The Literature of Juniper utilization for oils and specialty products: A report to the western juniper steering committee.

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**Scope:**

A computer based literature search was undertaken on junipers and their utilization throughout the world for essential oils, medicines, herbal products, pest control agents, and related specialty products.

**Strategy:**

Chemical abstracts were the primary focus of the searches. It has the world's largest databases and is not restricted to chemical utilization, but encompasses medicine, biology, and forest products as well. Also, Medline, Biosis, Forestry Abstracts, and Biological Abstracts were also utilized. Since there are over 700 papers with the key word Juniperus (since 1968), a long list of secondary key words for cross-reference were utilized to focus on appropriate papers. Most commonly, Juniperus or cedarwood oil were cross-referenced to key words such as essential oils, herbal medicine, pest control, flea, tick, fly, mosquito, mites (as well as their scientific names), veterinary, soap, insect, microbial, fungal, etc., etc.

The resulting papers recovered were compiled into the following sub groups, which were then used to compile this report.

reviews
wood essential oils
terpeneoid compounds
lignins
polyphenols
tropolones
medical
antimicrobial
insect
berries (analyses)
leaves (analyses)
regulatory

**Background:**

Aromatic oils from junipers have been used since antiquity for fragrance, flavoring, medicinal, insecticidal, and cosmetic purposes (1-5). Given the widespread occurrence of junipers across the northern hemisphere, it is not surprising to find equally widespread use by many different human cultures (6-11). Often the uses are unique to a particular area, but also many parallel uses are found. While there are over 60 known species of juniper, commercialization has been focused on a relatively few species and important products that are known worldwide.

Cade oil is produced from destructive distillation of *Juniperus oxycedrus* (Cade tree) wood which is native to the Mediterranean region (12-14). This empyreumatic oil contains mostly sesquiterpenes and phenols and is used in both human and veterinary dermatology products, shampoos, and cosmetics. Sometimes the oil is further purified (rectified) by an additional steam distillation.

The juniper berry oil of commerce is an essential oil produced by steam distillation of *Juniperus communis* berries (13-14). This oil is composed mainly of monoterpenes, including α-pinene, myrcene and sabinene as major components, lesser amounts of sesquiterpenes and other volatile compounds. Commercial production is carried out in several European countries including Italy, France, Germany, and Austria. Perhaps the most famous use of this product is to flavor gin and alcoholic bitters. It is generally recognized as safe for human consumption (GRAS) and also finds use in many other food products such as frozen deserts, gelatins, puddings, and meats (15). The berries themselves are compounded into spices and both berries and oils are used as phytopharmaceuticals in Europe, most commonly as a diuretic (16). Researchers are also investigating its potential antidiabetic activity (17). Aromatherapy seems to be a newer expanding market at the moment for this product (18). The berry oil is also used in many non-food/medicinal applications such as fragrances for soaps, perfumes and cosmetics (19).

Cedarwood oil is a generic term for the essential oil produced from the steam distillation of several different aromatic woods throughout the world (13,14,20). They can have quite different chemical and physical properties, although some are also similar. In the U.S., two major commercial products are produced (21). Virginiana cedarwood oil, which is produced from *Juniperus virginiana* heartwood, has also been called Tennessee, Eastern, and Southern red. Texas cedarwood oil is made from *Juniperus ashei*. Currently the U.S. products are contending with imported Chinese cedarwood oil produced from *Cupressus funebris*. All three oils are similar in that they contain the sesquiterpenes α-cedrene, β-cedrene, thujopsene, cedrol and widdrol as the major components, however, in quite different ratios. In one gas chromatography-mass spectrometry (GC-MS) analysis, the cedrol content of Virginiana oil was 22% while that of Texas oil was 19% and Chinese cedarwood oil was 9.6% (20). Cedrol content is one factor which influences price. Another is odor and color. Chinese oil contains a burnt note that also drives its price lower (21). Virginiana oil currently sells for $6.80/lb while cedrol is $12.75/kilo (22). Reportedly, U.S. producers are presently looking at new ways to help offset production costs. One is the use of the waste cedar fiber in the landscaping and soil amendment business (21).

Other sources of cedarwood oil throughout the world have been *Cedrus atlantica* (Moroccan), *Cedrus deodara* (Himalayan), and *Juniperus procera* (East African). The primary use of these oils is again for fragrance or as a chemical base for the fragrance and cosmetic industries. In the Pacific Northwest, steam distillation of western redcedar heartwood (*Thuja plicata*) has been done on a smaller scale from time to time to produce material for pet products.

