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Gas chromatography mass spectral analysis of free and glycosidically bound volatile compounds from *Juniperus oxycedrus* L. growing wild in Croatia

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Abstract

The essential oils in fresh needles and green and mature berries of *Juniperus oxycedrus* L. (Cupressaceae) were analyzed by GC–MS. A total of 36 compounds were identified from needles, representing 94.91% of the total oil. 15 compounds were identified in the green berry oil and 22 in the mature berry oil, which accounted for 94.33 and 90.94% of the total oil composition. The major component was α-pinene. The glycosidically bound volatile compounds amounted to 21 mg kg^{-1} in needles and 4 mg kg^{-1} from green berries. Only traces of aglycones were identified in mature berries. Sixteen volatile aglycones were identified in needle sample with 2-hydroxy-5-methylacetophenone as major component. A total of nine aglycones were identified in green berries. The major aglycones were 3-phenyl-2-propen-1-ol and myrtenol. There was no similarity between the glycosidically bound aglycones and the corresponding free compounds found in the essential oil.

Keywords: *Juniperus oxycedrus* L. (Cupressaceae); Glycosides; Essential oil; GC–MS

1. Introduction

Different *juniperus* species have been used in traditional medicine for centuries as incense, diuretics and remedies for indigestion. *Juniperus oxycedrus* L. (Cupressaceae) is a common shrub or small tree growing wild in stony places of Mediterranean and Near East countries. This plant is used frequently for the preparation of traditional medicinal brandy in Dalmatia. In the present paper, the gas chromatography mass spectral analysis (GC–MS) was applied to investigate the free and glycosidically bound volatile compounds from *J. oxycedrus* berries and needles. The study of those compounds may be of some interest for the pharmacology and food industry as potential resource of medicinal and aroma compounds.

The detailed study of the essential oil of *J. oxycedrus* berries has been realized by Hernandez, Del Carmen Lopez Martinez and Villanova (1987) and Mennut, Lamaty and Bessiere (1997). The essential oil content was 0.50–0.70% (v/w) and a total of 26–33 components were identified. These authors agreed that the principal components include α-pinene, β-myrcene, γ-muurolene and limonene. The most complete report on the leaf oil has been described by Stassi, Verykokidou, Loukis, Harvala and Philianos (1995) and Adams (1998). The leaf oil was dominated by α-pinene and cedrol with moderate amounts of dihydro-*p*-cymene-8-ol, α-terpineol and δ-cadinene.

Together with the essential oil, there is a growing interest for the study of glycosidically bound volatile compounds. The presence of those compounds in different plants has been well established earlier and many publications have dealt with their chemistry and distribution in the vegetable kingdom (Bilia, Rubio, Ladero Alvarez, Morelli, & Munoz Gonzales, 1994; Francis & Allcock, 1969; Guo et al., 1994; Grzunov, Mastelić & Ružić, 1985; Merkx & Baerheim Svendsen, 1990; Nishiya, Kimura, Takeya & Itokawa, 1992; Stahl-Biskup, Intert, Holthuijezen, Stengele & Schultz, 1993; Winterhalter, 1990). The glycosides are able to release volatile compounds by enzymatic or acid hydrolysis and can be considered as aroma precursors in plant materials. The glycosides with β-glycosidic linkage are more common than those with α-glycosidic linkage (Nirmala, Menon & Narayanan,

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1992) and the β-glucosidase from bitter almonds is commonly used for hydrolysis of the larger part of β-glycosides.

Our attention was focused to the investigation of the essential oils and glycosidically bound volatiles isolated from *J. oxycedrus* berries and needles. Because, to our knowledge, the research on the presence of the later compounds has not been reported to date, our aim was to isolate them and identify the composition of the aglycones and to show their possible similarity with the free compounds found in corresponding essential oil. Bound fraction was determined indirectly through release of aglycones by β-glucosidasic activity. Then, the compositions of both, free and released volatile aglycones, were analyzed by capillary gas chromatography coupled to mass spectrometry.

