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Composition of the essential oils of *Origanum majorana* and *O. vulgare* subsp *hirtum* growing wild in Turkey

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Abstract

The chemical composition of the essential oils isolated by hydrodistillation of the aerial parts of *Origanum majorana* L. and *O. vulgare* L. from Turkey has been examined by GC-MS. A total of 34 and 37 components were identified accounting for 97.80% and 98.61% of the oils of both *O. vulgare* and *O. majorana*, respectively. The oil of *O. majorana* contained carvacrol (43.12%), (E)-β-ocimene (13.03%), p-cymene (11.38%), linalool (10.07%) and thymol (4.46%) as main components. Major components of the *O. vulgare* oil were identified as carvacrol (48.38%), *p*-cymene (19.93%), γ-terpinene (16.29%), β-caryophyllene oxyde (1.92%), β-caryophyllene (1.16%), linalool (1.58%) and myrcene (1.32%). It is shown that carvacrol was the dominant constituent of both oils.

Key words: carvacrol, essential oil, linalool, *Origanum majorana*, *Origanum vulgare*, (E)-β-ocimene, *p-cymene*, γ-terpinene.

Introduction

Marjoram (*Origanum majorana* L.) and wild marjoram (*O. vulgare* L.) are perennial plants which grow wild in Western and Southern Turkey and other East Mediterranean countries (Davis 1982). *Origanum* species have been used in medicine and as spices since antiquity mainly because of their essential oils, which consist of a considerable amount of carvacrol and thymol (Sarer *et al.* 1982).

O. vulgare is well known as a plant with a medicinal value and as such is accepted officially in a number of countries. But the essential oil of *O. vulgare* has a limited use in perfumery, cosmetics as an aroma stimulator and strengthner (Fleischer & Sneer 1982).

Marjoram is frequently used as a flavouring agent in formulations of vermouths, bitters, confectionery, meat products and salads. Wild marjoram is one of the species used for the commercial seasoning. The dried product, the tincture and the essential oil are used by the flavour industry in various liquor formulations (Nykanen 1986; Akgül 1993). It is commonly known that the presence of essential oils and their composition determine the specific aroma of plants and the flavour of the condiment. Thus, the chemi-

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cal composition of essential oils is the most important criterion for spice identification and quality (Fleischer & Sneer 1982).

Most of the studies carried out on the oil show that the composition of these species can vary to a considerable extent, containing either carvacrol or thymol and in some cases, γ-terpinene as main component. *Origanum* species from many labiatae plants, which contain chiefly carvacrol and/or thymol together with considerable amounts of γ-terpinene and *p*-cymene, have been used for their essential oils in medicine and food industry (Heath 1981; Nykanen 1986; Akgül & Bayrak 1987).

During the past few years there has been a marked increase in the interest shown on many herbal spices and the most popular of them is oregano, which is used to enhance many kinds of foods (Fleischer & Sneer 1982). In this study, we report on the chemical composition of the essential oil obtained from the airdried parts of *O. majorana* and *O. vulgare* plants growing wild in Turkey.

Materials and methods

Plant material

Samples were collected from plants *O. majorana* and *O. vulgare* subsp *hirtum* growing wild in Içel (Büyükeceli-Gülnar) province of Southern Turkey, identified at the Department of Biology, Selcuk University, Konya, Turkey. The aerial parts were dried in the shade at room temperature.

Isolation of the essential oil

Dried aerial parts of the plants (about 100 g) were ground and subjected to hydrodistillation for 4 h using a Clevenger-type apparatus (Clevenger 1928). The oils extracted were dried over anhydrous sodium sulfate. Essential oil yields of *O. majorana* and *O. vulgare* subsp *hirtum* on dry weight basis were 3.83% and 4.40%, respectively.

Identification of components

For identification of components, analytical gas chromatography (GC) was performed using a DELSI 121 C apparatus fitted with a flame ionization detector and a CP WAX 51 fused silica column (25 m x 0.25 mm i.d., 0.25 μm film thickness). Temperature was programmed from 50° C at 5 minutes to 220° C at 3° C minute^{-1}. A CP W AX 51 fused silica WCOT column (60 m x 0.25 mm i.d., 0.25 μm) for GC/MS was used with helium as carrier gas. For GC/MS a CPWAX 52 fused silica CB column (50 m x 0.3 mm i.d.; 0.25 μm film thickness) was used with helium as carrier gas and coupled to a HP mass spectrometer: ionization energy 70 eV. Temperature programming was from 50°-240° C at 3° C minute^{-1}. The samples were injected at an injector temperature of 240° C. The components were identified by comparing linear retention indices with those from authentic compounds.