Carbon dioxide extraction of essential oils offers an energy efficient and environmentally attractive alternative to steam extraction. Comparison has been made of these two extraction procedures for juniper berries (111). Slightly different compositions were obtained in each case. Capitol set up costs are likely prohibitive for a small industry at this time, but this process should be kept in mind as its technology gets cheaper.

**Chemistry of *Juniperus* Wood and Bark:**

Sesquiterpenes have been the most investigated compounds of the various juniper heartwoods (23-36). This should not be surprising, considering the abundance of these compounds in the essential oils and their economic importance. Several types of sesquiterpenes are found in Juniper oils (Figure 1). In most, the major compounds have cedrane, thujopsene, or cuparane type C-15 skeletons. An exception is the oil from *J. oxycedrus* whose major components are cadinane type sesquiterpenes (37). GC-MS has become an important tool for the rapid analyses of essential oils and allowing for the comparisons of the product from various sources (38). This tool has been employed effectively in comparing juniper wood essential oils. In recent years, the essential oils from various juniper heartwoods have also played an important role as sources for new sesquiterpenes (39-45). In addition to several new structures, three new types of C-15 skeletons have also been discovered.

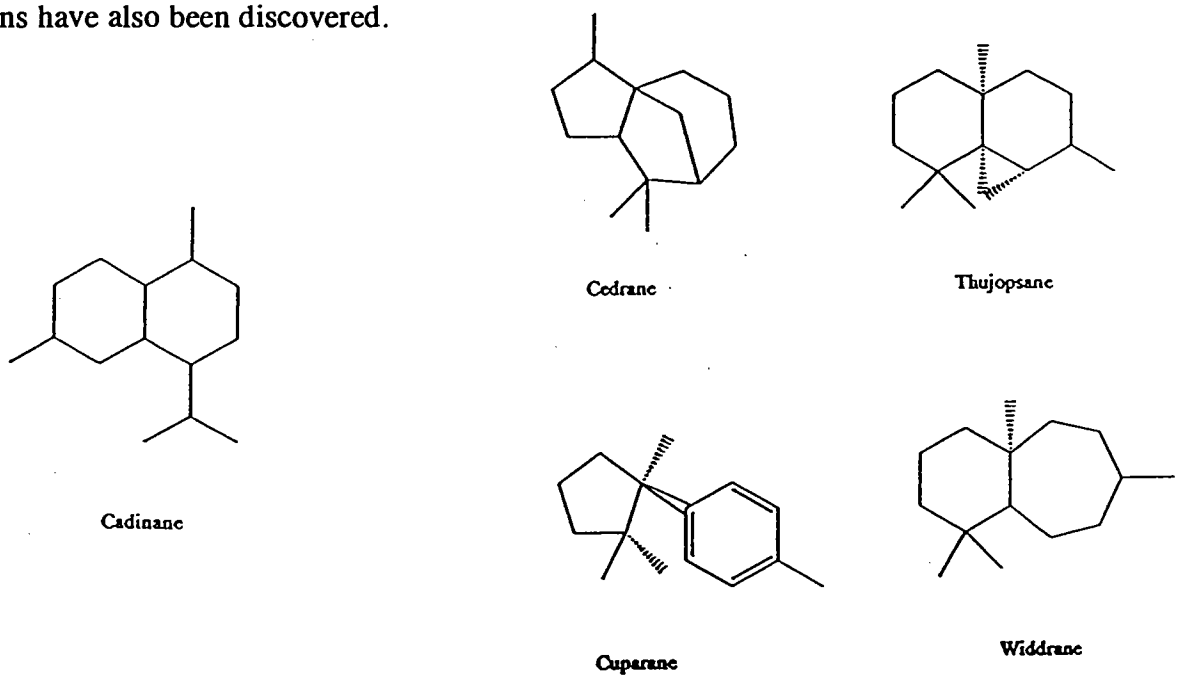


Figure 1. Sesquiterpene skeletal types found in Juniper wood essential oils.

