Asian Journal of Chemistry; Vol. 25, No. 8 (2013), 4361-4363

ASIAN JOURNAL OF CHEMISTRY

http://dx.doi.org/10.14233/ajchem.2013.13977

Chemical Composition of Essential Oils from Flower and Leaf of Korean Mint, *Agastache rugosa*

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(*Received*: 27 April 2012; *Accepted*: 11 February 2013) AJC-12964

This study aims at the qualitative and quantitative analysis of the essential oils extracted from the leaf and flower of *Agastache rugosa*. GC-MS analysis confirmed the presence of 43 compounds in the essential oils, 34 of which were common both in the leaf and in the flower. Seven compounds 2,6-dimethylheptane, 4-methyl-1-pentene-3-ol, δ-limonene, *trans*-ocimene, octenyl acetate, anethole and 2-methyl-5-isopropenyl-2-cyclohexenone were present only in the flower, while β-damascenone and allyl-3-methyl-2-butanoate were present only in the leaf. The leaf and flower extracts of *Agastache rugosa* contained 36 and 41 compounds that constituted 90.93 and 82.3 % of the oils, respectively. Methylchavicol was the predominant component both in the leaf (84.25 %) and in the flower (57.94 %); the other major components in the flower were anethole, L-limonene and *trans*-caryophyllene, while those in the leaf were 2-phenyl propionaldehyde and *trans*-caryophyllene.

Key Words: Korean mint, Flower, Leaf, GC-MS analysis, Essential oil, Methylchavicol.

INTRODUCTION

Korean mint, *Agastache rugosa* Kuntze belongs to the family Labiatae, is a perennial herb widely distributed in East and Southeast Asian countries. *Agastache rugosa* has been used in Chinese traditional medicine for the treatment of cholera, emesis and miasma and has been reported to have antitumor, antifungal, HIV integrase inhibitory and cytotoxic activities[1-7]. In addition, the leaves and flowers of *A. rugosa* are used as spices in fish-based foods and as a source of honey, respectively[8].

Estragole (*p*-allylanisole, methyl chavicol), a component of the essential oil extracted from *A. rugosa*, is used as a sweet condiment as well as in perfumes and salad dressings. Moreover, estragole is an important constituent of root beer and can be used to prepare other flavoring agents such as anethole and anisaldehyde. In Korea, the essential oil from *A. rugosa* has traditionally been used for medicinal purposes[8]. In addition, estragole is a major component (56-94 %) of the leaf of natural populations of *A. rugosa* grown in Ames, Iowa[9,10] and Ishikawa, Hyogo Prefecture, Japan[11,12]. In contrast, a population of *A. rugosa* (*A. rugosa* O. Kuntze var. *methyleugenolifera*) collected from Kitami, Hokkaido, Japan, has been found to contain 84-92 % methyl eugenol and only 2-6 % estragole[12]. Chemotypes of the essential oil components in individual *A. rugosa* samples collected from Scotland and the USA have been analyzed[13-15], but those in the case of Korean *A. rugosa* samples have not been analyzed in detail. The essential oil contents of the leaves and inflorescences of Korean *A. rugosa* are 0.29 and 0.38 % (in dry weight), respectively, with estragole constituting *ca.* 80 % of these essential oils[16,17]. Moreover, there is no change in the essential oil composition between the samples obtained during the vegetative and full- bloom stages[1]. In this study, we compared the characteristics of the volatile components in the flower and leaf of *A. rugosa*.

EXPERIMENTAL

Agastache rugosa plants were field-grown on the College Experimental Farm, Chungnam National University, Daejeon and their flowers and leaves were harvested at the flowering stage in August 2010. Fresh samples were stored frozen in sealed clear polyethylene plastic bags at -80 ºC until they were used. Collected samples were freeze dried at -80 ºC for at least 72 h.

Extraction of the essential oils: The freeze-dried flowers and leaves of *A. rugosa* (40 g) were subjected to simultaneous

steam distillation-extraction (SDE) for 3 h in a modified Likens-Nickerson apparatus, using a 1:1 *n*-pentane/diethyl ether mixture as the solvent. The obtained essential oil was dried over anhydrous sodium sulfate, filtered and stored at -20 ºC until further analysis.

