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Production, Yield and Derivatives of Volatile Oils from Eastern Redcedar (*Juniperus Virginiana* L.)

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Abstract: Eastern redcedar (*Juniperus virginiana L.*) is one of the most widely distributed indigenous conifers in southern states including Oklahoma, Arkansas and Texas. Eastern redcedar is also an important source of the volatile oils. This specie adversely influences the environment resulting in degrading grassland, displacement of native plant population and increasing wildlife hazard. Range-grown eastern redcedar has a poor quality. However, some mesic forested sites in Oklahoma produce trees suitable for lumber manufacture. Cedarwood oil has a significant commercial value in a broad range of applications from cold-remedy salves to room sprays and insecticides. Its extensive utilization in a broad range of products is attributable to its unique properties, such as its odor, repellency or toxicity to many pests, antibacterial, antifungal and antitermitic activities. Value-added oil based products from eastern redcedar can have a major economic incentive. It is important to evaluate recovery methods, processing and analysis of the composition as well as its yield. Therefore the objective of the article is to summarize findings of some of the past and current studies related above aspects of volatile oils from eastern redcedar.

Key words: Volatile oils, eastern redcedar, derivatives, recovery methods, oil compositions

INTRODUCTION

Volatile or essential oils are the concentrated aromatic compounds produced from leaves, seeds, barks, roots and the peels of the fruits of wide range of plant species. They vaporize upon contact with air. Volatile oils are recovered from plants by different methods, such as steam distillation and solvent extraction, depending on the present oil quality and the stability of the aromatic components. They can be redistilled and purified for the desired properties of end products. The volatile/essential oils and their derivatives are used for their aromatic property as flavorings in food, beverage products, as fragrances in perfumery and cosmetic products[1-3]. In the U.S. majority of the essential oils are obtained from agricultural plants and some from wildlife sources, such as balsam fir, pine and cedar trees. Eastern redcedar is also referred to as the "Virginia," "Tennessee" "Eastern" or "Southern Red Cedar" (distinguished from the Eastern White Cedar," *Thuja occidentalis* L.), is an important source of essential oil. The redcedar tree is a slow growing, evergreen, with a narrow, dense and pyramidal crown. It can grow to heights of 100 feet tall and five feet diameter in the Southeastern states. Eastern redcedar grows from central part of Virginia through North Carolina into Tennessee, central Kentucky and northern Alabama. It has also invaded in certain parts of Arkansas, southern Alabama, Mississippi, Texas, Florida, Iowa and Oklahoma in scattered form. As an example, more than seven million acre lands in Oklahoma are covered by eastern redcedar[4]. Oil obtained from wood of eastern redcedar (Virginian Cedarwood oil -"Red Cedarwood oil") has been known for a long time and used in a very broad range of products due to its unique properties, such as its odor, repellency or toxicity to many pests. Also cedarwood oil derivatives have been used in numerous industrial products in the form of fragrances and flavors. Therefore, today eastern redcedar is regarded as a potentially valuable and renewable resource for Oklahoma[4].

A number of research studies have been conducted on recovery, processing and analysis of oil from eastern redcedar in an effort to develop value-added products. This study will summarize the history of essential oils production from eastern redcedar and review some of related literature in the field. Properties of essential oils produced from the other tree species in the same family will be compared to those of the essential oils from eastern redcedar.

History of Production and Chemical Properties of Redcedar Oil: Eastern redcedar is known as Virginian Cedarwood Oil (VCO) or Cedarwood oil Virginia in the essential oils trade. VCO should not be confused with the other type of cedarwood oil obtained from the specie of *J. Ashei*-Texas Cedarwood oil (TCO), which

