International Journal of Plant Science and Ecology
Vol. 1, No. 4, 2015, pp. 138-141
http://www.aiscience.org/journal/ ijpse

Essential Oil Composition of *Marrubium vulgare* L. Cultivated in Egypt

Hussein A. H. Said-Al Ahl[1, *], Ahmed S. H. Gendy[2], Abeer A. Mahmoud[3], Hanaa F. Y. Mohamed[3]

[1]Medicinal and Aromatic Plants Research Department, National Research Centre, Dokki, Giza, Egypt
[2]HorticultureDepartment, Faculty of Agriculture, Zagazig University, Zagazig, Egypt
[3]Botany Department (Plant Physiology Section), Faculty of Agriculture, Cairo University, Cairo, Egypt

Abstract

The essential oil of the aerial parts of *Marrubium vulgare* L. (Lamiaceae) obtained by hydro distillation was analyzed by GC/MS in order to determine their chemical composition. Thirty-two components in the oil of *M. vulgare* were identified. The GC/MS revealed the presence of carvacrol (36.28%), β-phellandrene (15.49%), carvyl acetate (11.52%) which represents the main compounds of volatile oil extracted from *Marrubium vulgare* fresh herb. Other important compounds as trans-caryophyllene (4.06%), linalool (3.86%), α-terpinene (3.83%),β-pinene (3.53%), trans-sabinene hydrate (3.29%), β-thujone (2.93%), 1-octen-3-ol (2.48%), 1,8-cineol (1.49%), α-Pinene (1.44%) and borneol (1.12%) were reported in *Marrubium vulgare* essential oil cultivated in Egypt.

Keywords

Marrubium Vulgare, Essential Oil, Carvacrol, β-Phellandrene, Carvyl Acetate

Received: April 6, 2015 / Accepted: April 29, 2015 / Published online: May 25, 2015

@ 2015 The Authors. Published by American Institute of Science. This Open Access article is under the CC BY-NC license.
http://creativecommons.org/licenses/by-nc/4.0/

1. Introduction

Marrubium vulgare L. (Lamiaceae), commonly known as white horehound, is a perennial, herbaceous medicinal plant native to temperate regions.Itis distributed across Mediterranean Europe, southern America and northern Africa and used for medicinal purposes and as ingredient in skin cosmetics and cough lozenges (Zerbe et al., 2014). This plant was frequently employed as folk medicine to treat a variety of ailments, exhibits antispasmodic and antinociceptive effects. It possesses tonic, aromatic, stimulant, expectorant, diaphoretic and diuretic properties. It is helpful for bronchial asthma and nonproductive cough. It was formerly much esteemed in various uterine, visceral and hepatic affections and in phthisism (Adel et al., 2011). The plant is reported to possess hypoglycemic (Jorge et al., 2012), antibacterial (Masoodi et al., 2008), antidiabetic (Boudjelal et al., 2012), Gastroprotective activity (Oliveira et al., 2011) and many other reported biological activities. Essential oils extracted by distillation from aromatic plants are appreciated for their bioactive efficacy as fungicides, bactericides (Zarai et al., 2011), antioxidant (Jorge et al., 2012) and other biological activities. Marrubiin is diterpenoid lactone that constitutes the bitter principle of the horehound and many other medicinal plants of the family Lamiaceae, which are used in several countries to treat different pathologies (Marrelli et al., 2013). Extensive pharmacological studies have demonstrated that marrubiin displays a suite of activities including ant nociceptive (De Jesus et al., 2000), antioxidant, antigenotoxic (Mnonopi et al., 2011), cardio protective (Laonigro et al., 1979), vasorelaxant (El Bardai et al., 2003), gastroprotective (Paula de Olivera et al., 2011), antispasmodic (Zaabat et al., 2011), immunomodulating (Karioti et al, 2007), antioedematogenic (Hellen et al., 2006), analgesic (De Souza et al., 1998; Meyre-Silva et al., 2005), and antidiabetic

* Corresponding author
E-mail address: hussein_saidalahl@yahoo.com (H. A. H. Said-Al Ahl)

properties (Mnonopi et al., 2012).