2. Experimental

2.1. Reagents

The solvents (*n*-pentane, *n*-hexane, methanol, ethyl acetate, acetonitrile, diethyl ether) were of GC grade from Fluka. Ammonia was from Merck. β-Glucosidase from bitter almonds (5–8 U/mg) and octyl-β-D-glucopyranoside were from Fluka. *n*-alkanes C_8–C_{22}, purity >99%, were from Sigma. Amberlite XAD-2 (20–60 mesh, obtained from Fluka) was treated according to the procedure of Gunata, Bayanove, Baumes and Cordonier (1985).

Column chromatography was performed on silica gel obtained from Merck (Kieselgel 60, Merck, 0.040–0.063 mm). Thin layer chromatography (TLC) was performed on 0.2 mm precoated silica plates (Kieselgel 60, Merck) with hexane/ethyl acetate (85:15, v/v) as eluent. The volatile compounds were revealed using 2% vanillin in concentrated sulfuric acid.

2.2. Plant material

The *J. oxycedrus* L. needles and berries were collected in the region of central Dalmatia, near Split, in March 1998. The voucher specimen was deposited in the Laboratory of Organic Chemistry, Faculty of Chemical Technology, Split, Croatia.

2.3. Isolation of the essential oil

Upon the addition of internal standard, octyl-β-D-glucopyranoside (0.5 mg), the plant material was subjected to hydrodistillation using 500 ml of distilled water. This process was performed in a Clevenger type apparatus for three hours. Essential oil was dried over anhydrous sodium sulfate and stored under nitrogen in sealed vial, at −20°C until required.

2.4. Fractionation of the essential oil

An aliquot, 20 μl of the essential oil, was fractionated on a microcolumn of silica gel 60 (0.040–0.063 mm) and five fractions were obtained. 10 ml pentane was used for fractionation (fraction I) and mixtures of pentane–ether: 5 ml 5% ether (fraction II), 5 ml 10% (fraction III), 5 ml 50% (fraction IV) and 5 ml pure ether (fraction V). All fractions were concentrated to 0.5 ml, tested by TLC and 2 μl of each fraction was injected for GC–MS analysis.

2.5. Isolation of glycosidically bound volatile compounds

After the hydrodistillation, the remaining aqueous extract was filtered, and the filtrate was subjected to liquid–solid chromatography in a glass column (150×20 mm) containing Amberlite XAD-2 as adsorbent (Gunata et al., 1985). The column was washed with water (500 ml), and the retained components were subsequently eluted with methanol (100 ml). After evaporation of the solvent, the residue was redissolved in 2 ml citrate–phosphate buffer (pH 5). Remaining volatile compounds were removed with 4×5 ml of *n*-pentane. Prior to enzymatic hydrolysis, the absence of volatile compounds was tested by TLC and GC–MS.

2.6. Enzymatic hydrolysis

In a typical experiment, β-glucosidase from bitter almonds (10 mg, 5–8 U/mg; Fluka) was added to the glycosidic extract. The enzymatic hydrolysis was realized during 48 h at 37°C. The liberated aglycones were extracted from aqueous layer with 4×5 ml of pentane. The combined pentane extract was concentrated to 0.5 ml, and 2 μl were injected for GC–MS analysis.

2.7. Gas chromatography–mass spectrometry

The analyses of the volatiles were run on a Hewlett-Packard GC-MS system (GC: 5890 series II; MSD 5971A) on two fused-silica capillary columns of different polarity: HP-20 M (polyethylene glycol; 50 m×0.2 mm i.d.; film thickness 0.2 μm) and HP-101 (dimethylpolysiloxane; 25 m×0.2 μm i.d.; film thickness 0.2 μm). The columns were directly coupled to the MS. The carrier gas was helium, flow rate of 1 ml/min. For HP-20 M oven temperature was programmed: 70°C for 4 min, then 70–180°C at 4°C/min and subsequently held isothermal for 10 min. The temperature program for HP-101 column was as follows: 70°C for 2 min, then 70–200°C at 3°C/min, held isothermal for 10 min. Injection port temperature 250°C; detector, 280°C; split ratio 1:50. The ionization of the sample components was performed in the EI mode (70 eV) for both columns.