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is also produced commercially. Unlike VCO, (TCO) is a prime product of the recovery process. VCO has been known for a long time. Until the beginning of 20th century VCO oil remained as a by-product of pencil industry. One of the main VCO producers in the U.S is Texaroma Inc. in Lakey, Texas, utilizing the young trees as the raw material to produce oil. The oil obtained from the leaf of Virginian Cedar (VCLO) is not a commercial product like VCO. Due to its poisonous components, it can not be a substitute for the current commercial cedarleaf oil, which is produced from the northern white cedar or arborvitae, *Thuja occidentalis* and the western redcedar, *Thuja plicata.* Today, the commercial cedar leaf oil has been produced in the northeastern U.S., in the eastern Quebec, southeastern Ontario and British Columbia in Canada. The cedarleaf oil industry is small and directed by local farmers who distillate the oil using simple tools. In summary, the commercial VCO oil is distillated from waste wood of furniture industry in North Carolina and the young trees in Texas. The physicochemical properties of essential oils, such as odor, color, specific gravity, optical rotation and solubility in the organic solutions are the main criteria that determine their quality and end uses. The crude VCO is more aromatic and lasting, compared to TCO. Its odor varies from balsamic, sweet and soft to more woody and less balsamic. VCO is commonly used as an ingredient in room deodorants, disinfectants, insecticides, cleansers and other numerous industrial products due to its woody odor and fixative effect. VCO has the color of pale yellow to slightly orange and it is lighter than TCO. Some of the important properties of a good quality VCO is shown below[5].

Specific gravity at 15º/15ºC	0.949-0.961
Optical rotation at 20ºC	-25º27' to -37º15'
Refractive Index at 20ºC	1.5030 to 1.5067
Solubility	Soluble in 8-10 vol. of 90% alcohol;
0.5-4.5 vol. of 95% alcohol is required for solution.	

Klein observed that physicochemical properties of VCO obtained from fresh and old redcedar wood were different from each other[6]. The samples derived from fresh wood had a higher specific gravity and optical rotation than those of derived from the old wood. The higher optical rotations of the fresh wood samples were attributed to their higher cedrol content. Cedrol and cedrene (a mixture of α-cedrene and β-cedrene isomers) are the main components of VCO. Table 1 shows a comparison of the composition of commercial oils. Generally, crude VCO is distillated from waste fresh wood and contains large amounts of cedrol crystals. This product has a refractive index of 1.504 at 20 ºC and less viscous than TCO and it is used for cleaning microscope sections. VCLO had a specific smell of crushed Virginian cedarleaf. VCLO exhibited the following characteristics: Specific gravity of 0.902 at 15 ºC, refractive index of 1.4872 at 20 ºC; and optical rotation of +57.1º.

Recovery methods and oil yield: Currently VCO is recovered by several methods, such as steam distillation, continuous partial pressure, solvent extraction and super critical fluid extraction, for laboratory or commercial uses. Oil yield depends on the type of the recovery method used. It is also correlated to the oil content of the wood, which is affected by the other factors, such as, age of tree, growth/soil and harvesting conditions, time of harvesting, process control systems, tree segment that sample taken from, pretreatment and collecting materials and size of the wood sample. The influence of these major factors on the oil content/oil yield should be taken into the consideration while reviewing major recovery methods used in the laboratory or industry.

Steam distillation is the most commonly used and the simplest method of oil recovery due to its suitability to field conditions[7]. The principal feature of the steam distillation is that steam vaporizes the volatile oil/substances, which are insoluble or slightly soluble in water. Once the steam is cooled down and condensed, the oil separates. Klein reported the oil yield from eastern redcedar wood ranges from 1 to 3.5%, depending on the ratio of the heartwood to sapwood[6]. Eller *et al.* [8] found that wood produced the same yield of oil (3.5%) whether it is from virgin or secondary growth trees. Adams calculated oil yields from heartwood of eastern redcedar species based on both dry material weight and fresh material weight using a laboratory scale steam distillation unit[9-11]. Distillation was carried out for 20h. The yields from heartwood of eastern redcedar were 3.18 and 2.56% on dry and wet bases, respectively. A six hours solvent extraction (hexane followed by methanol) was also carried out on the same material and 4.01% yield on dry base was attained. Payne *et al.*[4] conducted a study to determine the quantity of VCO isolated by various methods including laboratory and commercial steam distillation. In this study, sawdust wood material was pretreated with several different procedures, such as blending, soaking in water for overnight. The samples, which were blended and then soaked overnight, resulted in the highest yield. A modified Abderhalden drying apparatus (ADA) method was developed to determine the oil yield and moisture content accurately[1]. It was found that the oil yield from ADA was greater than the one obtained through laboratory steam distillation (SDA). The results indicated that the oil yield from heartwood chips (4.86%) and that from sawdust (5.65%) were not significantly different while using ADA method[1]. However, in SDA method it was necessary to grind the wood samples to release the oil which is held in the ray cells of the wood. It was concluded that with SDA method disruption of these

