

Investigation of *Juniperus* Species of the United States for New Sources of Cedarwood Oil¹

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The 11 taxa of Juniperus with widespread distribution in the United States and with significant biomass production were investigated to determine their yields of cedarwood oil and the major components of commercial interest: alpha-cedrene, beta-cedrene, thujopsene, cuparene, cedrol, and widdrol. Taxa examined were J. ashei, J. californica, J. erythrocarpa, J. deppeana, J. monosperma, J. occidentalis var. occidentalis, J. o. var. australis, J. osteosperma, J. pinchotii, J. scopulorum, and J. virginiana. The volatile heartwood oils were removed by steam distillation and analyzed by gas chromatography-mass spectroscopy to determine their yields and composition. Cedarwood oil yields in J. erythrocarpa and J. scopulorum were comparable to those of the two species currently being utilized (J. ashei, J. virginiana).

Cedarwood oil is an important natural product for sources of components either to be used directly in fragrance compounding or as a source of raw components in the production of additional fragrance compounds. The oil is used to scent soaps, technical preparations, room sprays, disinfectants, and similar products; as a clearing agent for microscope sections; and with immersion lenses (Guenther 1952). The price varies but has generally been about \$3.50/lb. (Chem. Mkt. Rptr., 27 Jan 1986, \$3.50/lb, Texas oil; \$3.70/lb, Virginia oil) for the past several years. The domestic sources of cedarwood oil for the United States are central Texas (*J. ashei*, "Texas cedarwood oil") and eastern U.S. (*J. virginiana* L., "Virginia cedarwood oil"). *Juniperus ashei* occurs on thousands of acres in central Texas. Although extensive native forests of *J. virginiana* do not occur, young forests are forming as abandoned fields are invaded by redcedars. Birds carry the seeds, and an abandoned field can be infested in only a few years. These old fields will be an important future source of cedarwood.

Cedarwood oils (*Juniperus* species) of the western hemisphere have not been examined thoroughly or systematically. Many of the analyses of cedarwood oil were done by Runeberg (1960a-e, 1961) and associates (Pettersson and Runeberg 1961; Pilo and Runeberg 1960) (Table 1). Of the 16 taxa occurring in the United States, seven have been analyzed to some extent. For many years *J. ashei* was reported to contain only alpha-cedrene and cedrol (Guenther 1952; erroneously referred to as *J. mexicana* in Erdtman and Norin 1966). However, more recently, Kitchens et al. (1971) reported beta-cedrene, thujopsene, widdrol, pseudo cedrol, beta-chamigrene, prim cedrol, widdrene, isowiddrene, alpha-chamigrene, and cuparene (three isomers). Guenther (1952) mentioned that the Texas cedarwood oil industry began in 1929 in Rock Springs, Texas. Bomberger and Baker (1941) noted that "cedarwax" was often precipitated from Texas cedarwood oil and that

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TABLE 1. LITERATURE REPORTS ON COMPOSITION OF VOLATILE WOOD OILS OF *JUNIPERUS* SPECIES.^a

	ACDR	BCDR	THJP	CPRN	CDRL	WDDL	HNKA	WDAZ	Reference
<i>J. ashei</i> * (= <i>J. mexicana</i> in part)	+	+	+	+	+	+			Guenther 1952
									Windemuth, 1945; Kitchens et al. 1971
<i>J. californica</i> *	2.6		26.0	1.0	32.0 ^{52.0}	0.2	7.8	5.2	Pettersson and Runeberg 1961
<i>J. cedrus</i>			82.4	3.7	2.2	2.6			Runeberg 1960a
<i>J. chinensis</i>			11.6	4.3	72.9	6.0	0.05	2.6	Pilo and Runeberg 1960
<i>J. communis</i> *			37.0	3.0	2.0	1.0			Bredenberg 1961
<i>J. conferta</i>			+	+	+				Doi and Shibuya 1972
<i>J. excelsa</i>					+				Rutowski and Vinogradova 1927
<i>J. foetidissima</i>	58.3				8.3	5.0			Runeberg 1961
<i>J. horizontalis</i> *	+		+	+	+	+			Narasimhachari and von Rudloff 1961
<i>J. occidentalis</i> *	+				+				Kurth and Lackey 1948
<i>J. osteosperma</i> * (= <i>J. utahensis</i>)	12.7		47.8	12.5		13.5	3.4	0.1	Runeberg 1960b
<i>J. phoenicea</i>			79.3	2.9	7.2	0.1	1.4	3.6	Runeberg 1960c
<i>J. procera</i>	41.8			2.5	41.8				Pettersson and Runeberg 1961
<i>J. recurva</i>	3.5	0.9	5.1	1.8	49.0	16.7			Oda et al. 1977
<i>J. semiglobosa</i>		+			+				Goryaev et al. 1962
<i>J. thurifera</i>	23.3		15.5	3.9	27.1		7.8	2.7	Runeberg 1960d
<i>J. virginiana</i> *	35.0		30.0	2.0	4.0	2.0			Runeberg 1960e

