ELSEVIER

Food Chemistry 66 (1999) 143–145

Food Chemistry

www.elsevier.com/locate/foodchem

Chemical composition of the essential oil of marjoram (*Origanum majorana* L.) from Reunion Island

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Received 18 February 1997; received in revised form 16 September 1997; accepted 13 October 1997

Abstract

Capillary gas chromatography, mass and FTIR spectrometry were used to analyse the composition of the water-distilled essential oil of marjoram (*Origanum majorana* L.) grown in Reunion Island located in Indian Ocean (21°S 55°E). Among 45 compounds recorded by gas chromatography, GC–MS and GC–FTIR 43 components were identified. The essential oil was found to be rich in terpinen-4-ol (38.4%), *cis*-sabinene hydrate (15.0%), *p*-cymene (7.0%) and γ-terpinene (6.9%).

1. Introduction

Origanum majorana L. is a hardy perennial and herbaceous plant which grows wild in its natural areas: Egypt and eastern Mediterranean countries (Furia and Bellanca, 1971). It can be grown in Northern European areas with mulching materials (Hälvä, 1987). In tropical countries fresh plants can be affected by mould diseases.

Commercial *Origanum majorana* L. oil or sweet marjoram is used as a spice and condiment. The volatile aromatic compounds are employed in the food industry as a flavoring. The oil is used in perfumery for its spicy herbaceous notes.

The composition of oils from various *Origanum* species has been investigated (Lawrence, 1989; Nykanen, 1986; Komaitis, Infanti-Papatragianni, & Melissari-Panagiotou, 1992; Baser, Kirimer, & Tümen 1993; Ravid & Putievsky, 1986). It was postulated that the oils exist in two forms. One with terpinen-4-ol and sabinene hydrate as major components and the other with thymol and/or carvacrol as predominant compounds.

In the present study, the oil obtained from fresh flowering plants was analysed using gas chromatography, GC–MS and GC–FTIR.

2. Materials and methods

The plants were collected in the north-west of Reunion. Marjoram plants were grown in the fields of the Agricultural High School of Saint-Paul in 1993. Fresh flowering plants were hydrodistilled in a Clevenger-type apparatus for 5 h. The essential oil obtained was dried over anhydrous sodium sulfate and stored at −15°C until used. The gas-chromatographic determinations were run on Hewlett-Packard 5890 series II instrument using a BP-1 fused silica capillary column (50 m × 0.25 mm i.d.) coated with 0.25 μm of dimethyl siloxane. The oven temperature programme was 60°C rising at 2°C/min to a final temperature of 260°C. Peak areas and retention times were calculated using Hewlett-Packard Alice Software.

The essential oil was also examined by Intersmat IGC 120 FL gas chromatograph equipped with a flame ionization detector (FID), coupled with the Fisons Instruments Chrom-Card Software to determine the GC peak areas. Separation was performed on a Carbowax 20 M fused silica capillary column (50 m × 0.32 mm i.d.) coated with 0.20 μm of polyethylene glycol. We operated under similar above conditions. Retention indices (R.I.) of compounds were determined relatively to the retention times of a series of *n*-paraffin hydrocarbons with a logarithmic scale.

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0308-8146/99/$ - see front matter
PII: S0308-8146(98)00018-1

Table 1
Percentage composition of the essential oil of *Origanum majorana* L.