Table 1: Comparison of the compositions of commercial cedarwood oils*

Oil kind	α-cedrene	β-cedrene	Thujopsene	α-Selinene	β-Himachalene	Cedrol/ widdrol[a]	Total(%)
Chinese	29.4	8.68	23.6	1.44	3.47	14.2	80.8
Texas	20.5	5.96	29.4	2.11	3.86	26.6	88.5
Perfumers	16.7	5.71	27.7	1.96	4.33	31.5	88.8
Virginia	30.1	7.75	17.7	2.10	3.50	24.28	85.5

[a] Cedrol/widdrol is given as a single percentage because of difficulty in their separation and the integration of their gas chromatography peaks; widdrol is a minor contributor. *After Payne *et al.*[5]

Table 2: Comparison of the compositions of VCO obtained from wood chips by different methods*

Process	α-cedrene (%)	β-cedrene (%)	Thujopsene (%)	Cuparene (%)	Cedrol/ widdrol[a] (%)	
Steam distillation laboratory	0.99	0.408	1.13	0.856	68.2	0.332
Steam distillation Commercial	13.3	3.32	22.7	2.63	39.4	na
Abderhalden 100ºC, N_2[b]	3.93	1.83	5.19	2.40	67.0	0.545

[a]Cedrol/widdrol is given as a single percentage because of difficulty in their separation and the integration of their gas hromatography peaks; widdrol is a minor contributor. * Payne *et al.*[4].

cells had taken place. With ADA method, less time (1.5 h) was spent compared to that of SDA method (10-11h) for isolating the oil. The oil content which was obtained with ADA method varied significantly among the trees, ranging from 1.06 to 3.44% based on fresh material weight. Table 2 displays a comparison of the compositions of VCO obtained from chips by different methods. In conclusion, the studies on the oil yield employing ADA and SDA have shown no significant dependence on the extraction method. However, the effects of environmental factors on the content of the oil recovered with SDA were not studied in detail. Also more comprehensive work needs be done to improve the oil yield in SDA.

Solvent extraction is another commonly used extraction method that employs organic solvents, such as, hexane, methanol and acetone, to recover VCO and VCLO. Since the organic components of the oil can alter under high temperature during the steam distillation method, solvent extraction method is preferable to avoid such changes. Eller and King conducted a study on the effect of the SC- CO_2 extraction parameters on the VCO yield[8]. It was found that the oil yield increased with temperature ranging from 40ºC to100ºC and pressure ranging from 1,500 psi to 10,000 psi. It was noted that the relative density of CO_2 under the condition of 100ºC and 6,000 psi resulted in the highest oil yield of 4.6%. Also, this yield was higher than those of others obtained by steam distillation (3.5%). Cooling the restrictor from 100ºC to 80ºC increased the oil yield from 4.0 to 4.2%. It was found that VCO yield decreased with increased chip age significantly due to loss of volatiles by time. Volatile oil collections were applied on SC- CO_2 extracted chips, steam distillated chips and unextracted chips by using the volatile collection system described by Eller *et al.* to measure the efficiency of SC- CO_2 method. VCO was trapped on Super polymeric adsorbent[12]. Trapped volatiles were extracted from Super polymeric adsorbent with hexane. The extracts were consequently analyzed by gas chromatography (GC). The chromatograms of volatile collections indicated that SC- CO_2 extracted chips released no significant amount of oil. However, the unextracted chips released high amounts of volatiles. The volatile release rates for these tree different types of chips were also measured. The unextracted chips released volatiles at two orders of magnitude higher rate than SC- CO_2 extracted chips did and at four times higher rate than the steam distillated chips did. These results affirmed that SC-CO_2 was an effective method of extracting VCO.

