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EFFECT OF HYDRODISTILLATION AND MICROWAVE DISTILLATION EXTRACTION METHODS ON CHEMICAL COMPOSITIONS OF ESSENTIAL OIL OF PICKLING HERB AND MYRTLE PLANTS

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Abstract. *The essential oil composition of pickling herb and myrtle was investigated by GC and GC-MS. The most prominent components for pickling herb were methyl eugenol (84.18%), δ-3-carene (5.02%) and α-phellandrene (2.59%) for Clevenger method. In addition, the main constituents of oil obtained by microwave distillation were δ-3-carene (38.20%) and α-phellandrene (24.72 %). The major components of myrtle oil obtained by using Clevenger distillation method were linalool (28.28%), α-pinene (22.99 %), 1,8-cineole (14.94 %), linalyl acetate (9.36 %), limonene (4.87 %) and α-terpineol (4.48 %). According to another method, myrtle oil contained 39.32 % 1,8-cineole, 12.34 % α-terpineol, 9.98 % linalyl acetate, 6.87 % limonene, 6.57 % linalool, 5.27 % α-pinene and 3.49% α-terpenyl acetate. In addition myrtle oil, α-phellandrene, δ-3-carene, β-phellandrene contents of pickling herb oil extracted by Clevenger method were found high according to the same concentration of oil obtained by microwave.*

Key words: *pickling herb, myrtle, essential oil, hydrodistillation, solvent-free microwave distillation.*

INTRODUCTION

The genus *Echinophora* (Apiaceae) is represented in the flora of Turkey by 6 species including 3 endemic (Davis 1982) . It is called as “çörtük, turşu otu” in Turkish. The plant is also used in folk medicine to heal wounds and to treat gastric ulcers due to its antifungal, carminative, and digestive properties (Baytop 1984). In some previous study, the characteristic compounds of pickling herb (*E. tenuifolia*) essential oil were methyl

eugenol, α-phellandrene, δ-3-carene and p-cymene (Tanker et al. 1976, Akgület al. 1989, Özcan et al. 2002, Chalchat et al. 2007).

The myrtle tree (*Myrtus communis* L.), belongs to the Myrtaceae family and is a typical representative of the Mediterranean flora. In Turkey, myrtle trees are found growing in pine forests and along river beds, particularly in the Taurus mountains, from just above sea level to 500-600 m. The myrtle tree is called “hambeles”, “mersin” or “murt” in Turkish (Baytop 1984, Özek et al. 2000; Özcan & Chalchat 2004, Farah et al. 2006, Uyar 2006). Its fresh and/or dried leave oils are used in cosmetics, sauces, and confectionary and beverage industries (Baytop 1984, Karamanoğlu 1972, Mazza 1983, Duke 1988).

Recently, the chemical composition of the essential oils of *Echinophora tenuifolia* and *Myrtus communis* leaves have been reported (Akgül & Chialva 1989, Özcan et al. 2002, Chalchat et al. 2007, Özek et al. 2000, Özcan & Chalchat 2004, Farah et al. 2006, Uyar 2006). But, a literature search showed no previous studies of the chemical composition of oils obtained from these plants via microwave distillation. In comparison with hydrodistillation, microwave offers important advantages such as similar yields, shorter extraction times and substantial savings of energy. In addition, this proposed method reduces the environmental burden, with less CO_2 released into the atmosphere (Lucchesi et al. 2004, Benkaci-Ali et al. 2007).

The purpose of this study was to determine differences of essential oil yield and composition of pickling herb and myrtle plants by using microwave distillation and hydrodistillation.

MATERIAL AND METHODS

Plant material

Aerial parts of pickling herb (*E.tenuifolia L. spp. sibthorpiana*) and leaves of myrtle (*Myrtus communis* L.) plants were obtained from Konya (Selçuklu) and Mersin (Büyükeceli-Gülnar) in June and August, 2007, respectively. The samples were transported in bags, and were dried to constant weight at +4 ^{o}C until extraction. Specimens had been deposited in the Food Engineering museum of the University of Selçuk in Konya, Turkey.

