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Chemical Composition of *Satureja boissieri* Hausskn. ex Boiss. Species From Adıyaman (Turkey)

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ABSTRACT: The genus *Satureja* L. (Lamiaceae) includes about 200 species of herbs and shrubs, often aromatic, widely distributed in the Mediterranean area, Asia and boreal America (Rustaiyan et al*.,* 2004). *Satureja* is represented by sixteen taxa in Turkey (Davis, 1982; Tumen, 2000) and the endemism ratio is 33% in Turkey, where *Satureja* species are grown mainly in south and west Anatolia (Davis, 1982). The essential oil components of aerial parts of *Satureja boissieri* Hausskn. ex Boiss. was investigated by GC and GC-MS. The yield of plant is ca. 0.20 g L^{-1}. Thirty five component were identified representing 95.4% oil. Carvacrol (30.1%), thymol (21.8%), *p*-cymene (12.5%) and γ-terpinene (6.5%) were found as main constituents. The results were discussed in terms of natural products, renewable resources and chemotaxonomy.

Keywords: *Satureja*, essential oil, carvacrol, thymol, chemotaxonomy

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Adıyaman'dan (Türkiye) *Satureja boissieri* Hausskn. ex Boiss. Türünün Kimyasal Kompozisyonu

ÖZET: *Satureja* L. (Lamiaceae) cinsi yaklaşık 200 tür içerip daha çok Akdeniz, Asya ve Boreal Amerika'da yayılış gösteren otsu, çalımsı ve çoğunlukla aromatik bitkilerdir (Rustaiyan ve ark*.,* 2004). *Satureja* cinsinin ülkemizdeki endemizm oranı yaklaşık %33 civarındadır ve 16 takson ile temsil edilip, daha çok Anadolu'nun güney ve batı kesimlerinde yayılış gösterir (Davis, 1982). *Satureja boissieri* Hausskn. ex Boiss.'in topraküstü kısımlarının uçucu yağ içerikleri GC ve GC-MS ile araştırıldı. Bitkinin yağ verimi yaklaşık olarak 100 gramda 0.2 mL'dir. Toplam yağdan (% 95.4) otuz beş bileşen tespit edildi. Karvakrol (% 30.1), timol (% 21.8) ve *p*-simen (% 12.5) ve γ-terpinen (% 6.5) ana bileşenler olarak bulundu. Sonuçlar doğal ürünler, yenilenebilir kaynaklar ve kemotaksonomi açısından tartışıldı.

Anahtar Kelimeler: *Satureja*, uçucu yağ, karvakrol, timol, kemotaksonomi

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INTRODUCTION

Turkey is regarded as an important gene-centre for the family Lamiaceae. Lamiaceae is represented in Turkey by 45 genus, 574 species and the endemism ratio of the family is about 44% (Erik and Tarikahya, 2004). Most aromatic plants belonging to the family Lamiaceae, such as *Satureja*, *Origanum*, *Thymus*, etc., are used as herbal tea in Turkey (Baser, 1995). There are a number studies on chemical composition of Lamiaceae genus (Kilic et al*.,* 2011; Kilic and Bagci, 2008). *Satureja* L. is a genus of aromatic plants of the family Lamiaceae (the mint family). The genus has been a subject of much discussion among taxonomists and is variously treated. In Flora Europaea (Heywood and Richardson, 1972) recognized 5 genera in the region including *Calamintha* R.R. Mill*., Acinos* R.R. Mill*, Clinopodium* L., *Micromeria* Benth., and *Satureja* L. In the Flora of USSR (Shishkin, 1954), China (Li and Hedge, 1994), Turkey (Davis, 1982) and Iranica (Rechinger, 1982) a similar classification was adopted considering to some specific genera which were included on the basis of geographical distribution. *Satureja* in its narrow concept is a genus comprising 30 species, mainly distributed in Mediterranean Region but also extended to Irano-Turanian phytogeographical region. *Satureja* belongs to the tribe Mentheae within the subfamily Nepetoideae and includes about 284 species in the world (Nixon, 2006). Certain organic chemicals are derived from *Satureja* taxa, which are useful to humans. They have usually well known, and will be used by native inhabitants as spice, medicinal plant or source of essential oils. Medicinal properties and a large variety of specimens in the species increase the importance of diversity studies in this genus. Within the *Satureja* genus the genetic diversity has been dealt with using morphological characters (Hadian et al., 2011; Kasyani et al., 2012), enzyme electrophoresis (Attar et al., 2006; Arzi et al., 2010), chloroplast DNA restriction site analysis (Cantino, 1998) and molecular markers (Hadian et al., 2008; Hadian et al., 2010).

