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Research Article

Leaf and Flower Volatile Oil Components of Two Thyme Taxa *Origanum onites* L. and *Thymbra spicata* var. *spicata* L. in Turkey

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Abstract

Medicinal and aromatic plants have a special importance with volatile oil components. Lamiaceae family members are important in pharmacology and perfumery industry because they contain volatile and aromatic oil. *Origanum onites* L. and *Thymbra spicata* var. *spicata* L. are the most widely used and most exported species. The volatile components of the leaves and flowers of *Origanum onites* L. and *Thymbra spicata* var. *spicata* taxa were determined by Headspace Solid Phase Microextraction (HS-SPME) technique combined with gas chromatography/mass spectrometry (GC / MS). 33 different components of *Origanum onites* were identified and the main components were p-cymene (11.45%), γ-terpinene (11.89%), linalool (14.35%), thymol (20.03%) and carvacrol (26.91%), respectively. For *Thymbra spicata* var. *spicata* L., 36 different compounds were identified and the main components were p-cymene (11.72%), γ-terpinene (10.96%), linalool (13.44%), thymol (18.92%) and carvacrol (27.34%), respectively. Oxygen containing monoterpenes have been found to be high.

Keywords: *Origanum onites* L., *Thymbra spicata* var. *spicata* L., SPME, carvacrol, thymol, Turkey

İki Kekik Taksonu *Origanum onites* L. ve *Thymbra spicata* var. *spicata* L.'nın Yaprak ve Çiçek Uçucu Bileşenleri

Öz

Tıbbi ve aromatik bitkiler grubunda özellikle uçucu yağ içeriği açısından zengin olanların ayrı bir önemi bulunmaktadır. *Lamiaceae* familyası üyeleri uçucu ve aromatik yağ içermelerinden dolayı farmakoloji ve parfümeri sanayinde önemlidir. *Lamiaceae* familyasının ihracatı en çok yapılan ve uçucu yağ üretiminde kullanılan türleri ise; *Origanum onites* L. ve *Thymbra spicata* var. *spicata* L. dır. *Origanum onites* L. ve *Thymbra spicata* var. *spicata* L. taksonlarının yaprak ve çiçeklerinin uçucu bileşenleri gaz kromatografisi/kütle spektrometresi (GC/MS) ile kombine edilmiş Tepe Boşluğu-Katı Faz Mikro Ekstraksiyon (HS-SPME) tekniği ile belirlenmiştir. *Origanum onites*'te 33 farklı bileşen tespit edilmiş olup ana bileşenler p-cymene (%11.45), γ-terpinene (%11.89), linalool (%14.35), thymol (%20.03) ve carvacrol (%26,91)'dur. *Thymbra spicata* var. *spicata* L.'da 36 farklı bileşen tespit edilmiş olup ana bileşenler p-cymene (%11.72), γ-terpinene (%10.96), linalool (%13.44), thymol (%18.92) ve carvacrol (%27.34)'dur. Oksijenli monoterpenleri yüksek oranda bulunduğu belirlenmiştir.

Anahtar Kelimeler: *Origanum onites* L., *Thymbra spicata* var. *spicata* L., SPME, carvacrol, thymol, Türkiye

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Introduction

Volatile oils (essences, ethereal oils) and their aromatic extracts are widely used as a source of fragrance, food additives, cleaning products, in fragrance of cosmetics and medicines and also in taste industries, as a source of aroma-chemicals or as a starting material for synthesis of naturally identical and semi-synthetic beneficial aroma chemicals (Weiss, 1997). The Lamiaceae family members which are represented by 45 genera and 546 species in Turkey, are important in pharmacology and perfumery industry due to comprising volatile and aromatic oils (Secmen et al., 2000).

More than 15 plant species in Turkey are named and used as "thyme". The majority of these plants belong to *Thymus* of the Lamiaceae family, while others include the genus *Origanum*, *Satureja*, *Majorana* and *Thymbra* (Ozguven and Tansi, 1998; Kocabas and Karaman, 2001).