Hydrodistillation processing (Clevenger method)

Essential oils of pickling herb and myrtle leaves were obtained by the hydrodistilled method. The plant materials (about 150 g), cut into small pieces, were placed in a flask (2 L) together with double-distilled water (1.5 L). The mixture was boiled for 4 h. The extracted oil was dried over anhydrous sodium sulfate, and kept at -18 ^{o}C until analyzed.

Microwave processing

Microwave distillation was carried out at atmospheric pressure in a microwave laboratory oven (MILESTONE, Dry DIST, Solvent-Free Microwave Extraction Labstation), as described previously (Lucchesi et al, 2004; Benkaci-Ali et al., 2007). Each ground material (100 g) was heated until no more essential oil was obtained, using a power of 800 W for 10 min with the addition of 50 ml distilled water [14,16]. The volatile oil obtained was dried over anhydrous sodium sulfate. The yields (v/w) calculated on a dry weight basis of pickling herb and myrtle were 2.35% and 2.75% to 0.62 and 0.83% for Clevenger and microwave distillation, respectively.

Analytical GC

Identification of components was carried out with an analytical HP 5890 gas chromatograph equipped a flame ionization detector and a CP WAX 51 fused silica column (25 m x 0.3 mm; 0.25 µm film thickness). Temperature was set initially at 50 °C for 5 min and programmed to reach 220 °C at the rate of 3 °C/min. Injector temperature was 240 ^{o}C. Retention indices were determined relative to the retention times of a series on n-alkanes with linear interpolation.

GC-MS Analysis

A CP WAX 51 fused silica WCOT column (60 m x 0.3 mm) for GC/MS was used with helium as the carrier gas (flow rate 1 ml/min) and coupled to a HP mass spectrometer: ionization energy 70 eV. Temperature was programmed from 50 to 240 °C at 3°C/min. The components were identified by comparing linear Kovats indices, their retention times, and mass spectra with those obtained from the authentic samples and/or the MS library. The library search was carried out using a Wiley GC/MS Library of Essential Oil Constituents.

The percentage composition of the essential oil was computed from GC peak areas without correction factors. Qualitative analysis was based on a comparison of retention times and mass spectra with corresponding data in the literature (Adams, 2001).

RESULTS AND DISCUSSION

A total of 31, 21 and 35, 42 components, respectively, of pickling herb oil and myrtle oil by using hydrodistillation and microwave were identified by GC-MS analyses. The main components representing 97.15% to 99.1% and 97.54% to 95.19% of both oils, respectively, are given in Table 1 and Table 2. The oil yields changed from plant to plant and were different by distillation method. The yields increased somewhat with the microwave method. While pickling herb contained 2.35% and 2.75% oil for Clevenger method and microwave distillation, respectively, myrtle leaves contained 0.62 % and 0.83 % oil, respectively.

Table 1.

Chemical composition of pickling herb essential oil (%).

RT	RI	Constituents	Distillation methods	
			Clevenger	Microwave
6.75	838	4-hydroxy-4-methyl-pentan-2-one	0.20	0.22
9.74	926	Alpha-thujene	-*	0.24
10.01	939	Alpha-pinene	0.05	0.30
11.40	972	Sabinene	-	0.26
11.97	989	Myrcene	0.24	1.69
12.56	1005	Alpha-plellandrene	2.59	24.72
12.63	1008	Delta-3-carene	5.02	38.20
12.93	1017	Alpha terpinene	-	0.09
13.19	1025	p-cymene	1.11	1.97
13.35	1030	Limonene	0.26	1.27
13.39	1031	Beta-phellandrene	0.40	2.24
13.50	1034	1,8-cineole	0.09	-
13.56	1036	(Z)-beta-ocimene	0.34	1.22
13.83	1044	Benzene acetaldehyde	0.05	-
13.91	1046	(E)-beta-ocimene	0.23	0.82
14.30	1058	Gamma-terpinene	0.05	0.22
15.05	1081	Para mentha-2,4(8)-diene	0.11	0.37
15.18	1085	Terpinolene	0.30	1.34
15.34	1090	2,5-dimethylstyrene	0.07	-
15.64	1099	Linalool	0.12	0.06
16.05	1112	1,3,8-menthatriene	0.11	0.18
16.44	1125	Cis-para-menth-2-ene-1-ol	0.03	-
16.99	1144	Trans-para-menth-2-ene-1-ol	0.03	-
17.60	1164	Delta-terpineol	0.17	-
17.88	1173	Mentha-1,5-dien-8-ol	0.16	-
18.13	1182	Para cymene-8-ol	0.25	0.06
18.62	1198	Alpha terpineol	0.09	-
18.82	1205	Cis sabinol	0.41	0.20
18.97	1210	Verbenone	0.03	-
19.87	1242	Piperitenone	0.09	-
21.47	1299	Carvacrol	0.13	-
22.87	1353	Eugenol	0.10	-
23.95	1394	(Z)-jasmone	0.14	-
24.10	1399	Methyl eugenol	84.18	22.23
		Total	97.5	99.1

