	1026	5.02	5.02	4.83	4.59	5.24	4.82	1.97	4.01	4.08	2.98
(*Z*)- β – ocimene	11.22	1036	0.92	0.68	1.36	1.31	2.41	2.45	1.11	1.64	0.85	0.36
(*E*)- β –ocimene	11.53	1047	0.38	0.26	0.50	0.43	0.57	0.49	0.26	0.55	0.31	0.28
ɣ-terpinene	11.87	1058	10.27	11.01	9.80	10.19	11.03	10.19	5.31	9.50	8.06	6.54
Terpinolene	12.73	1085	2.88	2.68	2.67	2.59	2.50	1.96	1.19	2.56	2.05	1.89
Oxygenated monoterpenes												
1-octen-3-ol	9.41	977	0.37	0.37	0.57	0.31	0.65	0.79	0.36	0.56	0.48	0.33
(*Z*)-sabinene hydrate	12.09	1064	2.28	2.62	1.33	2.01	3.53	2.33	1.36	1.99	1.98	1.86
(*E*)-sabinene hydrate	13.11	1097	13.00	17.36	9.90	13.88	0.00	0.00	0.00	0.00	0.00	0.00
Linalool	13.14	1100	0.74	0.63	1.33	1.65	19.24	18.08	10.13	14.01	13.25	15.58
terpinen-4-ol	15.47	1179	2.63	2.13	9.81	3.36	6.48	6.09	2.77	7.56	9.84	9.03
α –terpineol	15.84	1191	2.90	2.42	3.37	2.94	0.13	2.65	1.15	2.94	3.40	-
thymol methyl ether	17.09	1236	5.52	4.81	3.60	5.36	3.06	3.71	1.82	3.67	3.55	2.96
carvacrol methyl ether	17.34	1246	0.20	0.16	5.58	0.19	4.70	5.58	2.35	5.89	5.95	5.31
Thymoquinone	17.51	1251	4.85	4.99	0.25	6.01	0.57	0.33	0.09	0.30	0.41	0.54
Linalyl acetate	17.69	1259	4.59	3.64	5.99	6.00	3.56	4.05	3.31	7.46	5.88	7.59
Phenolic monoterpenes												
Thymol	18.72	1295	0.28	0.18	7.46	0.34	4.28	5.10	2.05	6.01	8.26	11.52
Carvacrol	18.96	1303	4.62	4.21	0.35	5.83	0.20	0.24	0.07	0.29	0.56	1.11
Sesquiterpenes hydrocarbons												
Caryophyllene	22.14	1422	0.43	0.29	5.79	0.53	4.30	3.06	2.75	6.45	4.99	5.97
Aromadendrene	22.61	1441	0.58	0.52	0.92	0.77	0.37	0.60	0.28	1.11	0.53	0.77
α-humulene	22.98	1455.16	0.19	0.14	0.77	0.09	0.52	0.76	0.33	0.89	0.62	0.48
Alloaromadendrene	23.15	1462.36	0.84	0.84	0.10	1.78	0.07	0.20	1.02	0.12	0.24	0.22
α –muroleno	24.06	14.98.88	0.10	0.08	1.99	0.13	1.45	1.92	0.04	2.89	1.54	2.96
β –bisabolene	24.31	1510.3	0.00	0.00	0.17	0.00	0.44	0.66	50.00	0.14	0.20	0.16

The values (relative peak area percent) represent averages of three determinations.
LRI: Linear retention index relative; Tr: retention time (min) relative to standard mixture of n-alkanes.