Oil composition and its derivatives: Compositions of the volatile oils are especially critical to flavor and fragrance industry that deals with more than 3,000 aromatic compounds. Since VCO has a low volatility, high tenacity and woody smell, make it specifically preferred in flavor and fragrance mixes and tends to last longer. Some of the VCO components are identified with modern analytical methods, while some of the minor constitutes of the oil are still unknown. A study carried out by Flake *et al.* showed that VCO was composed of 80% cedrene, 3-14% cedrol and small amount of cedrenol[13]. It was found that all these compounds were related to Cedrene, sesquiterpene, were converted to cedrol and cedrenol during aging of VCO. Cedrene is a mixture of isomers, α-cedrene and β-cedrene. α-cedrene constitutes the major fraction of these isomers (75%) and contains endocyclic double bond. β-cedrene, however, contains exocylic methylene group. VCO also contains other small quantities of bicyclic sesquiterpenes. Cedrol, a relatively volatile molecule, is a type of alcohol. It occurs in the oil as a crystallible type (cedrol m.86º) and a liquid type (pseudocedrol). Flake *et al.* reported that the oil extracted from fresh wood had 12.5% cedrol, while the oil extracted from old wood had 4.8% cedrol[13]. This

Table 3: Comparison of the compositions of VCLO obtained from two different regions (Texas and Ontario)*

Components	Region	
	Texas	Ontario
	(% at 0.10-0.20 fresh yield)	
Safrole	18.7	1.5
Saninene	16.3	9.2
Methy eugenol	11.8	0.7
Elemol	7.6	22.8
α-Eudesmol, Elemicin, and β- Eudesmol	8.5	7.0
Me-vinyl anisole "Acetate-2"	6.1	0.3
4-Terpinenol	6.0	6.5
Elemol acetate	4.0	1.8
Methyl citronellate	2.5	8.5
Citronellol	2.3	9.3
γ-Terpinene	1.6	1.0
Linalool	1.4	0.7
γ-Eudesmol	1.2	0.6
α-Pinene	1.3	2.7
Limonene	0.9	17.8
3-Carene	0.8	0.4

* After Von Rudloff[18]

attributed to the loss of the cedrol by time. Cedrenol, a primary sesquiterpene alcohol, occurs in small amounts in VCO. In another study it was reported that various minor constituents i.e., cuparene, widdrol and curcumene, as well as thujopsene, present in VCO[6]. The percentages of commercially important components were given by Adams as α-cedrene (3.18%), β-cedrene (7.7%), thujopsene (27.6%), cuparene (6.3%), cedrol (15.8%) and widdrol (1.0%)[10]. It was noted that all of these compounds made up 85.7% of VCO, but 86.8% of TCO. Sesquiterpene fraction was also analyzed by Walker in detail[5]. By using numerous chemical analysis techniques, including column chromatography on alumina, distillation and gas chromatography, it was found new components in VCO, which were β-elemene, α and β-humulane, caryophllene, an "acorene", valencene, two cuprenes and cuparene.

The major constituents (cedrol and sesquiterpene hydrocarbons) did not play any significant role in the typical VCO odor. Adams confirmed this result in a study, when the VCO composition was compared with that of TCO and Chinese cedarwood oil (*J. funebris*) which shared the same major components (α-cedrene, β-cedrene, thujopsene, cedrol, widdrol) in similar quantities[14]. However, the oils differed in composition of the minor components. Especially, Chinese cedarwood oil had the minor components which caused its off-odor. Twenty-nine different compounds in VCO, 30 in TCO and 31 in Chinese cedarwood oil were reported in the literature[6,15]. 21.1% α-cedrene, 8.2% β-cedrene, 21.3% thujopsene, 1.6% cuparene, 22.1% cedrol and 2.3% widdrol were found in VCO, being different than reported in a related study[10].

Eller and King also proposed that SC-CO_2 method would result in less decomposition and better quality oil than commercial/lab steam distillation since no contact of oxygen occurs[8]. They reported higher cedrol content and lower thujopsene content and no widdrol content compared to the steam distillated samples. However, this difference can be due to the kiln-dried chips employed from which some amount of volatile compounds was already lost. It was found that there were no significant differences among the compositions of VCO obtained at different combinations of temperature and pressure. It was suggested that for better quality deterpenation (i.e. remove non-odorous compounds from the oil) silica could be used in the procedure, as has been described for sweet orange and lemon essential oils[16].