2.8. Identification and quantitative determination of components

Linear retention indices for all compounds were determined by coinjection of the sample with a solution containing a homologous series of C_8–C_{22} *n*-alkanes (Van Den Dool & Kratz, 1963). Individual constituents were identified by comparison of their retention indices with library data (Adams, 1995), with those obtained from known plant sources and those obtained from authentic components and by comparison of their mass spectra with those in the Wiley MS library. Because it is possible to have the same constituent in multiple different fractions, the content of components in each fraction of essential oils were calculated from a GC peak areas related to GC peak area of 1 mg menthol added as internal standard. Percentage of total (Table 1) was obtained by their addition. Preliminary GC–MS analysis showed the absence of menthol in essential oil samples.

The aglycone concentrations were calculated from the GC peak areas related to GC peak area of 1-octanol (from the internal standard octyl-β-D-glucopyranoside). Preliminary GC–MS analysis showed the absence of 1-octanol as potential aglycone in plant material.

Table 1
Percentage composition of the essential oil isolated from needles, green and mature berries of *J. oxycedrus* L

No.	Component	RI[a] HP-20M/HP-101	Percentage of total			Mode of identification[b]
			Fresh needles (Yield 0.05%)	Green berries (Yield 0.16%)	Mature berries (Yield 0.04%)	
1.	α-Pinene	1038/936	41.37	66.30	61.21	GC, MS
2.	β-Pinene	1102/972	0.53	2.04	2.37	GC, MS
3.	β-Myrcene	1149/986	0.67	4.90	5.73	GC, MS
4.	Limonene	1183/1023	0.06	1.43	1.95	GC, MS
5.	Terpinolene	1262/1079	–	–	0.28	GC, MS
6.	1-Octen-3-ol	1411/979	0.25	0.45	0.47	GC, MS
7.	α-Cubebene	1438/1340	0.49	0.72	0.45	GC, MS
8.	α-Campholene aldehyde	1452/–	0.15	–	0.29	GC, MS
9.	Camphor	1472/1122	0.20	0.53	0.96	GC, MS
10.	β-Bourbonene	1496/1369	0.21	–	–	GC, MS
11.	Sesquiterpenic hydrocarbon	1524/–	0.84	–	–	–
12.	Bornyl acetate	1550/1266	0.07	1.88	1.40	GC, MS
13.	α-Elemene	1554/–	0.63	–	–	GC, MS
14.	Terpinen-4-ol	1557/1175	0.15	0.17	0.54	GC, MS
15.	Calarene	1564/1402	–	1.76	0.62	GC, MS
16.	β-Caryophyllene	1585/1401	0.14	–	0.56	GC, MS
17.	*trans*-Verbenol	1606/–	–	–	2.97	GC, MS
18.	Allo-aromadendrene	1612/1447	0.44	–	–	GC, MS
19.	α-Terpineol	1624/1183	0.45	–	–	GC, MS
20.	α-Humulene	1638/1437	0.87	1.91	2.35	GC, M
21.	Sesquiterpenic hydrocarbon	1680/1466	4.40	9.87	5.22	MS
22.	α-Muurolene	1693/1482	0.16	–	0.30	GC, MS
23.	δ-Cadinene	1716/1503	1.44	1.32	1.22	GC, MS
24.	Myrtenol	1734/1196	0.05	–	0.31	GC, MS
25.	2-Tridecanone	1771/1475	0.34	–	–	GC, MS
26.	*trans*-Carveol	1776/1220	0.11	–	0.18	GC, MS
27.	Dihydrofarnesal	1961/1594	3.35	–	–	GC, MS
28.	Unknown	1982/–	1.22	–	–	–
29.	E-Nerolidol	1987/1552	0.23	–	–	MS
30.	Dodecenyl acetate[c]	1992/1651	6.32	–	–	MS
31.	Oxygen containing sesquiterpene	2015/–	1.28	–	–	–
32.	T-muurolol	2130/–	0.50	–	0.26	MS
33.	Oxygen containing sesquiterpene	2139/1604	0.27	0.06	–	MS
34.	*cis*-Farnesol	2168/1687	1.27	–	–	GC, MS
35.	*trans,trans*-Farnesal	–/1714	2.16	–	–	GC, MS
36.	3,7,11-trimethyl-6,10-dodecadien-1-ol	–/1684	2.15	–	–	MS
37.	Unknown	–/1922	1.25	–	–	–
38.	Manoyl oxide	–/1956	12.29	0.99	1.30	GC, MS
39.	Farnesol	–/1728	8.60	–	–	GC, MS
	Total		94.91	94.33	90.94	