A well-known folk medicine and spice plant, thyme is mainly used for meat dishes, vegetable dishes, various sauces and salads, cheeses and sausage production, digestive system and upper respiratory tract disorders, indigestion, loss of appetite and cough. It has antiseptic, sedative, gas expectorant, expectorant, cramp solvent properties. Also, it is a highly sought-after spice plant in food storage in recent years due to its antibacterial effects on bacteria causing food spoilage and food poisoning (Bayram et al., 2010). Thyme is a good source of pollen for bees and a source of quality grass in milk-giving animals (Ortiz and Fernandez, 1992).

SPME is a method for combining sample preparation, extraction and condensation in a single step without solvent. Significant gains were obtained in the processing time and costs by this method, while the diagnosis was improved. In addition, it has been observed that there are positive developments in the sample preparation stage and the results. The effectiveness of the SPME method depends on the type and thickness of the material covering the fiber portion in the syringe. Besides, the fact that SPME method is as short as 1-30 minutes shows its advantage over other methods (Vas and Vekey, 2004; Araujo et al., 2007; Donmez and Salman, 2017).

The most widely exported and used in the production of essential oil are *Origanum onites* and *Thymbra spicata* var. *spicata*. *Origanum onites* and *Thymbra spicata* var. *spicata* has been the subject of more researches, mainly due to its wide range and volatile fat content. In other studies, it is seen that different methods are used to determine the essential oil components. The aim of this study was to determine the volatile components, quantities and classes of fresh leaves and flowers of *Origanum onites* and *Thymbra spicata* var. *spicata* that were collected from Sutlegen province of Kas county in Antalya, Turkey.

Materials and Methods

Origanum onites and *Thymbra spicata* var. *spicata* specimens that were collected from stands where these are grown naturally in Sutlegen province of Kas county in Antalya, Turkey during the vegetation period of 2017-2018, constitute the material of the study. Collecting site is situated in the southwestern of Turkey (36° 28' N; 29° 38 E'). The leaves and flowers of the plant samples collected from the research area were placed in paper packaging and transferred to the laboratory on the same day without ever being exposed to sunlight. The collected plant materials were dried at room temperature (25 °C). The volatile components of leaves and flowers were determined by the Head Space Solid Phase Micro Extraction (HS-SPME) technique combined with gas chromatography / mass spectrometry (GC/MS). On the basis of the solid phase micro extraction technique, 2 g of the leaves and flower samples taken from each sample were placed in a 10 mL vial and stored at 60 °C for 30 minutes after the mouth was sealed with a silicone cap.

The SPME apparatus was passed through the headspace with 75 μm thin Carboxen/ Polidimethylsiloxane (CAR / PDMS) coated fused slica fiber to adsorb volatiles then injected directly into the capillary column of the Shimadzu 2010 Plus GC-MS device (Restek Rx-5, MS 30 m x 0.25 mm, 0.25 μm). The device is connected to the same brand mass selector detector operated in EI mode (70 eV). This procedure was repeated three times to compare the accuracy of the results and the results were given as average. Helium was used as carrier gas with a flow rate of 1.61 mL per minute. Injection and detection temperatures were set at 250°C. Retention Indices (RI) of volatile components are calculated according to the standard of C7-C30 alkane mixtures under the above chromatographic conditions. Identification of the compounds was performed by comparing the mass spectra and the compounds in the spectral library (Wiley, Nist, Tutor, FFNSC).