While VCO consists of mainly α-cedrene, β-cedrene, thujopsene, cuparene, cedrol and widdrol, VCLO does not contain large amount of these compounds. The studies on composition of VCLO have been generally used for chemosystematic (taxonomic) applications, such as analyses of hybridization and geographic variation[12,17]. Von Rudloff found that VCLO primarily consists of sabinene, limonene, α-pinene, γ-terpinene, terpinolene, 3-carene, myrcene, 4-terpinenol, citronellol, elemol, γ-, α- β-eudesmal and the aromatic ethers (estragole, safrole, methyl eugenol, methyl citronellate and elemicin)[18]. Small amounts of α-thujene, *p*-cymene, linalool, elemol acetate and δ-cadinene, as well as methyl vinyl anisole were also found. It was concluded that storing VCLO at the low temperature (about 0ºC) in the dark for several weeks changed the composition slightly. VCLO of the trees from Texas and Ontario showed significant differences in composition (Table 3).

Comer *et al.*[17] suggested that if the variation, which is due to the different environmental conditions, is controlled, even the oil from leaves of juvenile eastern redcedar seedlings would be utilized. Setzer *et al.*[9] found variations in VCLO composition between the sexes and habitats in the same region. However, they concluded that the variations between different sexes should be further investigated since they only used 5 trees for each sex. It was also noticed the composition of VCLO was qualitatively and quantitively different from the one reported by Von Rudloff[18] (Table 3). In this study, VCLO consisted of mostly safrole and methyl eugenol and a less amount of sabinene, terpineol, terpinolene, elemol, elemicin, isopropenhylanisole, cineol, eugenol, limonene, myrcene, camphor, linalool and menthenol. Instead of

methyl vinyl anisole, *o*-isopropenylanisole was detected.

Cedryl acetate and acetyl thujopsene are the main derivatives of VCO that have a market value. They are used in fragrances very frequently. Cedryl acetate has

many grades, differing in purity. Its ester content determines its quality/odor. It can be directly separated from VCO by distilling off the lower boiling sesquiterpene components[7,13]. Cedrol can be converted to cedryl acetate by acylation. Another method to produce cedryl acetate is complete acylation of VCO, which yields acetyl thujopsene. Acetyl thujopsene is also referred as vertofix, largely produced from TCO. VCO yields more cedryl acetate than acetyl thujopsene and cedryl acetate lasts shorter time in the fragrances. There is an opportunity to use these constituents of the oil as starting materials for the production of vanillin and helioptropine, which are valuable terpene materials. Methyl eugenol, used commonly in fragrance and flavor industry due to its ginger-like note, can be fractioned from VCLO. This can result in marketing of such constituents as a raw material for the other chemicals in fragrance and flavor industry.

CONCLUSION

Eastern redcedar is one of the most important sources of essential oils. Commercial oil produced from this specie is known as Virginian cedarwood oil (VCO). Compared to TCO, the crude VCO is more oily, lasting, softer and its yellow-orange color is lighter. Due to its woody smell, it is a common ingredient in perfumery. The minor components, such as thujopsenal, caryolan-1-ol and chamigrenal, contribute to VCO odor. VCO differs from TCO and Chinese cedarwood oil in the type and amount of the major and minor components, accounting for their different smells. Sapwood has been found to show higher percentage of cedrol and different components than that of heartwood. Therefore, sapwood of eastern redcedar would be a good source for cedrol production if studied more comprehensively. Composition of VCLO, containing components such as sabinene, limolenene, α-pinene and citronellol, significantly differs from that of VCO. Super critical fluid extraction (SFE) is the newer method that has the advantage of causing less degradation as compared to steam distillation. However, extraction of VCO/VCLO by using SFE method has not been studied extensively to achieve reliable results compared to other methods.