*nonidentified

Table 2.

Chemical composition of myrtle essential oil (%).

RT	RI	Constituents	Distillation methods	
			Clevenger	Microwave
6.75	838	4-hydroxy-4-methyl-pentan-2-one	0.21	0.23
7.21	852	E-hex-2-enal	0.26	0.13
7.28	854	Hexenol	0.20	0.19
9.34	914	İsobutyrate d'isobutyle	1.51	0.10
9.74	926	Alpha-thujene	0.13	-*
10.01	939	Alpha-pinene	22.99	5.29
11.57	977	Beta-pinene	0.16	0.14
11.97	989	Myrcene	0.71	0.09
12.44	1002	2-methylbutyrate d'isobutyle	-	-
12.56	1005	Alpha-plellandrene	0.10	0.15
12.63	1008	Delta-3-carene	-	0.23
13.19	1025	p-cymene	0.29	0.43
13.35	1030	Limonene	4.87	6.87
13.50	1034	1,8-cineole	14.95	39.32
13.56	1036	(Z)-beta-ocimene	0.22	-
13.91	1046	(E)-beta-ocimene	0.42	0.07
14.30	1058	Gamma-terpinene	0.06	0.15
14.73	1071	Cis linalol oxyde	0.21	-
15.18	1085	Terpinolene	0.16	0.06
15.24	1087	Trans linalool oxide	0.15	-
15.67	1100	Linalool	28.28	6.57
15.76	1103	3,7-dimethyl-1,5,7-octatrien-3-ol		0.07
16.96	1143	Cis sabinol	0.11	0.64
17.84	1172	Delta-terpineol	-	0.24
18.15	1182	Terpinene-4-ol	0.14	0.65
18.35	1189	Cis mentha-1(7),8diene-2-ol	-	0.27
18.62	1198	Alpha-terpineol	4.48	12.34
18.65	1199	Estragole	0.37	-
19.26	1221	Trans-carveol	-	0.25
19.38	1225	Nerol	0.57	-
19.43	1227	Citronellol		0.17
19.56	1231	Trans mentha-1(7),8diene-2-ol		0.15
20.13	1252	Linalyl acetate	9.36	9.98
20.29	1258	Geraniol	-	0.27
20.60	1268	Methyl citronelate	-	0.07
21.46	1299	Carvacrol	-	0.14

Table 2. (continued)

RT	RI	Constituents	Distillation methods	
			Clevenger	Microwave
22.04	1321	Methyl geraniate	0.15	0.43
22.13	1324	Myrtenyl acetate	0.10	0.18
22.87	1353	Alpha-terpenyl acetate	0.70	3.49
22.87	1353	Eugenol	-	0.08
23.01	1358	Neryle acetate	0.94	-
23.52	1377	Geranyl acetate	3.64	0.89
23.68	1383	Cis myrtanyle acetate	0.06	-
24.10	1399	Methyl eugenol	0.50	2.26
24.69	1423	Beta-caryophyllene	0.18	0.22
25.59	1459	Alpha-humulene	0.18	0.27
26.45	1494	E-methyl isoeugenol	-	0.10
27.47	1538	Flavesone	-	0.19
28.61	1586	Caryophyllene oxide	-	0.79
28.99	1603	Humulene oxyde isomere	-	0.10
28.24	1614	Humulene epoxide II	-	0.68
29.25	1615	Humulene-1,2-epoxyde	0.11	-
29.84	1641	caryophylla-4(12),8(13)-diene-5-beta-ol	-	0.16
		Total	97.54	95.19

*nonidentified

The percentage of the essential oil compositions of pickling herb oil obtained by the Clevenger and microwave distillation methods are given in Table 1.