The crude extract of *Marrubium vulgare* is widely used as antihypertensive treatment in traditional medicine. It has been shown to decrease systolic blood pressure in spontaneously hypertensive rats and to inhibit KCl-induced contraction in rat aorta (El Bardai et al., 2001).The essential oil and the extract obtained from the aerial parts of *Marrubium vulgare* have been shown to have strong antimicrobial and antioxidant activities (Firuzi et al., 2010). The main active ingredient that is produced and accumulated in the aerial parts of the plant is a diterpenoid known as marrubiin (Piccoli and Bottini, 2008). A substantial antioxidant, anticoagulant, antiplatelet and antiinflamatory effects have been attributed to marrubiin (Mnonopi et al., 2011). *Marrubiumvulgare*, also contains marrubenol and phenylpropanoid esters which have been shown to exhibit L-type calcium channel blocking and cyclooxigenase (COX) inhibitors activities (El-Baradai et al., 2003; Sahpaz et al., 2002). Further, phenylpropanoids have been proved to protect cardiomyocyte against hypoxia-induced death (Zhang et al., 2009). Biological activities of *M. vulgare* are attributed to an array of diterpenoids, sterols, flavonoids and phenylpropanoids (Meyre-Silva and Cechinel-Filho, 2010).The aim of this study was to identify andestablish the chemical composition of the essential oil of *Marrubiumvulgare* growing in Egypt.

2. Materials and Methods

2.1. Plant Material

Seeds of white horehound (*Marrubium vulgare* L.) were obtained from the HEM ZADENB.V - P.O. Box 4 - 1606 ZGVenhuizen - The Netherlands. Seeds were sown in the nursery on 15th February, 2012. On 10 April 2012, uniform seedlings were transplanted into the experimental farm of the Faculty of Pharmacy, Cairo University, Giza, Egypt, which represents clay-loamy soil. The flowering fresh herb was collected at the end of July.

2.2. Gas Chromatography/Mass Spectrometry (GC/MS)

Extraction of essential oil: Fresh herb (100 g) was extracted by water distillation using Clevenger apparatus for 2 h according to Guenther (1961).GC/MS Analytical Condition: The volatile oil analysis was carried out using gas chromatography-mass spectrometry instrument stands at the Central Laboratories, National Research Center with the following specifications. Instrument: a TRACE GC Ultra Gas Chromatographs (THERMO Scientific Corp., USA), coupled with a THERMO mass spectrometer detector (ISQ Single Quadrupole Mass Spectrometer).The GC/MS system was equipped with a TG-WAX MS column (30 m x 0.25 mm i.d., 0.25 μm film thickness). Analyses were carried out using helium as carrier gas at a flow rate of 1.0 mL/min and a split ratio of 1:10 using the following temperature program: 40 ^{o}C for 1 min; rising at 4.0 ^{o}C/min to 160 ^{o}C and held for 6 min; rising at 6 ^{o}C/min to 210 C and held for 1min. The injector and detector were held at 210 ^{o}C. Diluted samples (1:10 hexane, v/v) of 0.2 μL of the mixtures were always injected. Mass spectra were obtained by electron ionization (EI) at 70 eV, using a spectral range of m/z 40-450. Most of the compounds were identified using two different analytical methods: relative retention time and mass spectra (authentic chemicals, Wiley spectral library collection and NSIT library).

3. Results and Discussion

The essential oil of *Marrubium vulgare* growing in Egypt was subjected to detailed GC/MS analysis. Exactly 32 compounds, mostly aromatic, were identified, representing 99.39 % of the total oil. The major compounds were carvacrol 36.28%, β-phellandrene15.49% and carvyl acetate 11.52%. Other important compounds weretrans-caryophyllene (4.06%), linalool (3.86%), α-terpinene (3.83%), β-pinene (3.53%), trans-sabinene hydrate (3.29%), β-thujone (2.93%), 1-octen-3-ol (2.48%), 1,8-cineol (1.49%), α-Pinene(1.44%) andborneol (1.12%).