^a Species marked with an asterisk are native to the United States. Approximate percent concentration of key components was obtained when possible from the original reference. ACDR = alpha-cedrene; BCDR = beta-cedrene; THJP = thujopsene; CPRN = cuparene; CDRL = cedrol; WDDL = widdrol; HNKA = hinokiic acid; WDAZ = widdringtonia acid II.

the cedarwax was actually crystalline cedrol. In 1941 there was no market for cedrol and it was treated as a waste product (Bomberger and Baker 1941). This is an interesting point in that Texas cedarwood oil currently sells for \$3.50/lb, whereas cedrol sells for \$5.25/lb (Chem. Mktr. Rptr., 17 Jan 1986).

Juniperus californica was reported (Pettersson and Runeberg 1961) to have cedrol as the major component (52%) of the heartwood volatile oils (Table 1) with a considerable amount of thujopsene (26%). *Juniperus communis*, a small shrub in the United States, was reported to have mostly thujopsene (37%); however, it is not clear how the author (Bredenberg 1961) arrived at these percentages. *Juniperus horizontalis* is a prostrate plant that forms mats. Due to its low wood biomass, its oil composition is primarily of academic interest. Narasimhachari and von Rudloff (1961) reported that *J. horizontalis* contained alpha-cedrene, thujopsene, cuparene, cedrol, and widdrol but relative concentrations were not reported (Table 1). *Juniperus occidentalis* was examined by Kurth and Lackey (1948) who merely reported that the wood contained alpha-cedrene and cedrol. *Juniperus osteosperma* (referred to as *J. utahensis* by Runeberg [1960b]) had 47.8% thujopsene, with about equal amounts of alpha-cedrene, cuparene, and widdrol (Table 1). *Juniperus virginiana* wood was not directly analyzed by Runeberg (1960e). Using a commercial sample of cedarwood oil said to be from *J. virginiana*, he found mostly alpha-cedrene and thujopsene with a very small amount of cedrol (4%) (Table 1). However, the commercial cedarwood oil may have been precipitated or fractionally distilled to remove cedrol (see results below). Wenninger et al. (1967) analyzed the sesquiterpene hydrocarbons of American cedarwood oil (*J. virginiana*?) and reported that the oil contained 55–65% sesquiterpene hydrocarbons with alpha-cedrene and thujopsene as the major components. Runeberg (1960e) stated that the highest yield of oil, about 3.5% of the wood (dry wt.), was obtained from sawmill waste from older trees (i.e., trees with a greater ratio of heartwood to sapwood). Guenther (1952) obtained only a 0.2% yield by distilling sapwood of *J. virginiana*; he noted that young trees (commonly called "sap cedars") yielded less than 1% oil, compared with older trees (commonly called "virgin cedars"), which yielded 3.5%.

In many parts of the United States weedy junipers have invaded abandoned fields and overgrazed rangelands. They often occur in almost continuous stands for hundred of miles. The most important of these species are *J. ashei* Buch., *J. californica* Carr., *J. erythrocarpa* Cory, *J. deppeana* Steud., *J. monosperma* (Engelm.) Sarg., *J. occidentalis* Hook., *J. osteosperma* (Torr.) Little, *J. pinchotii* Sudw., and *J. virginiana* L. These have occupied millions of acres. An estimated 21.5 million acres of juniper-invaded grasslands (Smith and Rechenthin 1964) are in Texas alone. Ranchers are paid by the U.S. Department of Agriculture for juniper removal to improve range conditions. The opening of the canopy appears to be very important for forage production (Clary 1974). Cropping machinery is currently being used in the Texas cedarwood oil production plant (PAKS Corporation) at Junction, Texas, to harvest *J. ashei*. The trees are cut, chipped, and trucked to distillation units at Junction where the wood oils are steam distilled; the oils are shipped to International Flavors and Fragrances Corporation (New York) for processing. The fuel for distillation is a portion of the spent chips.

