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The co-occurrence and systematic significance of pregeijerene B and 8-alpha-acetoxyelemol in *Juniperus*

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Abstract

The identity of a major unknown component in *Juniperus* volatile leaf oils has recently been determined as pregeijerene B. The occurrence of pregeijerene B in *Juniperus* is shown to be highly correlated with 8- α -acetoxyelemol. These compounds are shown to be present in section *Sabina*, but absent in sections *Caryocedrus* and *Juniperus*. In section Sabina, these compounds are very common among the smooth leaf margined, one seeded junipers of the eastern hemisphere and the smooth leaf margined junipers of the western hemisphere. Pregeijerene B and 8- α -acetoxyelemol are thought to arise from 8- α -acetoxyhedycaryol, accounting for their co-occurrence.

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1. Introduction

For several years an unknown compound of M^+ 162 (KI 1274) has been reported (Adams, 1997; 1999; 2000a, b, c, d) in numerous *Juniperus* species. The compound occurs in large amounts (13.2%) in the leaf essential oil of *J. erectopatens* (Cheng and L.L. Fu) R. P. Adams from Yunnan, China (Adams, 1999).

Recently, the structure of this compound has been identified as (E, E, E)-1,7dimethylcyclodeca-1,4,7-triene (1) in *J. erectopatens* and given the common name of pregeijerene B (Cool and Adams, 2003). Pregeijerene B (1) is unusual in having

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three E, E, E double bonds.



A cursory examination of Juniperus leaf oils revealed that pregeijerene B seemed to co-occur with $8-\alpha$ -acetoxyelemol. The purpose of this paper is to examine the distribution of pregeijerene B in *Juniperus* and its co-occurrence with $8-\alpha$ -acetoxyelemol.

2. Materials and methods

The distribution of pregeijerene B and 8- α -acetoxyelemol was taken from previous reports (Adams, 1997; 1999; 2000a,b,c,d; 2001a) in *Juniperus* species. The reader is referred to Adams (2001b) for details on methodology.

3. Results and discussion

The complete listing of all *Juniperus* species is shown in Table 1. Pregeijerene B has an interesting distribution in *Juniperus* (Table 1). Notice that pregeijerene B and 8- α -acetoxyelemol are present in section *Sabina* but absent in all junipers in sections *Juniperus* and *Caryocedrus*. These compounds are present in all the smooth leaved, one seeded junipers of the eastern hemisphere (Table 1). However, they are found in only three two multi-seeded junipers with the highest concentration in *J. erectopatens*. Using RAPD data, Adams (1999) showed that *J. erectopatens* (described as *J. sabina* var. *erectopatens*) is actually not closely related to *J. sabina* but clusters loosely along with *J. procera*. Notice that both J. erectopatens and *J. procera* contain pregeijerene and 8- α -acetoxyelemol (Table 1).

The distribution among the smooth leaf margined junipers of the western hemisphere shows an interesting pattern in these compounds are absent in the Caribbean junipers, except for the anomalous *J. saxicola* of Cuba, but present in all the continental junipers, except *J. horizontalis*. About half of the seventeen serrate leaf margined junipers have pregeigerene B and $8-\alpha$ -acetoxyelemol (Table 1). Juni-

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Table 1

Distribution of pregeijerene B and $8-\alpha$ -acetoxyelemol in *Juniperus* leaf oils. Within subgroups, the species are listed in groupings to reflect relationships based on DNA fingerprinting data (Adams, 1999, 2000a,b,c,d, 2001a)

