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Running Head: Composition of the essential oil of three *Stachys* species from Turkey

Composition of the essential oil of *Stachys sericantha, S. gaziantepensis* and *S. mardinensis* (Lamiaceae) from Turkey

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ABSTRACT

The genus *Stachys* L. one of the largest genera of Lamiaceae and *Stachys* species are used by many people for the therapeutic value of their essential oils. In this study, water-distilled essential oil of three species of *Stachys* were analyzed. The analysis was performed by using a gas chromatography (GC-FID) and gas chromatography-mass spectrometry (GC-MS) systems, simultaneously. The major components were found as hexadecanoic acid (23.7%), dodecanoic

acid (11.3%) and caryophyllene oxide (10.7%) for *S. sericantha* P.H. Davis (endemic), α-pinene (53%), β-pinene (8.2%) for *S. gaziantepensis* M. Dinc and S. Doğu (endemic), Menthyl acetate (15.3%), isomenthone (15.0%), pulegone (10%), spathulenol (7.0%) and caryophyllene oxide (6.7%) for *S. mardinensis* (Post) R. Mill .

***Keywords*:** Essential oil, GC-FID, GC-MS, *Stachys sericantha*, *Stachys gaziantepensis*, *Stachys mardinensis*

INTRODUCTION

The genus *Stachys* L. one of the largest genera of the subfamily Lamioideae (Lamiaceae), includes about 300 species of annuals and perennials occurs in all parts of the world, except for Australia and New Zealand.[1] The *Stachys* genus is represented 87 species (112 taxa) belonging to 15 sections and 2 subgenera in Turkey. 52 species are endemic to Turkey and endemism ratio is 46%.[2-5] Most of the endemic taxa are east Mediterranean elements. The species name originates from the Greek and means “an ear of grain” referring to the inflorescence spike found in many members. They are consumed as herbal remedies in alternative medicine and wild tea (mountain tea) in Mediterranean regions. Many *Stachys* species are used in decoctions or infusions for the treatment of skin, stomach, ulcer, asthma, rheumatic disorders and vaginal tumors. Some members of genus have been reported to be used as anti-inflammatory and antibacterial agents. Moreover, their antianxiety, antioxidant and antinephritic properties have also been reported. [6] According to ethnobotanical studies, *Stachys* species generally consumed as herbal tea and a rich carbohydrate source in Europe and East Asia. A lot of *Stachys* species in

Turkey are widely used as herbal teas due to their antibacterial properties. Essential oil composition of the species is one of the main reasons of their consumption as tea in Anatolian ethnobotany [7-8], however, the species also consists of glycosides, saponins, polyphenols, tannins, phenolic acids, flavonoids and diterpenoids together with essential oils, mono and sesquiterpenoids. Therefore, the synergetic effects of the component chemicals could be the main reason of consumption of their flowers and aerial parts as tea in Anatolian culture. [6]

The essential oil compositions of the *Stachys* genus have been well documented in the literature and they mainly consist of sesquiterpenes and oxygenated sesquiterpenes The main components of the essential oil of the species were observed to be germacrene D, caryophyllenes, cadinene, spathuleneol and caryophyllene. Moreover, the monoterpenes such as α-pinene, β-pinene, phellandrene and carvacrol were also extracted from *Stachys* species. [6,9]

S. sericantha P.H. Davis is a member of section Eriostomum, endemic and Mediterranean element. Plant is perennial 35-70 cm long, corolla purple-pink coloured, 13-16 mm. It grows on stone slopes, in between 20-1300 m altitude in Antalya province and the flowering time is from June to July. [2] Plant is locally known as “dikenli cay” in the regions where it grows. [10] In Turkey, *S. sericantha* is used in the form of herb, infusion and decoction as aremedy for the treatment of cold, stomach ailments, fever and cough. [6] *S. gaziantepensis* M. Dinc and S. Dogu is an endemic penennial plant, included in section Infrarosularis. Plant is 10-30 cm long and corolla yellow coloured, 10-12 mm. It grows on marble rocks, between 1060 and 1300 m altitude in Gaziantep province. The flowering time is between second half of May and first half of June. *S. gaziantepensis* is also used as herbal tea for the treatment of cold in the regions where it grows. [11] *S. mardinensis* (Post) R. Mill is a member of section Fragilicaulis and Iranian-Turan element. Plant is perennial 10-60 cm long and corolla yellowish, 12-20 mm. It grows sloping limestone rocks and cliff crevices,

between 450 and 1200 m altitude in Mardin and Malatya provinces. The flowering time is from May to July. [2] Plant is locally known as "Kaya Pungu" or "Punge Tehta" and it used for headaches in the regions where it grows. [10,12] In the present paper, the chemical composition of the essential oils obtained from the aerial parts of endemic *S. sericantha*, endemic *S. gaziantepensis* and *S. mardinensis* from Turkey were reported. Mostprobably, this could be the first report on the essential oil chemical compositions of *S. gaziantepensis* and *S. mardinensis.*