The most prominent components for *E. tenuifolia* were methyl eugenol (84.18%), δ-3-carene (5.02%), α-phellandrene (2.59%) and *p*-cymene (1.11%) for the Clevenger method. The main constituents of oil obtained by microwave distillation were δ-3-carene (38.20%), α-phellandrene (24.72 %), methyl eugenol (22.23%) and terpinolene (1.34%).

The characteristic compounds of myrtle oil obtained by Clevenger method and microwave distillation are listed in Table 2 in order of their experimental retention times and indices. The major components of myrtle oil obtained by using Clevenger distillation method were linalool (28.28%), α-pinene (22.99 %), 1,8-cineole (14.94 %), linalyl acetate (9.36 %), limonene (4.87 %), α-terpineol (4.48 %) and geranyl acetate (3.64 %). According to another method, myrtle oil contained 39.32 % 1,8-cineole,

12.34 % α-terpineol, 9.98 % linalyl acetate, 6.87 % limonene, 6.57 % linalool, 5.29 % α-pinene, 3. 49% α-terpenyl acetate, and 2.26 % methyl eugenol. The main constituents of both pickling herb and myrtle oils extracted by the hydrodistillation method were generally found similar when compared with literature data (Akgül & Chialva 1989, Özcan et al. 2002, Chalchat et al. 2007, Özcan & Chalchat 2007, Başer et al. 1994, 1998, Chalchatet al. 1998, Asllani 2000, Senatore et al. 2006, Özcan & Akgül 2003).

While 1,8-cineole and α-terpineol contents of myrtle oil extracted with the Clevenger distillation method were high, α-pinene and linalool decreased. Linalyl acetate contents of both oils were found similar. In addition to myrtl oil, α-phellandrene, δ-3-carene, β-phellandrene contents of pickling herb oil extracted by Clevenger method were found high according to the same concentration of oil obtained by microwave. But, methyl eugenol content of pickling herb obtained by microwave was found too much high (84.18 %).

The several concentrations and higher number of compounds extracted by hydrodistillation, when compared to microwave, is probably related to the possible degradation of products by hydrolysis, oxidation, and *trans*-esterification because of longer extraction time and because a greater quantity of water is used (Benkaci-Ali et al. 2007). In addition, when the results were compared with the literature, the oils showed similarities and differences. The observed differences may be probably due to environmental, soil, locations and nutritional status of the plants as well as other factors that can influence the oil composition. In comparison with hydrodistillation, microwave offers important advantages such as similar yields, shorter extraction times and substantial savings of energy. In addition, this proposed method reduces the environmental burden, with less CO_2 released into the atmosphere (Benkaci-Ali et al. 2007).

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REFERENCES

Adams, R.P. (2001): Identification of Essential oil Components by Quadrupole Chromatography – Mass Spectrometry (GC/MS). Allured Publishing Corp., Carol Stream, IL..

Akgül, A., Chialva, F. (1989): Constituents of the essential oil of *Echinophora tenuifolia* L. subsp.*sibthorpian* (Guss.)Tutin from Turkey. Flavour and Fragrance Journal 4: 67-68.

Asllani, U. (2000): Chemical composition of Albanian myrtle oil (*Myrtus communis* L.). Journal of Essential Oil Research 12(2):140-142 .

Baytop, T. (1984): Therapy with Medicinal Plants in Turkey (Post and Present). İstanbul Univ.Publ. No.3255, İstanbul. [in Turkish]

Başer, K.H.C., Erdemgil, F.Z., Özek, T. (1994): Essential oil of *Echinophora tenuifolia* L. subsp. *sibthorpiana* (Guss.) Tutin. Journal of Essential Oil Research 6: 399-400.

Başer, K.H.C., Kürkçüoğlu, M., Malyer, H., Bıçakçı, A. (1998): Essential oils of six *Echinophora* species from Turkey. Journal of Essential Oil Research 10: 345-351.