The purposes of my study were to determine yields and composition of commercially important components of the cedarwood oils of some United States

juniper species and to evaluate the commercial potential of plants now considered noxious trees in rangelands. The following United States taxa were not analyzed: *J. communis* L., low in the production of wood biomass; *J. deppeana* f. *sperryi* (Correll) Adams (only three trees known in the United States); *J. flaccida* Schlecht. (rare in the United States, occurring only in Big Bend National Park); *J. horizontalis* Moench (prostrate habit, small amount of biomass); and *J. virginiana* var. *silvicola* (Small) Silba (found in coastal foredunes, limited distribution).

MATERIALS AND METHODS

Samples of wood and herbarium vouchers were collected from *J. ashei* (Adams 5007–5009, 9 km W of Ozona, Crockett Co., TX; Adams 5010–5016, 2 km E of Junction, Kimble Co., TX); *J. californica* 'A' (Adams 5067–5071, 13 km NE of I-40, Granite Mtns., San Bernardino Co., CA) and *J. californica* 'B' (Adams 5072–5076, 30 km SE of Yucca, Yuma Co., AZ) ('A' and 'B' refer to the two chemical races discovered by Vasek and Scora (1967) and reconfirmed by Adams, von Rudloff, and Hogge (1983) using leaf volatile oils); *J. erythrocarpa* (Adams 4987–4996, 32 km N of Alpine, Jeff Davis Co., TX); *J. deppeana* (Adams 4974–4983, 32 km NW of Ft. Davis, Jeff Davis Co., TX); *J. monosperma* (Adams 5027–5036, 2 km W of Santa Rosa, Guadalupe Co., NM); *J. occidentalis* (Adams 5077–5086, 8 km W of Juntura, Malheur Co., OR); *J. occidentalis* var. *australis* (Adams 5057–5066, 2 km W of Sonora Jct., Mono Co., CA); *J. osteosperma* (Adams 5047–5056, 25 km E of Monticello, San Juan Co., UT); *J. pinchotii* (Adams 4997–5001, 28 km E of Ft. Stockton, Pecos Co., TX; Adams 5002–5006, 10 km W of Sheffield, Pecos Co., TX); *J. scopulorum* (Adams 5037–5046, 5 km E of Clines Corner, Tarrant Co., NM); and *J. virginiana* (Adams 5017–5025, 7 km W of Bastrop, Bastrop Co., TX). Voucher specimens are deposited at the Science Research Center–Utah (SRCG).

The samples consisted of wood (20 cm long × 5–10 cm in diameter) and leaves (400 gm). All samples were kept cool (February collections) in the field and then frozen in the lab until analyzed.

The wood samples were separated into heartwood and bark/sapwood; each subsample was then kept separate. Portions of the heartwood, bark/sapwood, and leaves were dried (48 hr, 100°C) to determine the percent moisture. About 12 gm of the heartwood were steam distilled (20 hr) to remove the volatile oil (Adams 1975a). Percent yield was calculated as: $100 \times \text{oil wt.}/(\text{corrected dry wt. of wood distilled} + \text{oil wt.})$.

The volatile oils were subjected to fused silica capillary GLC and GCMS to identify the major components (Adams et al. 1979). The principal components of commercial interest are alpha-cedrene, beta-cedrene, thujopsene, cedrol, and widdrol.

RESULTS AND DISCUSSION

The volatile heartwood oil yields varied from 0.21% dry wt. (*J. pinchotii*) to 4.87% (*J. erythrocarpa*) (Table 2). The two species used commercially, *J. ashei* and *J. virginiana*, yielded 4.04% and 3.18%, respectively. In general, junipers from arid lands (*J. californica*, *J. osteosperma*, *J. monosperma*, and *J. pinchotii*) yielded less oil than some from more mesic areas (*J. scopulorum* and *J. virginiana*), which had fairly high yields. In contrast, *J. erythrocarpa*, of very arid habitats, had the highest yield. The observation of Guenther (1952) that young trees of *J. virginiana* yielded much less oil than older (mature) trees was confirmed. Yields from sapwood were less than 0.5% (Adams, unpublished), whereas the yield from heartwood was 3.18% dry wt. (Table 2).

Since differentiation in morphology and leaf oils of *J. ashei* has been reported (Adams 1975b, 1977) between Ozona and Junction, Texas, analysis was made on heartwood from both areas. The yield of steam distillable components was 4.92% dry wt. from Ozona versus 4.04% from Junction. Personnel at PAKS Corporation (Junction), pointed out trees that would be higher yielding: those with a strong central axis, older trees, and slower growing trees. They commented

TABLE 2. OIL YIELDS FROM *JUNIPERUS* HEARTWOOD (DRY WT. BASIS) AND PERCENTAGE COMPOSITION OF THE MAJOR COMMERCIALY IMPORTANT COMPONENTS (ALPHA-CEDRENE, BETA-CEDRENE, THUJOPSENE, CUPARENE, CEDROL, AND WIDDROL), THE SUM OF THESE COMPONENTS, AND THE RELATIVE % YIELD OF THE COMPONENTS (SUM \times PERCENT YIELD).^a