MATERIALS AND METHODS

Plant material

Plant materials were collected during the flowering period from Antalya (*S. sericantha*), Gaziantep (*S. gaziantepensis*) and Mardin (*S. mardinensis*) provinces of Turkey (Table 1). The voucher specimens are kept at the Herbarium of the Department of Biology, Necmettin Erbakan University, Konya, Turkey (NEÜ Herb.).

Isolation of essential oil

The essential oil from air-dried plant materials was isolated by hydrodistillation for 3 h, using a Clevenger-type apparatus to produce a small amount of essential oil of *S. sericantha* which was trapped in *n*-hexane. The yields of oils were 0.11% for *S. gaziantepensis* and 0.02% for *S. mardinensis* (Table 1). The obtained oils were dried over anhydrous sodium sulphate and stored at +4°C in the dark until analysed and tested.

GC-MS analysis

The GC-MS analysis was carried out with an Agilent 5975 GC-MSD system. Innowax FSC column (60 m x 0.25 mm, 0.25 μm film thickness) was used with helium as carrier gas (0.8 ml/min). GC oven temperature was kept at 60°C for 10 min and programmed to 220°C at a rate of 4°C/min, and kept constant at 220°C for 10 min and then programmed to 240°C at a rate of 1°C/min. Split ratio was adjusted at 40:1. The injector temperature was set at 250°C. Mass spectra were recorded at 70 eV. Mass range was from *m/z* 35 to 450.

GC analysis

The GC analysis was carried out using an Agilent 6890N GC system. FID detector temperature was 300°C. To obtain the same elution order with GC-MS, simultaneous auto-injection was done on a duplicate of the same column applying the same operational conditions. Relative percentage amounts of the separated compounds were calculated from FID chromatograms. The analysis results are given in Table. Identification of the essential oil components were carried out by comparison of their relative retention times with those of authentic samples or by comparison of their relative retention index (RRI) to series of *n*-alkanes. Computer matching against commercial (Wiley GC/MS Library, MassFinder 3 Library) [13,14] and in-house "Baser Library of Essential Oil Constituents" built up by genuine compounds and components of known oils, as well as MS literature data, [15,16] was used for the identification.

RESULTS AND DISCUSSION

As in our samples, the yield of essential oil of *Stachys* species is lower than other Lamiaceae family members (e.g. *Origanum*, *Satureja*, *Sideritis*, *Salvia*, *Thymus)* however, the consumption pattern of the species is reported to be quite similar to the other members of the family. Most of species of *Stachys* have a pleasant smell. They are consumed as herbal tea in the major parts of the world and Turkey and due to their moderate antibacterial, antifungal, anti-inflammatory and anti-oxidant capacity, they are widely used as herbal remedy in alternative medicine. [6]

In this study, the water-distilled essential oils from aerial parts of the three *Stachys* species were characterized by GC-FID and GC-MS. The compounds identified from the essential oils along with their relative percentages are listed in Table 2. According to our findings, the essential oil of *S. gaziantepensis* of which 89.8% of the composition was determined, was characterized by its high proportion of monoterpene hydrocarbons (71.1%). Of the 18 components detected, the most abundant compounds were determined to be α-pinene (53%) followed by β-pinene (8.2%). In *S. mardinensis* oil, 16 components were identified representing 82.8% of the total oil. The oil consisted mainly of oxygenated monoterpenes (62.0%) and oxygenated sesquiterpenes (15.7%). Menthyl acetate (15.3%), isomenthone (15.0%), pulegone (10%), menthol (8.4%), spathulenol (7.0%) and caryophyllene oxide (6.7%) were the major components. In *S. sericantha* oil, 59 components were accounted for 82.5% of the total composition of the oil. Fatty acid+esters (41.0%) and oxygenated sesquiterpenes (22.8%) are found as main components, hexadecanoic acid (23.7%), dodecanoic acid (11.3%) and caryophyllene oxide (10.7%).