Species	Pregeijerene B	8-α-Acetoxyelemol
Section sabina		
Smooth leaves, one seeded, eastern hemisphere		
J. carinata (Y. K. Yu & L. K. Fu) R. P. Adams	4.9	2.0
J. convallium Rehder & Wils.	_	_
J. coxii A. B. Jacks.	0.05	0.05
J. indica Bertol.	0.2	0.1
J. komarovii Florin	4.5	2.0
J. microsperma (Cheng & L. K. Fu) R. P. Adams	4.3	1.6
J. morrisonicola Hayata	1.3	4.4
J. pingii W. C. Cheng & Ferre	2.9	1.4
J. prezewalskii Kom.	3.7	3.9
J. pseudosabina Fisch. & C. A. Mey.	0.2	0.05
J. recurva BuchHam. ex. D. Don	2.2	1.9
J. saltuaria Rehder & Wils.	0.9	0.3
J. sauamata BuchHam. ex D. Don	5.5	4.7
J. tibetica Kom.	5.6	3.6
J. wallachiana Hook. f. & Thomson ex Brandis	t	0.8
Smooth leaves multi-seeded eastern homisphere		
L chinansis I		
J. chinensis E.	_	_
J. uuvunuu Fall.	-	-
J. erectoputens (Chelig & L. K. Fu) K. F. Adams	15.2	1.5
J. excelsa M. Bieb., Greece	_	—
J. excelsa MBleb., Georgia (CIS)	-	—
J. joetiaissima Willd.	-	-
J. jarkendensis Kom.	-	_
J. phoenicea L.	-	_
J. polycarpos K. Koch, Armenia	-	_
J. polycarpos K. Koch, Kazakhstan	-	_
J. polycarpos K. Koch, Pakistan	-	_
J. polycarpos K. Koch, Turkmenistan	-	_
J. procera Hochst. ex Endl.	0.3	3.5
J. sabina L.	-	—
J. semiglobosa Regel	_	_
J. semiglobosa var. talassica (Lipinsky) Silba	_	—
J. thurifera L.	0.4	0.3
Smooth leaf margins, western hemisphere		
J. barbadensis L.	_	_
J. bermudiana L.	_	_
J. blancoi Mart.	3.0	6.8
J. gracilior Pilger	_	_
J. gracilior var. ekmanii (Florin) R. P. Adams	_	_
J. gracilior var. urbaniana (Pilger & Ekman) R. P. Adams	_	_
J. lucayana Britton	_	_
J. horizontalis Moench.	_	_
J. mucronata R. P. Adams	2.2	2.3
J. saxicola Britt. & P. Wilson	0.9	3.3
		(continued on next page)

Table 1	(continued)	
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Species	Pregeijerene B	8-α-Acetoxyelemol
J. scopulorum Sarg.	7.6	8.7
J. virginiana L.	5.3	4.9
J. virginiana var. silicicola (Small) E. Murray	3.9	2.7
Serrate leaf margins, western hemisphere		
J. angosturana R. P. Adams	0.05	0.05
J. ashei Buch.	_	_
J. californica Carr.	0.3	0.1
J. coahuilensis (Mart.) Gaussen ex R. P. Adams	0.3	1.3
J. comitana Mart.	_	_
J. deppeana Steudel var. deppeana	_	_
J. durangensis Mart.	_	_
J. flaccida Schlecht, var. flaccida	_	_
J. gamboana Mart.	_	_
<i>J jaliscana</i> Mart	_	_
<i>I monosperma</i> (Engelm) Sarg	2.8	17
<i>I monticola</i> Mart	0.7	2.0
L occidentalis Hook var occidentalis	_	_
<i>L osteosperma</i> (Torr.) Little	_	_
<i>J. ninchotii</i> Sudw	0.1	_
J saltillensis M T Hall	0.4	0.5
J. standleyi Steyer.	_	_
Soction Juningues (= soct_orwoodrus)		
L bravifolia (Soub.) Ant		
J. ordrug Wabh & Parthal	_	_
J. ceurus webb & Berthol.	—	—
J. communis L. val. communis	—	—
J. communis val. saxaniis Pall.	_	_
J. Jormosana Hayata	_	_
J. MICHENSIS KOIUZ.	_	_
J. macrocarpa Sibin. & Sm.	-	-
J. navicularis Grand	-	-
J. oxycearus L.	-	-
J. oxycearus var. baaia H. Gay	-	-
J. rigiaa Siedold & Zucc.	-	_
J. rigida var. conferta Parl.	-	-
J. taxifolia Hook & Arn.	-	-
Section caryocedrus		
J. drupacea Labill.	_	-

perus pinchotii is the only species in this survey that contained pregeijerene but not $8-\alpha$ -acetoxyelemol.

Cool and Adams (2003) postulate that both pregeijerene B and 8- α -acetoxyelemol arise from 8- α -acetoxyhedycaryol leading to their co-occurrence. Pregeijerene B may be found in other *Cupressaceae* species so it is useful to publish the mass spectrum of pregeijerene B (Fig. 1). The Kovat's Index (KI) for pregeijerene B is 1277 on DB-5.



Fig. 1. Quadrupole mass spectrum of pregeijerene B.

The presence of these compounds in section *Sabina*, but absent in sections *Car*yocedrus and Juniperus has phylogenetic significance. Section *Caryocedrus* is thought to be ancestral to section Juniperus and section Juniperus is thought to be ancestral to section sabina (Adams, 2000b). So it seems likely that the enzymes for the synthesis of these compounds arose in section sabina. In section Sabina, these compounds are very common among the smooth leaf margined, one seeded junipers of the eastern hemisphere and the smooth leaf margined junipers of the western hemisphere.

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