RESEARCH REPORT *J. Essent. Oil Res.*, **7**, 49-52 (Jan/Feb 1995)

Comparisons of the Volatile Leaf Oils of *Juniperus rigida* Mig. from Northeastern China, Korea and Japan

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Received: July 1994

ABSTRACT: The composition of the leaf oils of *Juniperus rigida* from northeast China, Japan and Korea are reported. The Japanese samples were very similar to previous literature reports being rich in bornyl acetate (40.5-59.0%) and α-pinene (13.5-15.4%), but the samples from northeast China and Korea showed geographical differentiation in the production of increased amounts of α-pinene (39.7% and 53.5% respectively) and reduced amounts of bornyl acetate (1.3% and 2.8% respectively).

KEY WORD INDEX: *Juniperus rigida*, Cupressaceae, essential oil composition, bornyl acetate, α-pinene, geographical variation.

INTRODUCTION: *Juniperus rigida* Mig. grows in northeast China, Korea and Japan. It is a small tree with a strong central axis and is known as Du-song in China and as the Japanese juniper in Japan. The leaf oil of *J. rigida* has only been reported from Japan (1-3), where it contains over 50% bornyl acetate. The wood oil of *J. rigida* from Japan has been analyzed in considerable detail (1). Fretz et al. (4) reported on the monoterpenes from a cultivated putative plant of *J. rigida.*

Due to the widespread cultivation of this species in botanic gardens, an opportunity became available to examine the leaf oil from *J. rigida* originated from China, Japan and Korea. A comparison of the leaf oils is presented in this paper.

EXPERIMENTAL: Foliage was collected and voucher specimens [Heibei Provice, N.E. China, (via Beijing Botanic Garden), R. P. Adams, 6797-6799; Korea (via Kew Gardens), R. Adams, 5636; Japan (via Arnold Arboretum), R. P. Adams, 5515-5517] are deposited at BAYLU! and NWTU! The leaf oils were isolated by steam distillation (200 g foliage, FW)

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1041-2905/95/0001-0049$04.00/0

Table I. Comparisons of the chemical composition of the leaf oils of *Juniperus rigida* from Japan (literature, reference 1), Japan, Korea and Northeast China

RT	Compound	Percent total oil			
		Japan (1)	Japan	N.E. China	Korea
215	(E)-2-hexenal	-	1.7	2.4	1.3
301	tricyclene	0.9	0.5	0.2	0.1
307	α-thujene	-	-	t	t
319	α-pinene	13.5	15.4	39.7	53.3
337	α-fenchene	t	t	0.6	t
340	camphene	1.3	1.0	1.0	0.6
347	thuja-2,4(10)-diene	-	t	t	0.3
363	verbenene (=pinadiene)	-	t	0.1	t
379	sabinene	0.3	0.1	t	0.2
386	β-pinene	1.2	1.7	1.9	4.7
408	myrcene	3.1	6.2	11.2	4.8
427	δ-2-carene	-	0.2	0.8	1.0
435	α-phellandrene	0.6	0.3	1.0	1.2
444	δ-3-carene	0.3	t	t	t
457	α-terpinene	0.1	t	t	t
471	p-cymene	0.1	0.4	0.6	0.9
481	limonene	4.4	4.1	4.2	3.4
482	β-phellandrene	-	2.0	2.1	1.7
498	(Z)-β-ocimene	-	-	t	-
519	(E)-β-ocimene	-	0.5	t	-
535	pentyl isobutyrate	-	0.1	0.6	-
545	γ-terpinene	0.4	0.2	0.2	-
560	cis-sabinene hydrate	-	t	-	t
605	fenchone	0.1	t	1.6	t
608	terpinolene	0.5	0.7	0.8	0.3
614	2-nonanone	-	-	t	-
629	trans-sabinene hydrate	-	t	-	-
632	linalool	0.3	1.1	0.6	
664	endo-fenchol	-	0.7	0.8	0.4
667	cis-thujone	0.1	-	-	-
692	α-campholenal	-	0.2	0.2	0.8
724	trans-pinocarveol	-	0.2	0.2	0.5
727	cis-verbenol	-	t	t	0.2
734	camphor	1.9	1.7	0.6	1.8
735	trans-verbenol	-	-	t	-
746	camphene hydrate	-	0.7	0.2	0.2
764	isoborneol	0.4	t	t	t
781	pinocarvone	-	t	-	0.4
789	borneol	1.3	4.1	0.5	1.1
809	isopinocamphone	-	t	t	t
820	terpin-4-ol	1.2	0.7	0.6	0.3
826	naphthalene	-	t	t	0.2
837	p-cymen-8-ol	-	t	t	t
852	α-terpineol	0.7	0.5	1.1	0.3
867	myrtenol	-	0.2	t	0.8
894	verbenone	-	0.1	t	0.5
923	trans-carveol	-	0.1	t	0.2
930	endo-fenchyl acetate	0.4	1.2	0.6	0.4
950	citronellol	-	0.7	0.2	t
1011	piperitone	-	t	t	0.6
1018	geraniol	0.2	-	-	-
1035	unknown	-	0.2	0.8	t
1094	(E)-anethole	-	t	t	-
1099	bornyl acetate	59.0	40.5	1.3	2.8

Table I. (Cont.)