[a] RI, retention indices relative to C_8–C_{22} *n*-alkanes on polar HP-20 M and apolar HP-101 columns.
[b] GC, compared with retention indices of components of reference essential oils; MS, mass spectra.
[c] Correct isomer is not identified (by tentatively identification on the basis of the MS it is E3-dodecenyl acetate).

3. Results and discussion

3.1. Essential oil

Among other things, the aim of this work was to show that there is no similarity between the glycosidically bound volatile compounds and free volatile compounds found in the essential oil. In this purpose, all oil samples were fractionated on a microcolumn of silica gel, as described in experimental part. The fractionation procedure was necessary to show the possible presence of the traces of aglycones in essential oil, which could be masked by main components.

The amount of 0.05 g of the essential oil was obtained from 100 g of fresh needles. A total of 36 compounds were identified in needles, representing 94.91% of the total oil (Table 1). The major compound was α-pinene (41.37%). Manoyl oxide was the second most important compound with peak area of 12.29%. Other important compounds were farnesol (8.60%), mixture of dodecenyl acetate isomers (6.32%), unidentified sesquiterpenic hydrocarbon (4.40%) and dihydrofarnesal (3.35%).

The yields of the berry oils were 0.04% (mature) and 0.16% (green). The chemical composition of the oils isolated from green and mature *J. oxycedrus* berries are also reported in Table 1. Fifteen compounds were identified in the oil from green berries and 22 compounds from mature berries, which accounted for 94.33 and 90.94% of the total oil composition, respectively. Although it is evident that there is a difference in the yield and number of identified compounds, the general similarity in chemical composition between two berry oils is obvious. The major compounds were α-pinene (66.3–61.21%), unidentified sesquiterpenic hydrocarbon (9.87–5.22%), β-myrcene (4.90–5.73%), α-humulene (1.91–2.35%), bornyl acetate (1.88–1.40%) and δ-cadinene (1.32–1.22%).

On the other hand, there is a large difference in berry oils in comparison with needle oil. It was of interest to note that a survey of two type oils revealed that they possessed a wide variation in the number of identified compounds, their content and chemical composition.

3.2. Glycosidically bound volatile compounds

For the first report on the glycosidically bound volatiles in *J. oxycedrus* L., the glycosides isolation was performed from fresh needles, green and mature berries of common *J. oxycedrus* tree. Approximately, 21 mg kg^{-1} of volatile aglycones were released from needles

Table 2
Percentage composition of glycosidically bound volatile compounds in *J. oxycedrus* L. isolated by liquid–solid chromatography on Amberlite XAD-2 as adsorbent and hydrolyzed by means of β-glucosidase