Taxon	% oil	ACDR	BCDR	THJP	CPRN	CDRL	WDDL	Sum	Rel % yield
<i>J. ashei</i>	4.04	1.8	1.6	60.4	2.8	19.0	1.1	86.8	3.51
<i>J. californica</i> 'A'	0.63	4.9	2.7	19.7	6.4	8.0	8.0	49.7	0.31
<i>J. californica</i> 'B'	0.61	3.9	1.9	18.7	4.7	9.3	9.2	47.7	0.29
<i>J. deppeana</i>	2.69	16.9	3.9	14.9	3.9	26.4	1.0	67.0	1.80
<i>J. erythrocarpa</i>	4.87	1.9	1.6	67.9	3.0	8.5	0.5	83.5	4.07
<i>J. monosperma</i>	1.22	2.7	1.8	61.0	3.8	4.1	1.7	76.3	0.93
<i>J. occidentalis</i>									
var. <i>occidentalis</i>	2.33	8.8	2.6	18.9	1.5	38.9	1.6	72.3	1.68
var. <i>australis</i>	1.78	3.3	1.3	20.1	1.5	38.2	1.6	66.0	1.17
<i>J. osteosperma</i>	1.19	4.0	1.8	40.0	2.6	13.2	1.5	63.1	0.75
<i>J. pinchotii</i>	0.21	2.8	1.2	4.8	t	4.4	—	13.2	0.03
<i>J. scopulorum</i>	3.40	4.3	2.4	57.9	6.1	6.1	3.0	79.8	2.71
<i>J. virginiana</i>	3.18	27.2	7.7	27.6	6.3	15.8	1.0	85.7	2.72

^a ACDR = alpha-cedrene; BCDR = beta-cedrene; THJP = thujopsene; CPRN = cuparene; CDRL = cedrol; WDDL = widdrol.

that new growth of cedars is lower yielding than the "virgin cedars." The new cedars have considerably more sapwood than the older trees and morphologically are allied with the central Texas type as opposed to the western or Mexican type of *J. ashei* (Adams 1975b, 1977). If plantations of *J. ashei* are ever established, germplasm from the western portion (Ozona, TX, and westward) of the range of the species should be evaluated.

The major components of the wood oil of current economic use are alpha-cedrene, beta-cedrene, thujopsene, cuparene, cedrol, and widdrol. Considerable differences in composition were observed among the species studied (Table 2). Cedrol ranged from 4.4% (*J. pinchotii*) to 38.9% (*J. occidentalis* var. *occidentalis*); thujopsene, from 4.8% (*J. pinchotii*) to 61% (*J. monosperma*). The chemical races of *J. californica* had very similar wood oils in contrast to their volatile leaf oils, which are quite different (Adams et al. 1983). Whether these compositional values vary geographically is not known and was beyond the scope of this study.

The sum of alpha-cedrene, beta-cedrene, thujopsene, cuparene, cedrol, and widdrol gives an index for these currently used compounds. *Juniperus pinchotii* not only has very low wood oil yield but the six components of interest are a minor portion of that oil (13.2%). Most oils of the other species contain from 70–85% of these components. Post-multiplication of the percent yield by the sum of the six components/100 gives a relative percent yield (Table 2). On this basis, the most promising species for commercial oil are *J. erythrocarpa* (4.07%), *J. ashei* (3.51%), *J. virginiana* (2.72%), and *J. scopulorum* (2.71%).

Juniperus erythrocarpa is a multi-stemmed tree or shrub occurring in grasslands from trans-Pecos Texas west into New Mexico and Arizona (Adams and Zanoni 1979). The most extensive stands occur in Arizona, but even these populations are probably not dense or extensive enough for commercial use. Additional surveys using aerial photography and biomass data are needed to determine the feasibility of using *J. erythrocarpa* as a commercial source of cedarwood oil.

Juniperus scopulorum is essentially a sibling species of *J. virginiana* found in montane regions of western United States (Little 1971). It generally occurs in foothills of the Rocky Mountains at 4,000–8,000 ft elevation. Extensive, dense populations are the exception for this species. As with *J. erythrocarpa*, aerial photography and biomass data are needed to assess commercial viability.

Neither *J. erythrocarpa* nor *J. scopulorum* appears to be competitive with *J. ashei* and *J. virginiana* as sources of cedarwood oil on a large scale, but both might support small, local distillation facilities.

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