Satureja boissieri, one of these species, is erect, suffrutescent perennial plant. Stems virgate, (25-) 40-60 cm, ± simple, pubescent to recurved-pubescent. Leaves lineare-oblanceolate, 10-26 x 2-5 mm, obtuse, emucronate, scabridulous. Inflorescence linear-oblong, 8-20x1.5-3 cm. Verticillasters approximate, (2-) 6-17-flowered, axillary cymes ± lax, usually conspicuously pedunculate. Floral leaves as long to much shorter than verticllasters. Calyx distincly bilabiate to 1/3-1/2, tubular-campanulate, 4-6 mm, crisply pubescent; lower 2 teeth 1.5-2 mm, upper teeth 0.75-1 mm. Corolla white, c. 9 mm, tube exerted from calyx. Stamens subexerted from corolla. Fl. 7-8. It grows rocky volcanic slopes, *Quercus* scrub, 1250-1480 m. (Davis, 1982).

Satureja taxa are well known medicinal plants due to presence of secondary metabolites such as flavonoids, steroids, terpenoids and tannins they are known for their healing properties for a long time and have been used as traditional folk remedies to treat various ailments such as cramps, diarrhoea, infectious diseases, muscle pains and nausea indigestion (Bezic et al., 2009). The essential oil constituents show different ingredients in other genera i.e. *Acinos* with pulegone, menthone and germacrene D as the prominent constituents of their oils (Chalchat et al., 2004; Stojanovic et al., 2009), *Calamintha* with piperitone oxide and cis-piperitone oxide (Kitic et al., 2001); *Clinopodium* with cis-piperitone oxide, piperitone oxide, pulegone and isopulegone (Castilho et al., 2006). However terpenoids can be recognized as potential taxonomic markers at generic levels. More investigation on essential oil pattern in other genera within the *Satureja* complex will provide a better understanding of their phylogenetic relationships.

Satureja species have economic and medicinal importance because of their high essential oil content. With their pleasant fragrance, *Satureja* species are widely used as herbal teas and spices in Turkey. Members of this genus are called *kekik* in Turkish, and some species are exported as thyme (Satil et al*.,* 2002). In folk medicine, especially *S. cuneifolia* Ten*., S. thymbra* L. and *S. hortensis* L. are traditionally used as digestives and diuretics in various regions of Turkey (Baytop, 1999). The aerial parts of *Satureja* species have distinctive tastes and can be added to stuffing, meat, chicken, pies and sausages as a seasoning. *S. cuneifolia* is used to produce essential oil and aromatic water. This plant is also used as a condiment and herbal tea, owing to its stimulating, tonic and carminative effect (Eminagaoglu et al*.,* 2007). Since ancient times, herbs and spices have been added to different types of food to improve the flavour and organoleptic properties. Especially popular today is the concept of food that combines nutritional and medicinal benefits. Many natural compounds isolated from plants have demonstrated a wide spectrum of biological activities. Among these, various kind of natural substances, essential oils from