RT	Compound	Percent total oil			
		Japan (1)	Japan	N.E. China	Korea
1116	2-undecanone	-	0.2	4.8	-
1118	(E,Z)-2,4-decadienal	-	-	-	t
1176	(E,E)-2,4-decadienal	-	-	-	t
1197	methyl geranate	-	t	-	-
1264	α-terpinyl acetate	-	0.2	-	-
1267	α-longipinene	-	-	-	t
1275	citronellyl acetate	-	0.2	t	-
1303	neryl acetate	0.1	-	-	-
1352	geranyl acetate	0.2	0.2	-	t
1355	β-bourbonene	-	-	0.2	-
1442	β-caryophyllene	1.3	0.5	1.6	2.5
1527	α-humulene	0.9	0.3	1.3	1.7
1537	(E)-β-farnesene	-	0.3	0.4	t
1571	sesquiterpene	-	-	1.3	-
1585	γ-himachalene	-	t	-	-
1586	γ-muurolene	-	-	t	-
1594	germacrene D	0.6	0.9	2.3	t
1629	epi-cubebol	-	t	-	-
1632	bicyclogermacrene	-	0.2	-	-
1634	(E)-methyl isoeugenol	-	-	0.7	-
1643	α-muurolene	-	-	t	-
1653	germacrene A	-	t	t	-
1664	α-farnesene*	-	0.3	0.7	t
1676	γ-cadinene	-	0.3	0.6	-
1700	δ-cadinene	0.1	0.2	0.1	-
1777	germacrene B	-	0.1	0.2	1.0
1790	geranyl butyrate	-	-	0.3	-
1796	(E)-nerolidol	0.4	0.6	1.0	0.5
1821	germacrene D-4-ol	-	t	-	-
1825	spathulenol	-	0.4	0.3	-
1837	caryophyllene oxide	0.1	0.4	0.6	2.5
1876	cedrol	-	-	0.3	-
1897	humulene epoxide II	-	0.2	0.5	1.4
1951	γ-eudesmol	-	t	-	-
1973	epi-α-cadinol (=T-cadinol)	-	0.2	0.6	-
1976	epi-α-muurolol (=T-muurolol)	-	0.2	-	-
1993	β-eudesmol	-	t	-	t
2000	α-eudesmol	-	t	-	-
2003	α-cadinol	0.1	0.9	0.3	-
2053	sesquiterpene alcohol	-	0.7	-	-
2079	epi-α-bisabolol	-	-	0.7	-
2141	(Z,Z)-farnesol	-	t	t	t
2159	(E,E)-farnesol	2.0	1.8	1.4	0.3
2201	(E,Z)-farnesol	-	t	t	0.2
2841	abietatriene	-	0.4	0.5	0.5
2891	abietadiene	-	0.1	t	t
3296	trans-abietal	-	t	-	-
3297	trans-totarol	-	-	t	-
3333	trans-ferruginol	-	0.1	t	t

Compounds are listed in order of their elution from a DB5 column
* correct isomer not identified
t = trace (less than 0.1%)
– = unidentified components less than 0.05%

in a modified Clevenger apparatus (5) for 2 h. The oil samples were concentrated (diethyl ether trap removed when collected in a Clevenger unit) with nitrogen and stored at –20°C until analyzed. Mass spectra were recorded with a Finnigan Ion Trap (ITD) mass spectrometer, model 800, directly coupled to a Varian 6500 gas chromatograph, using a J&W DB5, 0.26 mm x 30 m, 0.25 micron coating thickness, fused silica capillary column (see reference 6 for operating details). Identifications were made by library searches of our volatile oil library, LIBR(TP) (6) using the Finnigan library search routines based on fit. Additional searches were made of the EPA/NIH mass spectral data base (7).

RESULTS AND DISCUSSION: Oil yields (2 h) were 0.11% (Japan), 0.30% (Korea), and 0.77% (N.E. China) (oil wt/extracted, oven dried foliage weight). The oil of the literature report from Japan (1) was very similar (Table I) to our *J. rigida* sample from Japan (but cultivated at the Arnold Arboretum). Both oils contained large amounts of bornyl acetate (59.0%, 40.5%) and moderate amounts of α-pinene (13.5%, 15.4%). The differences are mostly in the presence of small quantities that may have been unidentified in the previous report (1). However, on the mainland of Asia, the oils from northeast China and Korea are considerably different from the Japanses oils. Oils from northeast China (Heibei province) and Korea are quite low in bornyl acetate (1.3%, 2.8%) and dominated by α-pinene (39.7%, 53.3%). Otherwise, the oil from northeastern China is more similar to the oil from Japan than that from Korea. This is particularly noticeable in the sesquiterpenes (Table I). None of the samples had high amounts of sabinene as reported by Fretz et al. (4), which brings into question the identity of their material.

It appears that the isolation of *J. rigida* in Japan from the mainland has resulted in some divergence from *J. rigida* on the mainland and this is reflected in the leaf oils. Mass spectra for unidentified constituents: [ITMS, m/z (rel. int.): RT1035, 41(76), 53(12), 68(100), 81(1), 91(1), 99(20), 117(1); RT 1159, 41(100), 55(20), 71(47), 81(32), 95(72), 109(33), 121(36), 136(38), 154(9), 161(2), sesquiterpene; RT2076, 41(100), 55(28), 67(27), 79(40), 91(45), 109(32), 117(19), 131(18), 145(10), 159(28), 177(8), 187(3), 202(10), 220(3), sesquiterpene alcohol.

ACKNOWLEDGMENTS: Authors are grateful to Guo Shou Jun and Yao Jan for field and lab assistance. This research was supported by funds from US NSF grant INT8901632 and a Gansu Provincial Education Community Project.

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