GC and GC-MS analysis: GC-MS analysis was performed using a Finnigan Focus-polarisQ system (Thermo Fisher, USA) equipped with a DB-5MS column (60 m length × 0.32 mm inner diameter × 0.25 μm film thickness, J&W Scientific, Folsom, CA). Helium was used as the carrier gas at a constant flow rate of 1.0 mL/min. One microliter of the extract was injected in the splitless mode. The oven temperature was held at 40 ºC for 7 min increased to 240 ºC at the rate of 5 ºC/min and held at this temperature for 15 min. The injector and detector temperatures were 220 and 270 ºC, respectively. The mass detector was operated in the electron impact mode at an ionization energy of 70 eV, in the scan range 10-650 amu. The conditions for GC analysis (Agilent 6890 Series GC system; Agilent Technologies, PA, USA) were the same as those for GC-MS, except that the flame ionization detector (FID) was used in the former case. The relative proportions (in percentage) of the essential oil constituents were obtained by normalization of the FID peak area.

RESULTS AND DISCUSSION

A total of 43 compounds, 34 of which were common in the leaf and flower, were identified in the essential oils extracted from *A. rugosa*. Thirty-six and 41 components (constituting 90.93 and 82.3 % of the essential oils) were identified in the leaf and flower samples, respectively (Table-1,

TABLE-1
VOLATILE COMPOSITION OF ESSENTIAL OILS FROM THE LEAF AND FLOWER OF *Agastache rugosa*

No.	Retention time (min)	Identification			Leaf	Flower
		Chemical	m.f.	m.w.	Area (%)	
1	9.86	2-Ethyl-2,5-dimethylpentane	C_9H_{20}	128	0.35	0.55
2	10.80	5-Methyl-2-hexanol	$C_7H_{16}O$	116	0.10	0.14
3	11.04	2-Heptanol	$C_7H_{16}O$	116	0.07	0.1
4	12.88	2,3-Dimethyl-3-methoxybutan-1-ol	$C_7H_{16}O_2$	132	0.06	0.03
5	13.94	Isobutyric acid	$C_4H_8O_2$	88	0.29	0.18
6	16.74	2,6-Dimethylheptane	C_9H_20	128	–	0.02
7	18.92	2,3,3-Trimethylpentane	C_8H1_8	114	0.06	0.08
8	19.19	4-Methyl-1-pentene-3-ol	$C_6H_{12}O$	100	–	0.03
9	20.44	3-Methyl-3-pentanol	$C_6H_{14}O$	102	0.01	0.01
10	20.76	1-Octen-3-ol	$C_8H_{16}O$	128	0.27	0.27
11	21.05	3-Octanone	$C_8H_{16}O$	128	0.15	0.09
12	21.27	Myrcene	$C_{10}H_{16}$	136	0.01	0.03
13	22.27	δ-Limonene	$C_{10}H_{16}$	136	–	0.72
14	23.45	L-Limonene	$C_{10}H_{16}$	136	0.51	2.94
15	26.59	*trans*-Ocimene	$C_{10}H_{16}$	136	–	0.01
16	27.02	Octenyl acetate	$C_{10}H_{18}O$	170	–	0.04
17	27.18	Linalool	$C_{10}H_{18}O$	154	0.01	0.08
18	27.55	1-Octen-3-yl acetate	$C_{10}H_{18}O_2$	170	0.43	0.25
19	**31.93**	**Anethole**	$C_{10}H_{12}O$	148	–	14.9
20	**32.38**	**Methyl chavicol**	**$C_{10}H_{12}O$**	**148**	**84.25**	**57.94**
21	34.18	*cis*-Salvene	C_9H_{16}	124	–	–
22	34.66	2-Phenyl propionaldehyde	$C_9H_{10}O$	134	1.28	0.12
23	35.64	2-Methyl-5-isopropenyl-2-cyclohexenone	$C_{10}H_{14}O$	150	–	0.27
24	36.41	2-Ethenylbicyclo[2.1.1]hex-2-ene	C_8H_{10}	106	0.02	0.01
25	37.00	2,5-Dimethyl-5-hexen-3-ol	$C_8H_{16}O$	128	0.01	0.01
26	38.55	Santolina triene	$C_{10}H_{16}$	136	0.06	0.02
27	40.05	2-Methylbenzenemethanol	$C_8H_{10}O$	122	0.23	0.03
28	40.60	α-Damascenone	$C_{13}H_{18}O$	190	0.10	–
29	40.90	*cis*-Epoxy-ocimene	$C_{10}H_{16}O$	152	0.09	0.13
30	41.10	7-(1-(E)-prop-1-enyl)cycloocta-1,4-diene	$C_{11}H_{16}$	148	0.04	0.04
31	41.43	2-Benzene-2-butylethanol	$C_{12}H_{18}O$	178	0.08	0.02
32	41.92	1,2-Diethylbenzene	$C_{10}H_{14}O$	134	0.02	0.01
33	42.49	*trans*-Caryophyllene	$C_{15}H_{24}$	204	1.00	2.05
34	45.08	1.8-Nonadiyne	C_9H_{12}	120	0.44	0.5
35	45.67	3,7-Dimethyl-(E)-1,3,6-octatriene	$C_{10}H_{16}$	136	0.47	0.25
36	46.20	Allyl-3-methyl-2-butanoate	$C_8H_{12}O_2$	140	0.02	–
37	46.53	2-Methyl-5,7-dimethylene-1,8-nonadiene	$C_{12}H_{18}$	162	0.08	0.05
38	48.94	3(Z),6(Z),8(E)-Dodecatriene-1-ol	$C_{12}H_{20}O$	180	0.08	0.04
39	49.28	4-Tridecen-6-yne	$C_{13}H_{22}$	178	0.06	0.09
40	49.69	3,7,11-Trimethyl-1,6,10-dodecatrien-3-ol	$C_{15}H_{26}O$	222	0.01	0.01
41	51.43	(-)-Elema-1,3,11(13)-*trien*-12-ol	$C_{15}H_{24}O$	220	0.01	0.01
42	51.52	2-Phene-10-hydroperoxide	$C_{10}H_{16}O_1$	168	0.07	0.03
43	51.96	*trans*-Limonene oxide	$C_{10}H_{16}O$	152	0.14	0.07
		Total (%)			**90.93**	**82.3**