aromatic and medicinal plants receive particular attention as potential natural agents for food preservation. Moreover, essential oils are proven to have various pharmacological effects, such as spasmolytic, carminative, hepatoprotective, antiviral and anticarcinogenic (Bowles, 2004). The therapeutic properties of these species can mainly be attributed to their essential oil that includes different amounts of biologically active compounds such as carvacrol and thymol. The essential oils isolated from various species of *Satureja* have been shown to have high chemical polymorphism and various biological activities such as antibacterial and antifungal (Eftekhar et al*.,* 2009), antioxidant, analgesic and anti-inflammatory (Ghazanfari et al*.,* 2006), antispasmodic, antidiarrhoea, antidiabetic, antihyperlipidemic and reproduction-stimulatory activities (Abdollahi et al*.,* 2003), and improvement of fertility (Haeri et al*.,* 2006), treatment of recurrent aphthous stomatitis and protection against organophosphorus compounds (Basiri et al*.,* 2007). Therefore, there is a great interest in continuing researches on the essential oil of these plants from points of view of the chemical composition to biological properties. Literature review showed variation between chemical compositions of different *Satureja* species oils. The main components of *S. boissieri* (Kurkcuoglu et al*.,* 2001) oil from Turkey were carvacrol (40.8%) and γ-terpinene (26.4%). The main constituents of *S. brownei* (Rojas and Usubillaga, 2000) oil from Venezuela were pulegone (64.3%) and menthone (20.2%). The main compound of *S. parvifolia* (Viturro et al*.,* 2000) oil from Argentina was piperitone oxide and those of *S. boliviana* (Rojas and Usubillaga, 2000) oil were γ-terpinene, β-caryophyllene and germacrene D. Germacrene D has also been detected as the main compound of *S. coerulea* (Tumen et al*.,* 1998) oil from Turkey. The main components of *S. hortensis* (Baher et al*.,* 2002), cultivated in Iran, were carvacrol and γ-terpinene.

The aim of this study is to examine the chemical composition of the essential oils isolated from aerial parts of *S. boissieri* by GC-MS; and to evaluate the chemical data that might be helpful in potential usefulness and chemotaxonomy of this plant.

Plant material:

S. boissieri (Kilic-3210) was collected from in an island which behind the Atatürk dam wall, from Adıyaman / Turkey, on June 2011 at an altidude of 1100-1200 m.

Isolation of the essential oil:

Air-dried aerial parts of the plant material (100 g) were subjected to hydrodistillation using a Clevenger-type apparatus for 3 h.

Gas chromatographic (GC) analysis:

The essential oil was analyzed using HP 6890 GC equipped with and FID detector and an HP- 5 MS column (30 m × 0.25 mm i.d., film tickness 0.25 μm) capillary column was used. The column and analysis conditions were the same as in GC-MS. The percentage composition of the essential oils was computed from GC-FID peak areas without correction factors.

Gas chromatography / mass spectrometry (GC-MS):

The oils were analyzed by GC-MS, using a Hewlett Packard system. HP-Agilent 5973 N GC-MS system with 6890 GC in Plant Products and Biotechnology Research Laboratory (BUBAL) in Firat University. HP-5 MS column (30 m × 0.25 mm i.d., film tickness (0.25 μm) was used with helium as the carrier gas. Injector temperature was 250 ºC, split flow was 1 mL/min. The GC oven temperature was kept at 70 ºC for 2 min. and programmed to 150 ºC at a rate of 10 ºC/min and then kept constant at 150 ºC for 15 min to 240 ºC at a rate of 5 ºC / min. Alkanes were used as reference points in the calculation of relative retention indices (RRI). MS were taken at 70 eV and a mass range of 35-425. Component identification was carried out using spectrometric electronic libraries (WILEY, NIST). The identified compounds of the essential oils are listed in Table-1.

RESULTS AND DISCUSSION

The essential oil components of aerial parts of *Satureja boissieri* was investigated by GC and GC-MS. The yield of oil is ca. 0.20 mL / 100 g. Thirty five component were identified representing 95.4% oil. Carvacrol (30.1%), thymol (21.8%), *p*-cymene (12.5%) and γ-terpinene (6.5%) were identified as main constituents (Table 1). Carvacrol also reported main constituents of *S. bachtiarica* Bunge. (13.2%), *S. mutica* Fisch & C.A.Mey. (68.8%), *S. khuzistanica* Jamzad. (29.4%), *S. rechingeri* Jamzad. (77%), (Hadian et al*.,* 2012), *S. cuneifolia* Ten. (44.9%) (Oke et al*.,* 2009) and *S. boissieri* (40.8%) (Kurkcuoglu et al*.,* 2001). The major monoterpenes of *S. boissieri* were thymol (21.8%) and carvac-