In contrast to the heartwood sesquiterpenes, there have been relatively fewer studies on the other natural products in the wood and bark of junipers (23, 44-51). However, these studies have shown the occurrence of diterpenes and lignans in both the wood and bark of a number of different species. This indicates the likelihood of more widespread occurrence in the numerous other juniper species yet to be investigated, such as western juniper. The diterpenes found so far belong to three main structural types, the labadanes, abietanes, and totoranes (Figure 2). Recent studies also suggest that the junipers may be a rich and diverse source of new diterpenes which in some cases could be good starting materials for fragrance chemicals (48). In support of this view, several new diterpene structures have very recently been discovered in both juniper wood and bark (46,47,50,51). The lignan podophyllotoxin which is noted for anticancer activity has been known to occur in *Juniper communis* wood (23) and *Juniperus virginiana* leaves for some time (52). Recent discoveries of new lignans with related structures to podophyllotoxin in juniper heartwood and bark have been made (44,53).

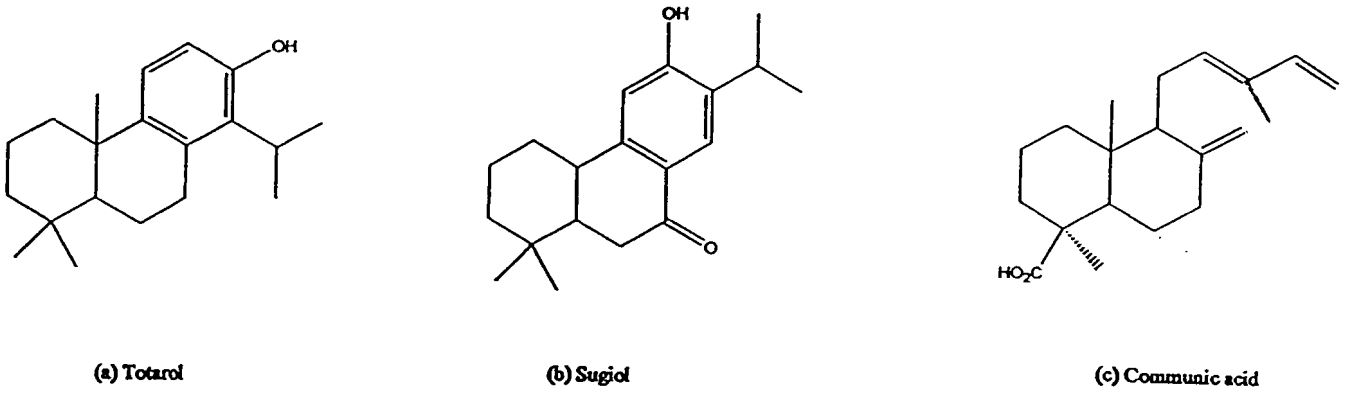


Figure 2. Diterpene types found in Juniper wood and bark.
(a) Totarane (b) Abietane (c) Labadane

Investigations of polyphenols in juniper barks have been surprisingly limited. The bark of J. communis was found to contain dimeric procyanidins (dimers of the catechin and epicatechin linked C4→C8) B1, B2, B3, and B4 (54). In an earlier study the same research group had found the associated flavanoid monomers (+)-catechin and (-)-epicatechin along with other flavanoids (+)-afzelechin, (-)-epiafzelechin, (+)-gallocatechin and (-)-epigallocatechin (55). These compounds all have important antioxidant properties.

**Biology of *Juniperus* extracts and plant parts:**

A wide range of biological activities have been explored for a relatively few juniper species. Wood oils, leaf oils, leaves, and berries have been most frequently studied. Table 1 summarizes papers referring to insect and arachnid activities. Table 2 lists papers with medicinal activities studied. Table 3 is a summary of publications of antimicrobial studies. The publications in Tables 1-3 are not all inclusive, but summarize the results of the literature search results. Many additional references on a given subject can be found within the papers listed here.

Table 1. Insect/Arachnid activities studied

| Organism | Material | Ref. |
|---|---|---|
| Mosquito | cedarwood oil | 56 |
| Mosquito | cedarwood oil | 57 |
| Mosquito | cedarwood oil (individual compounds) | 4 |
| Pulse Beetle | cedarwood oil | 58 |
| Flour Beetle | cedarwood oil | 63 |
| Carpet Beetle | cedarwood chests | 61 |
| Carpet Beetle | heartwood & leaf oil | 64 |
| Clothes moth | wood, wood oil, leaf oil | 61, 62, 64, 65 |
| Mites | leaf oil, other? | 65 |
| Fleas | | 65 |
| Ticks | wood oil | 67 |
| Termite | wood oil, leaves | 59, 66 |