Results and discussion

Chemical compositions of the volatile oils from *O. majorana* and *O. vulgare* subsp *hirtum* are listed in Table 1. In this study, a total of 34 and 38 compounds which account for about 97.8% and 98.6% of the essential oils of *O. vulgare* subsp *hirtum* and *O. majorana*, respectively, were found. The major components of *O. vulgare* were carvacrol, *p*-cymene and γ-terpinene, while carvacrol, (E)- β- ocimene, p-cymene, linalool and thymol were recorded as the main constituents of *O. majorana*.

Carvacrol was found to be the main component in the samples, its amount in *O. vulgare* subsp *hirtum* sample was higher. The percentages of *p*-cymene and *(E)*- β-ocimene, the second major components, were 19.9% and 13.0% in *O. vulgare* subsp *hirtum* and *O. majorana*, respectively.

Thymol which is responsible, together with carvacrol, for the phenolic character was found in lower percentage in both oils, but its amount in *O. majorana* oil was higher than that of *O. vulgare* subsp *hirtum*. The volatile oils of the samples were found to comprise practically the same compounds but a few, such as tricyclene, β-phellandrene, β-humulene, β-thujene, myrcene, (*E*)-β-ocimene, linalool and thymol amounts in *O. majorana* were higher compared with *O. vulgare*. *p*-cymene was found at high amounts in *O. vulgare* subsp *hirtum*.

Table 1. Chemical composition of essential oils of *Origanum majorana* and *O. vulgare* subsp *hirtum*

Compound	Percentage (%)	
	O. majorana	*O. vulgare*
tricyclene	0.14	-*
α-pinene	1.04	0.66
α -thujene	2.24	1.15
camphene	0.39	0.11
β-pinene	0.28	0.19
sabinene	0.08	-
δ-3-carene	0.12	0.09
α-phellandrene	0.28	0.13
myrcene	2.58	1.32
α-terpinene	2.21	1.08
limonene	0.41	0.33
β-phellandrene	-	0.21
γ-terpinene	-	16.29
1,8-cineole	0.88	-
(*E*)-β-ocimene	13.03	0.08
p-cymene	11.38	19.93
terpinolene	0.16	0.06
octen-1-ol	-	0.24
octanol	0.04	-
cis-linalool oxide	0.02	-
octen-3-ol	0.20	-
cis-thujanol-4	0.88	0.41
trans-linalool oxide	0.04	-
trans-thujanol-4	0.18	0.18
linalool	10.07	1.58
trans-*p*-menth-2-en-1-ol	0.02	-
β-caryophyllene	0.28	1.66
terpinen-4-ol	1.32	0.63
α-humulene	-	0.09
carvacrol methylether	0.03	-
Cis-*p*-menth-2-en-1-ol	0.04	-
trans-pinocarveol	0.02	-
estragole	0.10	-
borneol+α-terpineol	2.32	0.25
carvone	0.06	0.06
p-cymen-9-ol	0.05	-
p-cymen-8-ol	0.02	0.10
thymyl acetate	-	0.02
iso-caryophyllene oxide	-	0.11
β-caryophyllene oxide	0.08	1.92
methyleugenol	-	0.04
humulene oxide II	-	0.12
spathulenol	0.04	0.12
eugenol	-	0.05
thymol	4.46	0.21
carvacrol	43.12	48.38

*non identified. The data are percentage composition in volatile oil.

The essential oils from *O. majorana* contained a-pinene, a-thujene, camphene, α-pinene, δ-3-carene, myrcene, limonene, (E)-β-ocimene, *p*-cymene and terpinolene as the most important monoterpene hydrocarbons, while the oil from *O. vulgare* subsp *hirtum* showed lower amount of monoterpene hydrocarbons (except for *p*-cymene). Among the monoterpenic hydrocarbons in *O. vulgare* subsp *hirtum* oil, *p*-cymene and γ-terpinene were the most important. The amount of oxygenated compounds such as 1, 8-cineole, octen-3-ol, linalool (except for *O. majorana*), terpinene-4-o1, p-menth-2-en-1-ol and carvone in both samples were found low.