Benkaci-Ali, F., Baaliouamer, A., Meklati, B.Y., Chemat, F. (2007): Chemical composition of seed essential oils from Algerian *Nigella sativa* extracted by microwave and hydrodistillation. Flavour and Fragrance Journal 22: 148-153.

Chalchat, J.C.; Garry, R.P.; Michet, A. (1998): Essential oils of myrtle (*Myrtus communis* L.) of the Mediterranean littoral. Journal of Essential Oil Research 10: 613-617.

Chalchat, J-C., Özcan, M.M., Dağdelen, A., Akgül, A. (2007): Variability of essential oil composition of *Echinophora tenuifolia* subsp. *sibthorpiana* Tutin by harvest location and year and oil storage. Chemistry of Natural Compounds 43: 225-227.

Davis, P.H. (1982): Fora of Turkey and the East Aegean Islands. Vol 4, Edinburgh Universty Pres, Edinburgh, 9 pp.

Duke, J.A., (1988): Handbook of Medicinal Herbs, CRC Press, Boca Ruton, FL,U.S.A. pp198-199.

Farah, A., Afifi, A., Fechtal, M., Chhen, A., B. Satrani, B., Talbi, M. (2006): Fractional distillation effect on the chemical composition of Moroccan myrtle (*Myrtus communis* L.) essential oils. Flavour and Fragrance Journal 21: 351-354.

Karamanoğlu, K. (1972): Pharmaceutic Botanic. Ankara Univ. Ecz. Fak. Yay., Ankara.

Lucchesi, M.E., Chemat, F., Smadja, J. (2004): Solvent-free microwave extraction of essential oil from aromatic herbs: comparison with conventional hydrodistillation. Journal of Chromatography A 1043: 323-327.

Mazza, G., (1983): Gas chromotographic-mass spectrometric investigation of the volatile components of myrtle berries (*Myrtus communis* L.). Journal of Chromatography A 264: 304-311.

Özcan, M., Chalchat, J-C., Akgül, A. (2002): Composition of the essential oil of *Echinophora tenuifolia* L.subsp.*sibthorpiana* (Guss.) Tutin from Turkey. Journal of Essential Oil Research 14: 23-24.

Özcan, M.; Akgül, A. (2003): Essential oil composition of Turkish pickling herb (*Echinophora tenuifolia* L. subsp.*sibthorpiana* (Guss.) Tutin). Acta Botanica Hungarica 45: 163-167.

Özcan, M., Chalchat, J-C. (2004): Effect of collection period on the flavour profiles of the leaves of myrtle tree (*Myrtus communis* L.) growing wild in Turkey. Research Journal of Chemical and Environmental Sciences 8(1):70-73.

Özek, T., Demirci, B., Başer, K.H.C. (2000): Chemical composition of Turkish myrtle oil. Journal of Essential Oil Research 12: 541-544.

Pare, J.R.J., Belanger, J.M.R., Stafford, S.S. (1994): Microwave-assisted process (MAP): a new tool for the analytical laboratory. Trends in Analytical Chemistry 13:176-184 .

Senatore, F., Formisano, C., Napolitano, F., Rigano, D., Özcan, M. (2006): Chemical composition and antibacterial activity of essential oil *Myrtus communis* L. growing wild in Italy and Turkey. Journal of Essential Oil Research 9(2):162-169.

Tanker, N., Tanker, M., Şener, B., Baerheim-Svendsen, A. (1976): Determination by gas chromatography of *Echinophora tenuifolia* L. subsp.*sibthorpiana* (Guss.) Tutin essential oil. Journal of Faculty of Pharmacy of Ankara University 6: 161-180. [in Turkish].

Uyar, B. (2006): Mersin Bitkisinin (*Myrtus communis* L.) yaprak, meyve ve taze dallarının aroma bileşenleri ve yaprak uçucu yağ ve ekstraktlarının antibakteriyal etkisinin belirlenmesi [*Determination of the essential oils of leaves, fruits and fresh stems of myrtle plant (Myrtus communis L.) and antibacterial activity of extracts and essential oils of the leaves*]. Msc Thesis, S.Ü. Graduate School of Natural and Applied Sciences Department of Food Engineering, p33, Konya-Turkey.