Further study is needed to develop better understanding of oil recovery methods. The variability between trees, within a tree and among growth locations is not known in any detail. In particular, there is a lack of information on the influences of environmental conditions on quality/quantity of VCO. Unlike VCO, VCLO is not produced commercially due to its carcinogenic substances. However, by means of fractionation the useful volatiles can be recovered from this type of oil. Since large amounts of leaves are harvested along with the wood, research and development on VCLO may result in value-added products. Therefore, comprehensive studies are necessary to determine variability between trees, within the tree and among sites regarding both VCO/VCLO to generate value-added products based on eastern redcedar. The development of new processing methods would result in an increase in redcedar utilization and would provide a significant incentive.

REFERENCES

1. Baucard, G.R. and R.W. Serth, 1991. A continuous steam stripping process for the distillation of essential oils. Perfumer and Flavorist, 16: 2-8.
2. Coppen, J.J.W., 1995. Cedarwood oils. In Non-Wood Forest Products (1)-Flavours and fragrances of plant origin. Food and Agriculture Organization of the United Nations, Rome.
3. Simon, J.E., 1990. Essential oils and culinary herbs. In Advances in New Crops. Timber Press, Portland, OR., pp: 472-483.
4. Payne, K.W., R. Wittwer, S. Anderson and E.J. Eisenbraun, 1999. Use of a modified abderhalden apparatus for comparing laboratory and industrial methods of isolating eastern redcedar essential oil. Forest Product J., 49: 90-92.
5. Walker, G.T., 1968. Cedarwood oil. The American Perfumer and Essential Oil Rev., 5: 347-52.
6. Kitchens, G.C., J. Dorsy and K. Kaiser, 1971. Cedarwood oil and its derivatives. The Givaudanian, 1:3-6, 9.
7. Mater Engineering, Ltd., 1992. Oil of cedarwood from *Juniperus virginiana* L. In Opportunities for Rural Development Using Eastern Redcedar in Southwestern Iowa, pp: 4-17.
8. Eller, F.J. J.W. and King, 2000. Supercritical carbon dioxide extraction of cedarwood oil: A study of extraction parameters and oil characteristics. Photochem. Anal., 11: 226-231.
9. Adams, R.P., 1987a. Investigation of *Juniperus* species of the United States for new sources of cedarwood oil. Econ. Bot., 41: 48-54.
10. Adams, R.P., 1987b. Yields and seasonal variation of photochemicals from *Juniperus* species of the United States. Biomass, 12: 129-39.

11. Adams, R.P., 1991. Analysis of Juniper and other forest tree oil. In Modern Methods of Plant Analysis New Series-Essential Oils and Waxes. Springer-Verlag Berlin Heidelberg, Germany, 12: 131-157.
12. Eller, F.J., R.J. Bartelt, B.S. Shasha, D.J. Schuster, D.G. Riley and J.H. Davis, 1994. Aggregation pheromone for the pepper weevil, *Anthonomus eugenii* Cano. J. Chem. of Ecol., 20: 1537-1555.
13. Flake, R.H., E. Von Rudloff and B.L. Turner, 1973. Confirmation of a clinal pattern of chemical differentiation in *Juniperus virginiana* from terpenoid data obtained in successive years. Recent Advances in Phytochem., 6: 215-228.
14. Adams, R.P., 1991. Cedarwood oil-analyses and properties. Modern Methods of Plant Analysis, 12: 159-71.
15. Floreno, A., 1995. Cedarwood oil hike shouldered by buyers. Chemical Marketing Reporter. New York.
16. Dugo, P., L. Mondella, K.D. Bartle, A.A. Clifford and G.P.A. Breen, 1995. Determination of sweet orange and lemon essential oils with supercritical carbon dioxide using silica gel as an absorbent. Flavors and Fragrances J., 10: 51-58.
17. Comer, C.W., R.P. Adams and D.F. Van Haverbeke, 1982. Intra-and interspecific variation of *Juniperus virginiana* and *J. scopulorum* seedlings based on volatile oil composition. Biochem. System. Ecol., 10: 297-306.
18. Von Rudloff, E., 1975. Volatile leaf oil analysis in chemosystematic studies of North American conifer. Biochem. System. Ecol., 2: 131-67.
19. Setzer, W.N., K.W. Whitaker and R.O. Lawton, 1991. A chemical ecological study of the components of the essential oil of the eastern redcedar from three habitats in Huntsville, Alabama. Castenea, 84: 104-110.