No.	Component	RI[a] HP-20M/HP-101	Percentage of total			Mode of identification[b]
			Fresh needles (Yield 0.0021%)	Green berries (Yield 0.0004%)	Mature berries (Yield: traces)	
1.	3-Methyl-3-buten-1-ol	1212/–	3.33	–	–	GC, MS
2.	3-Methyl-2-buten-1-ol	1281/–	3.09	–	–	GC, MS
3.	1-Hexanol	1315/–	1.88	–	–	GC, MS
4.	3-Hexen-1-ol	1346/–	4.22	–	–	GC, MS
5.	1-Octen-3-ol	1412/980	4.03	–	–	GC, MS, CO
6.	Thymoquinone	1686/1222	–	7.07	–	GC, MS
7.	Methyl salicylate	1715/–	–	2.59	–	GC, MS
8.	3-Methyl-2-butenoic acid	1730/1027	1.49	–	–	GC, MS
9.	Myrtenol	1734/1195	–	25.69	Traces	GC, MS
10.	α-methylbenzyl alcohol	1752/–	2.67	–	–	GC, MS
11.	Benzyl alcohol	1811/1099	3.35	2.83	–	GC, MS, CO
12.	2-Phenylethanol	1848/1153	3.64	2.23	–	GC, MS, CO
13.	2,3-Dimethylphenol	1905/–	1.57	–	–	MS
14.	Ethylbenzaldehyde	1922/1826	4.08	–	–	MS
15.	*O*-methoxybenzyl alcohol	2082/1309	3.02	1.39	–	GC, MS
16.	Eugenol	2099/1377	1.50	9.13	–	GC, MS, CO
17.	2-Hydroxy-5-methylacetophenone[c]	2123/1336	29.49	–	–	MS
18.	3-Phenyl-2-propen-1-ol	–/1372	1.81	29.13	–	GC, MS, CO
19.	4-Hydroxy-3-methoxybenzaldehyde	–/1502	–	3.77	Traces	GC, MS, CO
20.	3,5-Dimethoxyphenol[d]	–/1531	2.04	–	–	MS
	Total		71.21	83.83	Traces	

[a] RI, retention indices relative to C_8–C_{22} *n*-alkanes on the polar HP-20 M and apolar HP-101 columns.
[b] GC, compared with retention indices of known components; MS, mass spectra; CO, identity confirmed by comparison with authentic compounds.
[c] Identification by comparison of the MS and retention time determined by.
[d] Jean, Garneau, Collin, Bouhajib and Zamir (1993) and Young and Paterson (1995).

glycosidic extract, 4 mg kg^{-1} from green berries and only traces of aglycones from mature berries glycosidic extract. As shown in Table 2, 16 volatile aglycones were identified in needles sample. The major aglycone is 2-hydroxy-5-methylacetophenone (29.49%). Other important aglycones are 3-hexen-1-ol (4.22%), ethylbenzaldehyde (4.08%), 1-octen-3-ol (4.03%), 2-phenylethanol (3.64%), benzyl alcohol (3.35%), 3-methyl-3-buten-1-ol (3.33%), etc. For comparison, a total of nine volatile aglycones were identified from green berries glycosidic extract. The major aglycones are 3-phenyl-2-propen-1-ol (29.13%) and myrtenol (25.69%). Eugenol (9.13%) and thymoquinone (7.07%) represent also important percentage content.

Comparison of the yields and the chemical and percentage composition of released aglycones from *J. oxycedrus* needles and berries shows a noticeable difference. The glycosidically bound volatile compounds in the green berries amount to 0.0004%, thus, representing only one fifth of those compounds in needles (0.0021%). Additionally, the major compounds are different. It is interesting that in mature berries the volatile aglycones have no been found.