Table 2. Medicinal activities

| | | Ref. |
|---|---|---|
| medical warming | berries | 68 |
| anti-fertility effects | berries | 69 |
| anti-neoplastic | leaves | 70 |
| anti-schistosoma | wood oil | 71 |
| anti-fertility | berries | 72, 87 |
| anti-herpetic | berries | 73 |
| anti-platelet/vasoreloxing | ? | 74 |
| cytotoxicity | leaves | 75 |
| hypertension | berries | 76, 88 |
| muscle relaxant | ? | 77 |
| dermatology | cade oil | 78 |
| cytotoxic agent | ? | 79 |
| platelet aggregation | leaves | 80 |
| kidney medicine | berries | 81 |
| anti-cancer | leaves | 82 |
| abortive | leaves | 83 |
| ethnobotany | ? | 84 |
| mouth care (toothpaste) | ? | 85 |
| platelet-activating | cedrol | 86 |
| anti-inflammatory | berries | 7 |
| hypoglycemic | berries | 17 |
| diuretic | berries | 89 |

Table 3. Antimicrobial studies

| | | |
|---|---|---|
| western juniper | wood, bark, leaves | 90 |
| other species of juniper | | 1, 93, 94, 95, 96, 97, 98,101, 102 |
| cedrol | | 91 |
| cedrane compounds | | 92 |
| diterpenes of juniper | | 98, 99, 100 |
| antimicrobial effects on deer | | 103 |

**Previous Studies on *J. occidentalis*:**

Kurth and Lackey, at Oregon State University, were the first to investigate the extractives of western juniper (104). They isolated the sesquiterpenes cedrol and cedrene from the heartwood. Also found in the heartwood extracts were the phytosterols α and β-sitosterol and other incompletely identified substances. In a later study on the essential oil, the yields varied depending on the height of the heartwood within the tree trunk (105). The average yield of essential oil was 2.26% from heartwood taken at the bottom of the trunk while the yield was only 1.09% on samples from the top of the trees. Cedrol content of the essential oil, varied from 15 to 40%. In later studies, *J. occidentalis* was included in a rather expensive investigation of *Juniperus* species from the United States as potential new sources of "cedarwood oils" (38). Essential oils from the heartwood of 11 species were analyzed and compared by GC-MS. *J. occidentalis* was found to have an essential oil yield of 2.3% and the major commercially important compounds identified as α-cedrene (8.8%), β-cedrene (2.6%), thujospene (18.9%), cuparene (1.5%), cedrol (38.9%) and widdrol (1.6%). *J. occidentalis* was shown to be one of the richest sources of cedrol.

Biological studies have indicated that some antibacterial activity was shown by *J. occidentalis* heartwood extracts against *S. aureus, B. subtilis*, and *M. smegmatis* (90). Only minor antifungal properties were found. These studies did not test activity toward wood rotting fungi. Termaticidal activity has also been demonstrated for western juniper heartwood extracts (66). Recently, a study by the Principal Investigator, has shown that the essential oil of western juniper has effective acaricidal activity in killing tick larvae and nymphs (67). The active compounds were identified as cedrene-13-ol(A) and cedrene-15-ol(B). Interestingly, the other major cedranes in the oil, including cedrol, were not active in this case (Figure 3).