The chemical compositions of *M. vulgare*essential oil from various origins have been the subject of many studies. The literature reveals the occurence of several chemotypes. From Lithuania, (Z)-β-farnesene, β-caryophyllene, (E)-2-hexenal, α-humulene and germacrene-D were the main components of *M. Vulgare* essential oil (Weel et al., 1999). From Czech Republic, the main constituents of *M. vulgare*essential oil were β-caryophyllene and germacrene-D (Nagy and Svajdlenka, 1998). In Poland, the main components of the oil of *Marrubium vulgare* were E-caryophyllene, germacrene D and δ-amorphene (Zawislak, 2012). In Tunisia, β-bisabolene (28.3 %), β-caryophyllene (7.8 %), (E)-β-farnesene (7.4 %) and 1,8-cineole (4.8 %) were the major constituents of *M.vulgare* oil, Hamdaoui et al. (2013). From different region of Iran, the main constituants of *M. vulgare* essential tricyclene, β-pinene, bisabolol, β-elemone and isomenthon-8-thiol(Saleh and Glombitz, 1989), bisabolene, β-caryophyllene, and E-β-farnesene (Asadipour et al., 2005), caryophyllene oxide, β-caryophyllene and germacrene D (Khanavi et al., 2005), β-bisabolene, β-caryophyllene, germacrene-D and E-β-farnesene (8.3%) (Mahnaz et al., 2005), γ-eudesmol, germacrene, D-citronellyformate, β-Citronellol, geranyltiglate, geranylformate (Bokaeian et al., 2014).From Libya, carvacrol, E-β-farnesene and thymol (EL-Hawary et

al., 2013).In Algeria, eugenol and β-bisabolene (Belhattab et al., 2006).In Egypt, Salama et al., (2012) reported that thymol and γ-cadineneas major components.Finally, this study indicated that carvacrol, β-phellandrene, carvyl acetate as the main component of the oil of *Marrubium vulgare*. The available literature does not indicate carvacrol and carvyl acetate as the main component of the oil of *Marrubium vulgare*. The results presented from previously studies indicated that the essential oils obtained from *Marrubium vulgare* showed significant variability in their chemical composition depending on location and stages of development as mentioned before (Lawrence, 1981; Tucker andMaciarello, 1990; Mockute et al., 2003).

Table 1. Principal constituents of *Marrubium vulgare* essential oil.

Compound	%	Compound	%
β-thujene	2.93	camphore	0.64
α-Pinene	1.44	borneol	1.12
camphene	0.64	terpinen-4-ol	0.97
sabinene	0.25	2,6-dimethylheptadecane	0.18
1-octen-3-ol	2.48	α -terpineol	0.33
β-pinene	3.53	thymol	0.34
3-octanol	0.16	carvacrol	36.28
α-phellandrene	0.60	trans-caryophyllene	4.06
α-terpinene	3.83	α-humulene	0.19
carvyl acetate	11.52	(+)-carvomenthene	0.30
limonene	0.82	α-cubebene	0.47
1,8-cineol	1.49	7-epi-sesquisabinene hydrate	0.27
β- phellandrene	15.49	caryophyllene oxide	0.67
trans- sabinene hydrate	3.29	cubenol	0.19
linalool	3.86	α-copaene	0.22
cis- sabinene hydrate	0.59	*E*-β-farnesene	0.24

4. Conclusion

It can be concluded that the major constituents of *Marrubium vulgare*L. essential oil grown in Egypt from the aerial parts were carvacrol, β- phellandreneandcarvyl acetate.

References

[1] Adel, K., Zied Z., Ahmed, B., Néji G., Mohamed, D. G. and Radhouane(2011). Chemical composition and antioxidant activity of *Marrubiumvulgare* L. essential oil from Tunisia.African Journal of Biotechnology, 10(19): 3908-3914.

[2] Asadipour, A., Mehrabani, M., Nazeri, V. andM.Tabarraii(2005). Composition of the essential oil of *Marrubiumvulgare*L.Ulum-i-Daroei, 2:77-82.

[3] Belhattab, R., Larous, L., Figueiri, A.andA.Pedro (2006). Essential oil composition and glandular trachoma of *Marrubiumvulgare* L. growing wild in Algeria.J. Essent. Oil Res., 18: 369-373.

[4] Bokaeian, M., Saboori, E., Saeidi, S., Niazi, A.A., Amini-Borojeni, N., Khaje, H.S. Bazi(2014). Phytochemical analysis, antibacterial activity of *Marrubiumvulgare* L. against *Staphylococcus aureus* in vitro.Zahedan Journal of Research in Medical Sciences, 16 (10): 60-64.

[5] Boudjelal, A., Henchiri, C., Siracusa, L., Sari, M.andG.Ruberto(2012). Compositional analysis and in vivo anti-diabetic activity of wild Algerian *Marrubiumvulgare* L. infusion.Fitoterapia, 83(2): 286-292.

[6] De Jesus, R.A.P.,Cechinel-Filho, V., Oliveira, A.E. V. Schlemper(2000). Analysis of the antinociceptive properties of marrubiin isolated from*Marrubiumvulgare.* Phytomedicine, 7: 111-115.