Some of the volatile compounds are very common as aglycones in different plants. The aglycones such as aliphatic alcohols, 2-phenylethanol, benzyl alcohol, eugenol, linalool, geraniol, nerol and α-terpineol might be considered as ubiquitous in aglycone fractions (Stahl-Biskup et al., 1993). Our investigations and global results concerning the chemical and percentage composition of aglycones in different species from the Cupresaceae family, shows a noticeable differences. The most common aglycones occurring in this species are the phenylpropyl derivatives and related compounds. For example, regarding the main aglycones, in *J. oxycedrus* L. needles (this work) it is 2-hydroxy-5-methylacetophenone (29.49%) and in berries it is 3-phenyl-2-propen-1-ol (29.13%), in *Cupressus sempervirens* L. (Miloš, Mastelić & Radonić, 1998; Miloš and Radonić, 1996) it is thymoquinone (27.78%) in both leaves and cones, in *Cupressus arizonica* Green (Miloš & Mastelić, 1998) and in *Juniperus communis* L. (Mastelić, Miloš, Kuštrak & Radonić, 1998) it is 3-phenyl-2-propen-1-ol (31.20 and 36.76%).

Finally, the comparison of *J. oxycedrus* bound and free volatile compounds shows that there is no similarity between the free compounds found in the essential oils (Table 1) and the corresponding glycosidically bound aglycones (Table 2). Only myrtenol and 1-octen-3-ol, which were identified in small quantity in the essential oil, were also identified in aglycone fractions. The same observation was achieved with *C. sempervirens* L., *C. arizonica* Green and *J. communis* L. For examined species of the Cupressus family it is obvious that glycosidically bound volatiles appear independently of essential oil.

3.3. Mass spectra of tentatively identified and unknown compounds in Table 1

11. Sesquiterpenic hydrocarbon (RI 1524/–), MW: 204; *m/z*: 161(100), 91(66), 105(59), 133(58), 204(57), 189(41), 77(37).
21. Sesquiterpenic hydrocarbon (RI 1680/1466), MW: 204; *m/z*: 91(100), 161(95), 105 (71), 77(61), 133(30), 53(23), 204(20).
28. Unknown (RI 1982/–), MW 220: *m/z* 96(100), 109(98), 67(97), 138(76), 43(74), 81(50), 123(48), 164(8), 220(6).
29. E-nerolidol MW (RI 1987/1552), MW: 222; 69(100), 93(95), 107(66), 41(63), 136(31), 161(30), 121(25), 222(2).
30. E3-dodecenyl acetate in mixture (RI 2015/–), MW:226; *m/z*: 43(100), 41(50), 67(47), 82(30), 96(26), 109(16), 138(14), 166(8), 226(6).
31. Oxygen containing sesquiterpene (RI 2015/–), MW: 222; *m/z*: 119(100), 161(82), 106(54), 92(44), 204(44), 147(7), 222(6).
32. T-muurolol (RI 2130/–), MW: 222; *m/z*: 95(100), 43(83), 121(80), 204(62), 161(54), 105(47), 77(45), 139(9), 189(9), 222(7).
33. Oxygen containing sesquiterpene (RI 2139/1604), MW: 222; *m/z*: 161(100), 119(67), 43(55), 105(53), 79(48), 95(41), 204(39), 69(23), 55(19), 133(11), 147(9), 189(9), 222(2).
36. 3,7,11-trimethyl-6,10-dodecadien-1-ol (RI-/1684), MW 224; *m/z* 69(100), 82(73), 95(53), 123(44), 181(24), 109(21), 224(2).
37. Unknown (RI-/1922), MW: 278; *m/z*: 149(100), 41(11), 69(7), 93(6), 55(6), 105(6), 223(5), 205(4), 257(4), 275(3), 278(2).

3.4. Mass spectra of tentatively identified compounds in Table 2

13. 2,3-dimethylphenol (RI 1905/–), MW: 122; m/z: 43(100), 107(66), 77(40), 122(39), 91(36), 134(30).
14. Ethylbenzaldehyde (RI 1922/1826), MW: 134; 107(100), 134(80), 43(67), 77(36), 91(19), 32(19).
17. 2-hydroxy-5-methylacetophenone (RI 2123/1336), MW 150; *m/z*: 135(100), 150(31), 77(30), 107(5).
20. 2,6-dimethoxy-4-(2-propenyl)-phenol (RI–/1531), MW 194; *m/z*: 194(100), 91(59), 119(35), 179(34), 77(33), 147(12), 131(8).

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