Results and Discussion

Volatile components, amounts and classes of leaves and flowers of *Origanum onites* L. and *Thymbra spicata* var. *spicata* were determined according to SPME (solid-based micro extraction) technique. 33 different components of *Origanum onites* were determined. Of them, p-cymene (11,45%), γ-terpinene (11,89%), linalool (14,35%), thymol (20,03%) and carvacrol (26,91%)' were found as main components. Also, 36 components were determined for *Thymbra spicata* var. *spicata*. p-cymene (11,72%), γ-terpinene

(10,96%), linalool (13,44%), thymol (18,92%) and carvacrol (27,34%) were main components. Oxygen containing monoterpenes were found to be high (Table 1).

Table 1. SPME results for *Origanum onites* L. and *Thymbra spicata* var. *spicata* L.

No	RI	RT	Constituents	O. onites %	T. spicata %	Formula	Category
1.	<700	1.670	2-Methylpropenal	-	0,17	C_4H_8O	AA
2.	<700	1.873	Acetic acid	0,25	0,17	$C_2H_4O_2$	FA
3.	<700	2.178	2-Butenal	0,08	0,08	C_4H_6O	AAI
4.	<700	2.220	3-Methylbutanal	-	0,11	$C_5H_{10}O$	AA
5.	<700	2.311	2-Methylbutanal	-	0,12	$C_5H_{10}O$	AA
6.	<700	2.531	1-Penten-3-one	-	0,13	C_5H_8O	AAI
7.	<700	2.680	Pentanal	0,06	-	$C_5H_{10}O$	AAI
8.	703	2.695	Furan	-	0,17	C_6H_8O	AA
9.	751	3.606	(E)-2-Pentenal	-	0,07	C_5H_8O	AAI
10.	801	4.596	Hexanal	0,07	0,08	$C_6H_{12}O$	AA
11.	850	6.085	(E)-2-Hexenal	0,13	0,83	$C_6H_{10}O$	AA
12.	927	8.495	α-Thujene	0,79	1,16	$C_{10}H_{16}$	MH
13.	933	8.734	α-Pinene	0,94	0,82	$C_{10}H_{16}$	MH
14.	957	9.365	Camphene	0,40	0,11	$C_{10}H_{16}$	MH
15.	964	9.788	Benzaldehyde	0,06	0,08	C_7H_6O	AAI
16.	978	9.535	β_Pinene	0,18	0,18	$C_{10}H_{16}$	MH
17.	979	9.805	3-Octenol	1,29	1,09	$C_8H_{16}O$	AAI
18.	986	10.445	Vinyl amyl ketone	-	0,15	$C_8H_{14}O$	AAI
19.	991	10.943	β-Myrcene	2,33	2,51	$C_{10}H_{16}$	MH
20.	1007	10.647	α-Phellandrene	0,38	0,39	$C_{10}H_{16}$	MH
21.	1009	10.742	.δ.3-Carene	0,14	0,10	$C_{10}H_{16}$	MH
22.	1013	10.896	2,4-Heptadienal	0,59	0,49	$C_7H_{10}O$	AA
23.	1018	11.102	α-Terpinene	1,84	1,57	$C_{10}H_{16}$	MH
24.	1025	12.255	**p-Cymene**	11,45	11,72	$C_{10}H_{14}$	MH
25.	1030	12.414	Limonene	1,37	1,32	$C_{10}H_{16}$	MH
26.	1031	12.632	1,8-Cineole	0,08	0,09	$C_{10}H_{18}O$	OM
27.	1046	13.163	β-Ocimene	0,37	1,34	$C_{10}H_{16}$	MH
28.	1058	13.544	**γ-Terpinene**	11,89	10,96	$C_{10}H_{16}$	MH
29.	1067	14.020	trans-Sabinene hydrate	0,16	0,53	$C_{12}H_{20}O_2$	FA
30.	1101	15.265	**Linalool**	14,35	13,44	$C_{10}H_{18}O$	OM
31.	1180	18.290	Terpineolene	0,41	0,44	$C_{10}H_{16}$	MH
32.	1198	18.852	α-Terpineol	0,36	0,20	$C_9H_{18}O$	AAI
33.	1300	22.455	**Thymol**	20,03	18,92	$C_{10}H_{14}O$	OM
34.	1317	22.715	**Carvacrol**	26,91	27,34	$C_{10}H_{14}O$	OM
35.	1418	26.905	Caryophyllene	2,32	2,38	$C_{15}H_{24}$	SH
36.	1454	26.966	.α.-Humulene	0,17	0,12	$C_{15}H_{24}$	SH
37.	1458	27.453	Aromadendrene	0,21	-	$C_{15}H_{24}$	SH
38.	1508	29.716	β-Bisabolene	0,12	0,62	$C_{15}H_{24}$	SH
39.	1557	29.735	Germacrene B	0,11	-	$C_{15}H_{24}$	SH
40.	1584	32.035	Caryophyllene oxide	0,16	-	$C_{15}H_{24}O$	OS
TOTAL				**100**	**100**		
Component				33	36		
AA: Aromatic alcohol				0,79	1,97		
AAI: Aromatic aldehyde				1,85	1,80		
FA: Fatty acid methyl ester				0,41	0,70		
MH: Monoterpene hydrocarbon				32,49	32,62		
OS: Oxygenated seskiterpen				0,16	-		