According to our results in Table 2, monoterpene hydrocarbones (71.1%) are charactereized by a high content in the oil of *S. gaziantepensis* while oxygenated monoterpenes (62.0%) are charactereized by a high content in the oil of *S. mardinensis.* Fatty acid+esters (41.0%) and

oxygenated sesquiterpenes (22.8%) are large amount in the oils of *S. sericantha.* In the earlier a study,[9] published essential oil composition of twenty-two *Stachys* species (mountain tea) which one of them is *S. sericantha*. It was identified to have 25 components which were accounted for 81.7% of the total composition of the oil. It consisted of sesquiterpenes (74.6%) and oxygenated sesquiterpenes (2.3%). Germacrene-D (32.4%), β-caryophyllene (23.2%) and α-cadinene (7.1%) as the main components. However, the essential oil composition of our samples was found to be quite different from those already reported. Both of *S. sericantha* are collected from Antalya region. But our samples are growing in Konyaaltı at 100 m and the other is growing in Kemer at 1200 m. Various factors, both endogenous and exogenous, can affect the composition of the essential oil of *S. sericantha.* We believe that the time of flowering, geographical and climatic factors may be very important. [17]

According to findings of Goren et al., [6,9] the oil composition of studied total 78 *Stachys* species in world are mainly consist of sesquiterpenes and oxygenated sesquiterpenes. The main components of the essential oil of the species were observed to be germacrene D, β-caryophyllene cadinene, spathulenol and caryophyllene oxide. Spathulenol and caryophyllene oxide were determined only in our *S. mardinensis* samples while caryophyllene oxide is found *S. sericantha.* Moreover, the monoterpenes such as α-pinene, β-pinene, phellandrene and carvacrol were also extracted from *Stachys* species. [6] The monoterpenes (71.1%), α-pinene and β-pinene, were also extracted from our *S. gaziantepensis* samples. As the main compounds, hexadecanoic acid in eight *Stachys* species and pulegone in one *Stachys* species (*S. setifera*) were reported by Goren et al. [6] However, dodecanoic acid, menthol, isomenthone and menthyl acetate as the main compounds are idendified herein oil of *Stachys* species for the first time.

Monoterpene compounds have therapeutic properties such as anti-inflammatory, antiseptic, antiviral, and antibacterial. In regards to cosmetics, monoterpene hydrocarbons are less valuable than oxygenated compounds in terms of their contribution to the fragrance of the essential oil. Conversely, the oxygenated compounds are highly odoriferous and, therefore, the most valuable. [18]

CONCLUSION

In the Anatolia folk medicine, *Stachys* species are used by many people in various villages and towns for the therapeutic value of their essential oils. In this study, water-distilled essential oil of three species of *Stachys* (Lamiaceae) was analyzed by GC-FID and GC-MS. The essential oil of *S. gaziantepensis* (endemic) was characterized by monoterpene hydrocarbons. Fatty acid+esters and oxygenated sesquiterpenes were found in *S. sericantha* (endemic) oil while oxygenated monoterpenes and oxygenated sesquiterpenes were identified from *S. mardinensis* oil.

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Table 1. *Stachys* spp. used in this study

Stachys spp.	Plant part	Collection site	Collection date	Yield (%)	Herb. No
* *S. sericantha*	Aerial part	Antalya:Konyaaltı	2010	tr	3338
**S. gaziantepensis*	Aerial part	Gaziantep:Şehitkamil	2012	0.11	3467
S. mardinensis	Aerial part	Mardin:Samandağ	2010	0.02	2472

*endemic

Table 2. The composition of the essential oil of *S. sericantha*, *S. gaziantepensis* and *S. mardinensis*

RRI	**Compound**	Ss %	Sg %	Sm %
1032	α-Pinene	0.3	**53.0**	-
1035	α-Thujene	-	0.9	-
1118	β-Pinene	0.9	**8.2**	-
1132	Sabinene	-	1.0	-
1174	Myrcene	-	3.7	-
1203	Limonene	-	3.0	-
1255	γ-Terpinene	-	0.6	-
1280	*p*-Cymene	-	0.7	-
1475	Menthone	-	-	1.5
1497	α-Copaene	0.1	-	-
1503	**Isomenthone**	-	-	**15.0**
1528	α-Bourbonene	tr	-	-
1535	β-Bourbonene	0.6	-	-
1553	Linalool	0.2	-	-
1574	**Menthyl acetate**	-	-	**15.3**
1591	Bornyl acetate	0.1	-	-
1594	Isomethylacetate	-	-	4.3
1600	β-Elemene	0.6	-	-
1610	Calarene (= *-gurjunene*)	0.1	-	-
1612	β-Caryophyllene	4.2	-	-
1632	Neoisomenthol	-	-	2.2
1638	**Menthol**	-	-	**8.4**
1648	Myrtenal	0.1	-	-
1662	**Pulegone**	-	-	**10.0**
1664	Nonanol	0.1	-	-