rol (30.1%). Thymol also reported main constituents of *S. bachtiarica* (28%), *S. mutica* (29.8%) (Hadian et al*.,* 2012); *S. atropatana* Bunge. (62.1%)*, S. mutica* (62.6%) (Gohari et al*.,* 2005) and *S. mutica* (26.5%), *S. macrantha* C.A.Mey. (8.1%), *S. intermedia* C.A.Mey. (32.3%) (Sefidkon et al*.,* 2005). These oils are characterized by the presence of thymol, carvacrol, *p*-cymene and γ-terpinene.

The major compound of *S. montana* L. oil was the phenolic monoterpene carvacrol (45.7%). Other important compounds were the monoterpenic hydrocarbons *p*-cymene (12.6%), γ-terpinene (8.1%) and the oxygen-containing compounds carvacrol methyl ether, borneol, thymol and thymol methyl ether. The volatile oil of *S. cuneifolia* was characterized as β-cubebene (8.7%), limonene (8.3%), α-pinene (6.9%), spathulenol and β-caryophyllene (Skocibusic and Bezic, 2004). In our study carvacrol (30.1%), thymol (21.8%), *p*-cymene (12.5%) and γ-terpinene (6.5%) were identified as main constituents (Table-1).

Table I. Chemical profiles of *Satureja boissieri* (%).

No	Compounds	*RRI	*S. boissieri*
1	α-thujene	1016	0.5
2	α-pinene	1023	1.7
3	Camphene	1034	0.2
4	Sabinene	1052	0.1
5	β-pinene	1056	1.6
6	Mrycene	1063	1.2
7	α-phellandrene	1077	0.1
8	*p*-cymene	1093	12.5
9	Limonene	1097	1.9
10	1,8-Cineole	1095	3.1
11	β-ocimene	1100	0.1
12	γ-terpinene	1115	6.5
13	Terpinolene	1138	0.3
14	Linalool	1145	0.1
15	Trans-pinocarveol	1178	0.1
16	Camphor	1184	0.2
17	Borneol	1200	1.8
18	α-terpineol	1215	0.1
19	Trans-carveol	1231	0.5
20	Bornyl acetate	1282	1.0
21	Thymol	1297	21.8
22	Carvacrol	1300	30.1
23	β-bourbenene	1365	0.3
24	β-Caryophyllene	1393	2.3
25	β-farnesene	1415	0.4
26	α-humulene	1418	0.2
27	Aromadendrene	1421	0.3
28	*n*-decanal	1425	0.1
29	Germacrene D	1432	1.8
30	β-selinene	1441	0.2
31	Bicyclogermacrene	1443	0.4
32	α-cadinol	1458	0.2
33	Spathulenol	1495	2.3
34	Caryophyllene oxide	1498	1.1
35	α-copaene	1534	0.3
		Total	**95.4**

*RRI: Relative Retention Index.

This study showed that the genus *Satureja* had a considerable variation in essential oil composition and this study demonstrates the occurrence of the carvacrol (30.1%), thymol (21.8%), *p*-cymene (12.5%) and γ-terpinene (6.5%) chemotype of *S. boissieri* in eastern Anatolian region of Turkey (Table-1). Some of the *Satureja* species showed different chemotype of essential oil, like carvacrol (40.8%), γ-terpinene (26.4%), and *p*-cymene (14.5%) chemotype in *S. boissieri* from Turkey (Kurkcuoglu et al*.,* 2001) and carvacrol (53.5% and 52-56%) chemotype in *S. pilosa* and *S. icarica* from Turkey (Tumen et al*.,* 1998). Chemical analysis has shown that the essential oil of *Satureja* taxa has chemical variatons and different oil profiles. These differences might be originated from the genetic, local, climatic and seasonal factors (Perry et al*.,* 1999). The findings showed that the genus *Satureja* had a considerable variation in essential oil composition and this study demonstrates the occurrence of the carvacrol / thymol chemotype in the Eastern Anatolian region of Turkey. Moreover this result is significant to chemotaxonomic evaluation of the genus and family patterns.

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