No. Pics	Compounds	Area percentage	R.I. apolar		R.I. polar	
			Ref.	Exp.	Ref.	Exp.
1	α-Thujene	0.35	938	926	1036	1020
2	α-Pinene	0.61	942	934	1039	1023
3	Camphene	0.04	954	948	1083	1063
4	Sabinene	4.94	976	970	1130	1121
5	β-Pinene	0.39	981	975	1124	1108
6	Myrcene	1.64	986	983	1156	1162
7	α-Phellandrene	0.05	1002	1000	1177	1168
8	α-Terpinene	2.75	1016	1014	1180	1181
9	p-Cymene	7.01	1020	1017	1272	1267
10	Limonene	1.36	1030	1026	1206	1202
11	*cis*-Ocimene	0.25	1025	1029	1228	1212
12	*trans*-Ocimene	0.04	1038	1039	1250	1244
13	γ-Terpinene	6.89	1057	1053	1251	1248
14	Terpinolene	1.82	1085	1082	1287	1278
15	*trans*-sabinene hydrate	3.49	1078	1085	1464	1464
16	Linalool	0.23	1092	1085	1506	1501
17	*cis*-Sabinene hydrate	14.95	1092	1091	1546	1556
18	α-Pinene oxyde	tr	1096	1095	—	—
19	*trans*-para-Menth-2en-1-ol	1.84	1111	1115	—	1586
20	Camphor	tr	1136	1138	1518	1535
21	*cis*-para-Menth-2-en-1-ol	0.79	1128	1130	1562	1565
22	Terpinen-4-ol	38.40	1175	1173	1628	1609
23	α-Terpineol	4.88	1185	1181	1661	1702
24	*trans*-Piperitol	0.40	1181	1186	1675	1679
25	Isoborneol	0.03	—	1188	1660	1656
26	*cis*-Piperitol	0.26	1190	1196	1741	1719
27	Nerol	0.19	1218	1214	1757	1760
28	Geraniol	0.19	1243	1236	1797	1764
29	Linalyl acetate	0.30	1246	1240	1538	1535
30	unknown	0.07	—	1249	—	—
31	Bornyl acetate	0.06	1279	1274	1584	1578
32	*trans*-Sabinyl acetate	0.11	1278	1276	1651	1643
33	Thymol	0.15	1287	1284	2100	2095
34	5-Caranol *	tr	—	1286	—	—
35	Unknown	0.07	—	1300	—	—
36	*cis*-dihydro-α-Terpinyl acetate *	tr	—	1325	—	—
37	α-Terpinyl acetate	0.02	1333	1337	1687	1687
38	Neryl acetate	0.05	1345	1342	1699	1708
39	Geranyl acetate	0.11	1364	1359	1735	1728
40	β-Caryophyllene	1.16	1428	1424	1617	1632
41	α-Humulene	0.02	1465	1457	1682	1673
42	Bicyclogermacrene	0.60	1498	1500	1768	1750
43	Spathulenol	0.25	1564	1566	2153	2171
44	Caryophyllene oxyde	0.11	1576	1582	2000	1985
45	Acorenone *	tr	1655	1661	—	—

* Tentative identification from mass spectra data; tr, trace (< 0.02%); R.I., retention indices.

GC–MS analysis were carried out on Fisons Instruments mass spectrometer (VG 70-250 SQ) equipped with the Wiley Library sofware coupled with a Hewlett-Packard gas chromatograph 5890 series II. Capillary GC conditions as above were employed here. Mass spectra were recorded with the significant MS operating parameters: ionization voltage, 70 eV; ion source temp, 190°C; scan mass range, 35–285 u. Unknown compounds were identified using published mass spectra and retention indices.

GC–FTIR analyses were carried out on a Nicolet 20 SXC Instrument with the Nicolet Aldrich Vapor Phase Library Software, coupled with Perkin–Elmer 8500 gas chromatograph. Separation was performed on a HP-1 fused silica capillary column (50 m × 0.32 mm × 0.1 μm). GC/FTIR conditions were the same as those used for GC analysis.

3. Results and discussion

The hydrodistillation of fresh flowering plants gave a colourless oil with a yield of ~1 ml per 100 g. The compounds identified and their relative proportions are listed according to their order of elution on BP-1 column in Table 1. Out of the 45 constituents separated by GC, 43 were identified. The identification of all components was based on their mass spectra and their retention indices measured on the BP-1 and Carbowax 20 M fused silica capillary columns.

In this study, the most prominent components were terpinen-4-ol (38%), *cis*-sabinene hydrate (15%), *p*-cymene (7%) and γ-terpinene (7%), together with sabinene (5%), α-terpineol (5%) and α-terpinene (3%). They constituted about 80% of total essential oil.

Investigation of the essential oil of marjoram has shown that the volatile aroma composition varies with the origin of the plant (Nykanen, 1986). Therefore, it should be noted that in acidic solution sabinene hydrate rearranges to give terpinen-4-ol and small amounts of γ-terpinene, α-terpinene and *p*-cymene (Granger et al., 1975; Fischer, Nitz, & Drawert, 1987). Nervertheless, Granger et al. (1975) have studied the various conditions, and concluded that only little artefact formation is caused by steam distillation.

However, according to earlier work, there exist two main chemotypes. One consists mostly of monoterpene alcohols and the other of phenols. In the first chemotype, terpinen-4-ol, either alone or together with other monoterpene alcohols such as *cis*- and *trans*-sabinene hydrate, has been found to be the main volatile components (Sarer et al., 1982). For the second chemotype marjoram oils rich in phenols consisted mainly of thymol (Jolivet et al., 1971) and/or carvacrol (Baser et al., 1993; Sarer et al., 1982).

The present study shows that essential oil of *Origanum majorana* L. grown in Reunion Island with a tropical climate, presents characteristics of the first chemotype.

Acknowledgements

The authors wish to thank Mr Auge for growing and collecting plant material.

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