No	RI	RT	Constituents	O. onites %	T. spicata %	Formula	Category
OM: Oxygenated monoterpene				61,37	59,79		
SH: Sesquiterpene hydrocarbon				2,93	3,12		

***RI:** Retention index, ***RT:** Retention time.

In previous studies, Erdemgil (1992) determined carvacrol as main essential oil component of *Origanum onites*. Also, carvacrol, γ-terpinene and β- bisabolene were found as main components by Ruberto et al. (1993). Results of other studies that were conducted the essential oil components of *Origanum onites*, are in parallel with our study. Unlike other studies, the main components were p-cymene, linalool and thymol in our study.

Kilic (2006) were determined carvacrol, p-cymene, β-myrcene, γ-terpinene, α-terpinene and trans-caryophyllene as main essential oil components of *Thymbra spicata* var. *spicata* by hydrodistillation and GC / MS analysis. Tumen et al. (2011) found carvacrol as main component of *T. spicata* var. *spicata*. Carvacrol and thymol were found as main components in study of Al-Sheibany et al. (2005). Also, carvacrol, thymol, γ-terpinene and p-cymene were determined by Ravid and Putievsky (1985), carvacrol and γ-terpinene were found by Markovic et al. (2011) as main components of *Thymbra spicata*. In the study of Akgul et al. (1999), carvacrol, thymol, γ-terpinene and p-cymene were found as main components. Carvacrol, thymol, γ-terpinene and p-cymene were found by Fleisher and Fleisher (2005) and also carvacrol, α-thujene, myrcene, γ-terpinene and p-cymene were determined as main component of *T. spicata* by Barakat et al. (2013). All these results support our research. By the way, linalool was determined as different.

Conclusions

According to the SPME (solid-based micro extraction) technique, 33 components were determined for *Origanum onites* and also 36 for *Thymbra spicata* var. *spicata.* In both taxa, p-cymene, γ-terpinene, linalool, thymol and carvacrol were found to be main components. The results of the study were compared with leaf and flower samples obtained from different regions and analyzed by hydrodistillation method. They are collected from the nature in an inappropriate and dense manner due to the most widely used and used in the production of essential oil. This situation jeopardizes the extinction of species and narrows its natural distribution areas. Local people and traders should be made aware of the conscious collection and consumption. In addition to the therapeutic properties of the essential oils, it is necessary to take into consideration the harmful aspects and the place and dosage of the essential oils should be well adjusted. Thyme is also consumed as a well-known folk medicine, tea and spice plant. It is recommended that people with sugar and blood pressure should pay attention to their dose while drinking their tea. Detailed studies should be carried out in these areas in order to be used more extensively in the food, cosmetic and pharmaceutical industries.

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