[7] De Oliveira, A.P., Santin, J.R., Lemos, M., Júnior, L.C.K., Couto, A. G., Bittencourt, C.M. D., FilhoV.C. and S. F. Andrade (2011). Gastroprotective activity of methanol extract and marrubiin obtained from leaves of*Marrubiumvulgare*L. *(Lamiaceae).* J. Pharm. Pharmacol., 63: 1230–1237.

[8] De Souza, M.M., De Jesus, R.A.P.,Cechinel-Filho, V. and V.Schlemper(1998). Analgesic profile of hydroalcoholic extract obtained from*Marrubiumvulgare.* Phytomedicine, 5: 103–107.

[9] El Bardai, S., Lyoussi, B., Wibo, M. and N. Morel(2001). Pharmacological evidence of hypotensive activity of*Marrubiumvulgare*and*Foeniculumvulgare* in spontaneously hypertensive rat. Clin. Exp.Hypertens., 23: 329-343.

[10] El Bardai, S., Morel, N.,Wibo, M., Fabre, N.,Labres, G.,Lyoussi, B. and J. Quetin-Leclercq (2003). The vasorelaxant activity of marrubenol and marrubiin from*Marrubiumvulgare.* Planta Med., 69:75-77.

[11] EL-Hawary, S., EL-Shabrawy, A., Ezzat, S. andF. EL-Shibany(2013). Gas chromatography-mass spectrometry analysis, hepatoprotective and antioxidant activities of the essential oils of four Libyan herbs.Journal of Medicinal Plants Research,7(24): 1746-1753.

[12] Firuzi, O., Javidnia,K.,Gholami,M., Soltani,M.R.Miri(2010). Antioxidant activity and total phenolic content of 24 Lamiaceae species growing in Iran.Nat. Pro.Commun, (5):261.

[13] Guenther, G. (1961). The essential oils V.III. Robert E.D. Nastrand Comp. Inc. New York.

[14] Hamdaoui, B., Wannes, W.A., Marrakchi, M., Brahim, N.andB. Marzouk (2013). Essential oil composition of two tunisianhorehound species:*Marrubiumvulgare*L.and*Marrubiumaschersonii*Magnus.Journal of Essential Oil Bearing Plants, 16 (5): 608-612.

[15] Hellen, K., Stulzer, H.K., Tagliari, M. P., Zampirolo, J. A., Cechinel-Filho, V. and V. Schlemper (2006). Antioedematogenic effect of marrubiin obtained from *Marrubium vulgare*. J. Ethnopharmacol., 108: 379–384.

[16] Jorge, V., Francisco, A., Adrián,T.S., Alejandra,A.C., Marisa,E.C., Irene,J.H., Angélica,F.F., Samuel,E.S. and A.R. Ortiz (2012).Acute hypoglycemic effect of ethanolic extracts from*Marrubiumvulgare*. Phytopharmacology, 3(1): 54-60.

[17] Karioti, A.,Skopeliti, M.,Tsitsilonis, O.,Heilmann, J. and H. Skaltsa(2007). Cytotoxicity and immunomodulating characteristics of labdanediterpenes from *Marrubium cylleneum* and *Marrubium velutinum*. Phytochemistry, 68: 1587-1594.

[18] Khanavi, M., Ghasemian, L., HosseinyMotlagh, E., Hadjiakhoondi, A. and A. Shafiee (2005). Chemical composition of the essential oils of *Marrubium parviflorum* Fisch. & C.A. Mey and *Marrubium vulgare* L. from Iran. Flav. Fragr. J., 20: 324-326.

[19] Laonigro, G.,Lanzetta, R.,Parrilli, M.,Adinolfi, M. and L.Mangoni (1979). The configuration of the diterpenespiro ethers from *Marrubium vulgare* and from *Leonotisleonurus*. *Gazz*. Chim. Ital., 109: 145-150.

[20] Mahnaz, K., Ghasmian, L., Motlagh, E., Abbas, H. and S.Abbas (2005). Chemical composition of *Marrubium parviforum* Fisch. & C.A. Mey. and *Marrubium vulgare*L. from Iran. Flavour Frag. J., 20(3): 324-326.

[21] Marrelli, M.,Conforti, F.,Rigano, D., Formisano, C., Bruno, M.,Senatore, F. and F.Menichini(2013). Cytotoxic properties of*Marrubiumglobosum*ssp. libanoticum and its bioactive components. Nat. Prod. Comm., 8: 567–569.