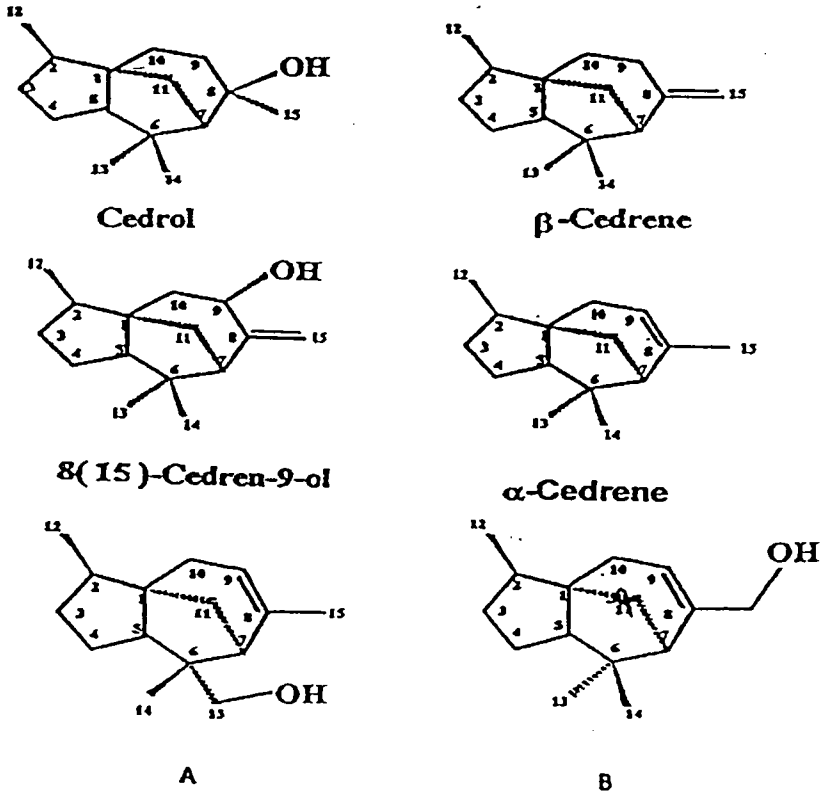


Figure 3.

Cedrane series from Western juniper.

Western juniper does not contain tropolone type compounds in its wood, unlike other junipers and cedars that do. Tropolones are compounds that chelate metals and are toxic to fish and some plants. These substances corrode metals, cause excessive blade wear, and stain with iron (106). The fact that they are not present is of significant value. Tropolones cause significant problems for Western red cedar, Alaska cedar, and Port Orford cedar utilization and waste disposal.

A detailed GC-MS analysis of western juniper leaf oil was reported by Von Rudloff *et al* in 1980 (107). The volatile components of the leaves are quite different from those of the heartwood essential oil. The major component is bornyl acetate (21-39%) with significant amounts of p-cymene and sabinene. Bornyl acetate and p-cymene are relatively cheap commodity chemicals. Sabinene, on the other hand, is relatively expensive and targeted use of leaf oils may be explored on sabinene activity.

There have been no analyses carried out on western juniper berries. An email conversation with Robert Adams (Texas) confirmed that he has not published a GC-MS profile of western juniper berries that I may have missed.

**Possible regulations and precautions:**

Use of Juniper products is cited as gras "generally accepted as safe" in several articles referenced here. Also, many report uses suggesting little or no problems. However, a precautionary note is warranted. Other papers warn about possible toxic effects (108). Junipers have a "toxicology" profile as well which must be considered. The chemical makeup of extracts from various junipers may be similar at times, but the exact composition may be unique to western juniper. Therefore, the chemical makeup of any western juniper product which may impact human health needs to be considered.

In 1993, the EPA (Environmental Protection Agency) determined that the currently registered uses of cedarwood oil for pest control of moths, mildew, and fleas doesn't cause an unreasonable risk to humans (109, 110). The documents review regulatory history and indicate that "cedarwood oil is a distilled extract from the cured cedarwood obtained from *Juniperus virginiana* oil and other species of cedar." This statement would seem to open the d s a true cedar and both are closely related species of J d to as "cedar" in common American slang. One needs t ow they affect a potential product from western juniper.

**Recommendations:**

The essential oil from western juniper heartwood is a valuable resource due to its high cedrol content and overall similarity to *Juniperus virginiana* oil. However, it seems that production of oil alone will not be competitive. A more complete and perhaps unique utilization of this resource is needed. The following are suggested avenues.

Separation of cedrol from the heartwood oil and and use of the other sesquiterpenes in another value added product. The other sesquiterpene fraction needs to be examined for biological activity.

Juniper oils have a history of widespread pest control use. Western juniper oil and its various fractions need to be tested against all pests that would provide a market outlet. Because of the variability of composition in other junipers, even those pests that have been previously tested need to be tested for western juniper. This is perhaps one of the more promising areas for investigation.

The residue from steam distillation, which is called marc, needs to be investigated as a value added product. The steam distillation process extracts mono and sesquiterpene compounds, but leaves diterpene and higher molecular compounds that may still have value and biological activity. Diterpenes from other junipers have been shown to have antimicrobial activity. Could the marc be dried and used for its absorbent and antimicrobial activity? The marc from *Juniperus virginiana* is used for landscaping and as a soil amendment. Western juniper marc will have to find a higher value. As a pest control agent it will have to operate in contact fashion, as the volatile compounds will be gone.

Antioxidants (proanthocyanidins) of the type found in the currently sold health food supplements were shown to be abundant in the bark of western juniper by recent preliminary tests in my laboratory at Oregon State University. The present commercial market is now supplied mostly from pine bark and grape seed extracts. Prices are currently around $1000 per kilogram for food grade material. The resource needs to be evaluated for extractable amount, removal without pitch, and exact molecular weight distribution. These substances are oligomeric and a certain oligomer profile is desired by the industry.

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