Fig. 1). Seven of the 43 compounds-2,6-dimethylheptane, 4-methyl-1-pentene-3-ol, δ-limonene, *trans*-ocimene, octenyl acetate, anethole and 2-methyl-5-isopropenyl-2-cyclohexenone were present only in the flower samples, while β-damascenone and allyl-3-methyl-2-butanoate were present only in the leaf.

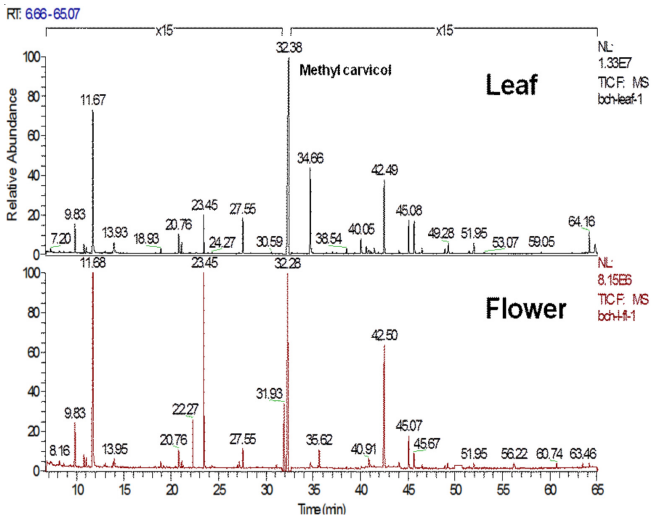

Fig. 1. GC-MS chromatogram of the essential oils from leaf and flower of *Agastache rugosa*

The most predominant component in the leaf and flower extracts was methylchavicol (84.25 and 57.94 %, respectively). The other prominent compounds in the flower were anethole (14.9 %), L-limonene (2.94 %), *trans*-caryophyllene (2.05 %), δ-limonene (0.72 %) and 2-ethyl-2,5-dimethylpentane (0.55 %), while those in the leaf were 2-phenyl propionaldehyde (1.28 %) and *trans*-caryophyllene (1 %). From these results, it was apparent that the methyl-chavicol content of the leaf of *A. rugosa* was 1.5 times that of the flower. Among the seven previously mentioned compounds present in the flower, anethole (14.9 %) and δ-limonene (0.72 %) were the most prominent. The 2-phenyl propionaldehyde content of the leaf was 10.7 times that of the flower. On the other hand, L-limonene, *trans*-caryophyllene and 2-ethyl-2,5-dimethylpentane contents of the flower were 5.8, 2.1 and 1.6 times those of the leaf, respectively. Previously some works refer to quantitive and quantitative determination[18,19] (volatile constituents, variation of estragole) from *A. rugosa*, they found difference among the cultivars collected from various territories. In this study, we also found a difference among the leaf and flower that agree with the results of previous study[20,21].

Conclusion

The essential oil content varied widely between the leaf and flower parts of *A. rugosa*. Compounds such as methyl-chavicol and 2-phenylpropionaldehyde were present in large amounts in the leaf, while L-limonene, *trans*-caryophyllene and 2-ethyl-2,5-dimethylpentane were more predominant in the flower. Hence, once the constituents of the essential oils are known, it is possible to choose the appropriate part of the plant the leaf or the flower and prepare essential oils that are rich in the desired component(s).

ACKNOWLEDGEMENTS

This work was supported by Priority Research Centers Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Education, Science and Technology (2010-0029642).

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