Chemical compositions of wild marjoram and marjoram have been studied by several authors. Sarer *et al.* (1982) reported that the most prominent components of *O. majorana* were carvacrol (65%) and thymol (4%). In another paper, Nykanen (1986) determined the flavour composition of marjoram. Most major components were *cis*-sabinene hydrate and 4-terpineol together with *trans*-sabinene hydrate, α-terpineol and linalool. The main components in the oil from *O. majorana* collected from Halkis area (Greece) were terpinen-4-ol (37.1%), *p*-cymene (12.05%), a-terpineol (7.15%), carvacrol (3.60%), *trans*-sabinene hydrate (2.14%), *cis*-sabinene hydrate (1.43%) and thymol (0.7%) (Komaitis *et al.* 1992). Jolivet *et al.* (1971) established 46.7% of phenolic compounds consisting of thymol (as major component) and carvacrol in an Italian marjoram oil sample. Baser *et al.* (1993) determined carvacrol (78.27-79.46%), *p*-cymene (4.68-4.31%), γ-terpinene (4.84-3.72%), myrcene (1.93-1.59%) and thymol (0.48-1.53%) in the oils from *O. majorana* collected in Içel and Antalya (Southern Turkey). Charai *et al.*(1996) reported that the oil contained linalool (32.68%), terpinen-4-ol (32.30%), *p*-cymene (8.0%), sabinene (4.57%), fenchone (4.89%) and α-terpineol (3.26%) as the major constituents.

O. vulgare is generally known to yield essential oil with high contents of carvacrol or thymol. Essential oils of three different compositions contained carvacrol, thymol and 4- terpineol as main compounds (Nykanen 1986; Wilkins & Madsen 1991).

As reported previously (Fleischer & Sneer 1982; Fleischer & Fleischer 1988), carvacrol (67.3%), γ-terpinene (15.0%), *p*-cymene (5.9%), borneol (1.1%), β-caryophyllene (1.0%) and terpinen-4-ol (0.7%) were identified as the major components of *O. vulgare* subsp *hirtum* oil. Lawrence (1984) analysed the composition of the essential oils of two subpecies of *O. vulgare*. He found that essential oil of *O. vulgare* from Turkey is rich in carvacrol. Sabinene, *cis*-ocimene, caryophyllene and germacrene D have been reported to be among the most prominent components of the essential oil of wild marjoram (Lawrence 1984).

The results of analysis of *O. vulgare* subsp *hirtum* collected from 19 locations were summarized by Baser *et al.* (1993) with only ranges of oil yields and percentage amounts of four major components. Sezik *et al.* (1993) found the major components of the oil of *O. vulgare* subsp *hirtum* as carvacrol (70.47%), while subsp *gracile* contained β-caryophyllene (17.54%) and germacrene D (12.75%). Baser *et al.* (1994) summarized the results of the analysis of *O. vulgare* subsp *hirtum* collected from 23 locations in which carvacrol (23.43-78.73%) was the major component. Chalchat & Pasquier (1999) have reported, sabinene (6.5-26.0%), β-caryophyllene (6.6-10.6%), thymol (0.6-9.6%), germacrene D (13.7-25.7%) as the major compounds from *O. vulgare* oils.

The carvacrol contents of *O. majorana* and *O. vulgare* oils were lower when compared with that of literature (Sarer *et al.* 1982; Fleischer & Sneer 1982; Baser *et al.* 1993), it was higher according to that of Komatitis *et al.* (1992).

Percentage of *p*-cymene and myrcene in the oil is high when compared with the data given for marjoram oil in the literature (Komaitis *et al.* 1992). Thymol content of *O. majorana* oil was found higher than those of earlier reports (Komaitis *et al.* 1992; Baser *et al.* 1993).

In the present study, β-terpinene, *p*-cymene and γ-caryophylene contents of *O. vulgare* subsp *hirtum* were lower in comparison with other reports (Fleischer & Sneer 1982). Thymol was present in low percentage and carvacrol content was significantly high in both the oils. These variations may be due to the environmental and growth conditions that can influence the oil composition. Baser *et al.* (1993) reported that the Turkish oregano is characterized by high oil yield and high carvacrol content. This study too shows that the oils of *O. majorana* and *O. vulgare* subsp *hirtum* are typically high in carvacrol content.

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