1687	α-Humulene	0.9	-	-
1695	(*E*)-β-Farnesene	-	1.0	-
1704	γ-Muurolene	0.6	-	-
1706	α-Terpineol	0.7	-	-
1726	Germacrene D	0.6	3.5	-
1733	*cis*-Piperitone oxide	-	-	2.9
1740	Valencene	0.1	-	-
1740	α-Muurolene	0.3	-	-
1748	Piperitone	-	-	1.6
1755	Bicyclogermacrene	-	1.8	2.7
1765	(*E*)-2-Undecanal	0.2	-	-
1766	Decanol	tr	-	-
1773	δ-Cadinene	-	1.0	-
1784	(*E*)-α-Bisabolene	0.2	-	-
1785	7-*epi*-α-Selinene	-	tr	-
1804	Myrtenol	0.1	-	-
1838	(*E*)-β-Damascenone	0.2	-	-
1868	(*E*)-Geranyl acetone	0.3	-	-
1900	*epi*-Cubebol	0.1	-	-
1941	α-Calacorene	0.2	-	-
1949	Piperitenone	-	-	0.8
1958	(*E*)-β-Ionone	0.1	-	-
1973	Dodecanol	0.1	-	-
1984	γ-Calacorene	0.2	-	-
1992	2-Phenylethyl isovalerate	1.0	-	-
2008	**Caryophyllene oxide**	**10.7**	-	**6.7**
2037	Salvial-4(14)-en-1-one	0.4	-	-
2056	Ethyl tetradecanoate	0.1	-	-
2071	Humulene epoxide-II	1.4	-	-
2081	Humulene epoxide-III	0.2	-	-

2130	Salviadienol	0.7	-	-
2131	Hexahydrofarnesyl acetone	1.6	-	0.4
2144	Spathulenol	-	1.9	**7.0**
2187	T-Cadinol	-	-	2.0
2209	T-Muurolol	0.2	-	-
2210	Copaborneol	-	2.0	-
2239	Carvacrol	-	4.4	-
2255	α-Cadinol	-	1.8	-
2262	Ethyl hexadecanate	0.3	-	-
2278	Torilenol	1.2	-	-
2298	Decanoic acid	0.6		-
2312	9-Geranyl-*p*-cymene	-	-	2.0
2324	Caryophylla-2(12),6(13)-dien-5α-ol (=*Caryophylladienol II*)	1.2	-	-
2369	Eudesma-4(15),7-dien-4β-ol	1.0	-	-
2384	Farnesyl acetone	0.5	-	-
2389	Caryophylla-2(12),6-dien-5α-ol (=*Caryophyllenol I*)	1.2	-	-
2392	Caryophylla-2(12),6-dien-5β-ol (=*Caryophyllenol II*)	3.5	-	-
2445	14-Oxo-calamenene	0.2	-	-
2500	Pentacosane	0.7	-	-
2503	**Dodecanoic acid**	**11.3**	-	-
2512	Benzophenone	0.5	-	-
2607	14-Hydroxy-δ-cadinene	0.9	-	-
2622	Phytol	0.7	-	-
2670	Tetradecanoic acid	4.3	-	-
2679	Manool	0.2	-	-
2700	Heptacosane	0.7	-	-
2800	Octacosane	0.2	-	-
2822	Pentadecanoic acid	0.7	-	-
2857	Palmito-γ-lactone	0.4	-	-
2931	**Hexadecanoic acid**	**23.7**	1.3	-

	Monoterpene Hydrocarbones	*1.2*	***71.1***	-
	Oxygenated Monoterpenes	*1.2*	*4.4*	***62.0***
	Sesquiterpene Hydrocarbones	*8.9*	*7.3*	*2.7*
	Oxygenated Sesquiterpenes	***22.8***	*5.7*	***15.7***
	Diterpenes	*0.9*	-	-
	Fatty acid+esters	***41.0***	*1.3*	-
	Others	*6.5*	-	*2.4*
	Total	**82.5**	**89.8**	**82.8**

RRI Relative retention indices calculated against *n*-alkanes % calculated from FID data
tr Trace (< 0.1 %); Ss=*S. sericantha*, Sg= *S. gaziantepensis* and Sm=*S. mardinensis*