[22] Masoodi, M. H., Ahmed, B., Zargar,I.M., Khan, S.A., Shamshir, K. And P.I.Singh (2008). Antibacterial activity of whole plant extract of *Marrubium vulgare*. African Journal of Biotechnology, 7 (2): 086-087.

[23] Meyre-Silva, C. andV. Cechinel-Filho (2010). A review of the chemical and pharmacological aspects of the genus *Marrubium*. Curr. Pharm. Des., 16: 3503–3518.

[24] Meyre-Silva, C.,Yunes, R.A., Schlemper, V., Campos-Buzzi, F. V. Cechinel-Filho(2005). Analgesicpotential of marrubiin derivatives, a bioactive diterpene present in *Marrubium vulgare* (Lamiaceae).Farmaco, 60: 321-326.

[25] Mnonopi, N.,Levendal, R.A., Davies-Coleman, R.T. and C.L. Frost (2011). The cardioprotective effects of marrubiin, a diterpenoidfound in *Leonotisleonurus* extracts. J. Ethnopharmacol., 138: 67-75.

[26] Mnonopi, N.,Levendal, R.A.,Mzilikezi, N. and C.L.Frost(2012). Marrubiin, a constituent of*Leonotusleonurus,* alleviates diabetic symptoms. Phytomedicine, 19: 488-493.

[27] Nagy, M.andE. Svajdlenka(1998). Comparison of essential oils from*Marrubiumvulgare* L. and *M. peregrinum* L. J.Essent. Oil Res., 10: 585-587.

[28] Piccoli, P. N. and R. Bottini (2008). Accumulation of the labdanediterpenemarrubiin in glandular trichome cells along the ontogeny of *Marrubium vulgare* plants. Plant Growth Regul, (56): 71.

[29] Sahpz,S., Garbacki,N., Tits,M. and F.Bailleul (2002). Isolation and pharmacological activity of phenylpropanoid esters from *Marrubiumvulgare*. J.Ethnopharmacol.,(79):389.

[30] Salama, M.M., Taher, E. E. and M. M.El-Bahy(2012). Molluscicidal and mosquitocidal activities of the essential oils of *Thymus capitatus*HOFF. ET LINK. And *Marrubium vulgare* L. Rev. Inst. Med. Trop. Sao Paulo, 54(5):281-286.

[31] Saleh, M.M.AndK.W.Glombitza(1989).Volatile oil of *Marrubiumvulgare* and its anti-schistosomalactivity. Planta Med., 55: 105-108.

[32] Weel, K.G.C., Venskutonis, P.R., Pukalskas, A., Gruzdiene, D.andJ.P.H. Linssen(1999). Antioxidant activity of horehound *(Marrubiumvulgare*L.*)* grown in Lithuania.Fett/Lipid, 10: 395-400.

[33] Zaabat, N., Hay, A.E.,Michalet, S.,Darbour, N.,Bayet, C.,Skandrani, I.,Chekir-Ghedira, L., Akkal, S. and M.G. Dijoux-Franca (2011). Antioxidant and antigenotoxic properties of compounds isolated from *Marrubiumdeserti* de Noe.J. Food Chem. Toxicol., 49: 3328–3335.

[34] Zarai, Z., Kadri, A., Chobba,I.B., Mansour,R.B., Bekir, A., Mejdoub, H. and N.Gharsallah(2011). The in-vitro evaluation of antibacterial, antifungal and cytotoxic properties of *Marrubiumvulgare* L. essential oil grown in Tunisia. Lipids in Health and Disease, 10: 161.

[35] Zawiślak, G. (2012). Chemical composition of essential oils of *Marrubiumvulgare* L. and *Marrubiumincanum*Desr. grown in Poland.Chemija,23. (2): 136-140.

[36] Zerbe, P., Chiang, A., Dullat, H., O'Neil-Johnson, M., Starks, C., Hamberger, B. and J.Bohlmann(2014). Diterpene synthases of the biosynthetic system of medicinally active diterpenoids in *Marrubiumvulgare*.The Plant Journal, 79: 914-927.

[37] Zhang,J., Liu,A.,Hou,R., Zhang, J., Jia,X.,Jian,W. and J. Chen(2009).Salidroside protects cardiomycte against hypoxia-induced death: AHIF-1 α-activated and VEGF-mediated pathway.Eur.J.Pharmacol.,607 (1): 6-14.