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Essential Oils from Leaves of Two Paraguayan Rutaceae: *Zanthoxylum hyemale* A. St. Hil. and *Z. naranjillo* Griseb.

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Abstract

Aerial parts of the species *Zanthoxylum hyemale* and *Zanthoxylum naranjillo* which present the same botanical apparence were subjected to hydrodistillation. Twenty nine compounds have been identified by GC/MS analysis of the essential oils. Sesquiterpenes such as spathulenol and bicyclogermacrene are the main components.

Key Word Index

Zanthoxylum hyemale, *Zanthoxylum naranjillo*, Rutaceae, essential oil composition, linalool, β-elemene, bicyclogermacrene, spathulenol, β-caryophyllene, germacrene D, caryophyllene oxide.

Plant Name

Zanthoxylum hyemale A. St Hil., *Zanthoxylum naranjillo* Grisb.

Source

The plant material, *Zanthoxylum hyemale* A. St. Hil. (syn. *Fagara hyemalis* Engl.) and *Zanthoxylum. naranjillo* Griseb. (syn: *Fagara naranjillo* Engl.) were collected by A. Fournet in February and March 1996 near Piribebuy, Department of Cordillera (Paraguay) and identified by N. Soria (Department of Botany, National University of Asuncion, Paraguay). Voucher specimens (AF 968, AF 980, AF 970 and AF 984) have been deposited at the Herbarium of Chemical Sciences Faculty, Asuncion, Paraguay (FCQ). *Z. hyemale* and *Z. naranjillo* are shrubs or trees (up to 15 m) belonging to the Rutaceae familiy and growing in the tropical South America (Paraguay, North of Argentina, South of Brazil and Uruguay). In Paraguay, *Z. hyemale* and *Z. naranjillo* are respectively called by the Guarani people *kuratu ra*, which means *kuratu* = coriander and *ra* = look like; and *tembetary moroti*, which means *tembé* = lip, *itá* = stone, *y* = abbreviation of *yvyr* = tree, *moroti* = white (bark color) (1). The tea of leaves of both species associated with the hierba maté, *Ilex paraguayensis* St. Hillaire (Aquafoliaceae) is used in Paraguay as painkiller and also as emetic, sudorific and to favor the salivation (2).

Plant part

The fresh leaves of *Z. hyemale* and *Z. naranjillo* were subjected to hydrodistillation in a French-pharmacopea type apparatus respectively to produce oil in 1.67% and 0.27% yield. Then, the obtained oils were dried over anhydrous sodium sulfate.

Previous Work

None.

Present Work

GC/MS analysis was carried out on a ATI UNICAM 610 gas chromatograph combined with a ATI UNICAM 120 mass spectrometer under the following conditions : column BPX5 (25 m x 0.22 µm, 1 µm film thickness) programmed from 60°C (5 min) to 250°C (10 min) at 3°C/min; gas carrier : Helium (15 Psi); ionizing voltage : 70 eV ; injector temperature: 240°C ; detector temperature: 250°C. Four samples were 1/10 diluted

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Table I. Chemical composition of the essential oils from aerial parts of *Zanthoxylum hyemale* and *Zanthoxylum naranjillo*

Component	Kovats indices BPX5	Kovats indices DB-5 (8)	Z. *hyemale* AF 968	Z. *hyemale* AF 980	Z. *naranjillo* AF 970	Z. *naranjillo* AF 984
α-thujene	933	931	t		t	0.1
α-pinene	941	939	0.2	1.0	t	t
sabinene	983	983	t	0.1	t	t
β-pinene	990	980	t	0.8	t	t
β-myrcene	995	991	2.5	t	0.1	0.4
α-phellandrene	1017	1005	t	t	t	t
para-cymene	1037	1026	t	t	0.1	0.3
limonene	1040	1031	t	t	0.1	0.2
β-phellandrene	1043	1031	0.3	t	t	0.1
cis-β-ocimene	1051	1040		0.2	t	
linalool	1112	1098	5.9	t	t	0.5
terpinen-4-ol	1203	1177	t			t
α-terpineol	1219	1189	t	t		
δ-elemene	1352	1339	t	0.8	0.3	0.8
α-cubebene	1360	1351	0.1	0.2	0.2	0.2
α-copaene	1392	1376	1.7	1.6	1.3	0.8
β-elemene	1406	1391	5.2	3.1	8.0	2.9
β-caryophyllene	1443	1418	5.7	2.0	9.8	0.2
β-gurjunene	1450	1432	0.4	0.7	0.3	0.5
aromadendrene	1462	1439		0.3		0.7
α-humulene	1480	1454	1.2	0.7	2.0	0.5
alloaromadendrene	1485	1461	0.4	0.4	0.2	0.3
germacrene D	1510	1480	20.4	24.9	17.2	32.5
bicyclogermacrene	1519	1494	21.1	23.9	16.0	16.4
δ-cadinene	1540	1513	0.6	5.0	0.9	1.7
cis-nerolidol	1573	1534	0.5	0.3		0.2
germacrene D-4-ol	1607	1574		10.4		
spathulenol	1612	1576	19.4	3.8	22.5	19.6
caryophyllene oxide	1619	1581	1.8	0.7	5.7	0.9

Compounds are listed in order of their elution from BPX5 column
t= trace (<0.05 %)

(v/v) in hexane or chloroform before injection (0.3-0.5 µL).

Components identification was based on comparison of the mass spectra and Kovats indice with those given by the literature (8).

The complete chemical composition of these essential oils are rich in sesquiterpenes (near 80%), specially in sesquiterpenic hydrocarbures (near 60%) as can be seen in Table I. Main components of the essential oil of Z. *hyemale* are germacrene D (20-25%) and bicyclogermacrene (20-25%). The percentage of some compounds vary with the samples: spathulenol and δ-cadinene are present at concentrations respectively from 3.8 - 19.4 % and from 0.6 - 5.0 %, germacrene D-4-ol is detected only in one sample.

Main components of essential oils of Z. *naranjillo* vary with the samples, namely germacrene D (17-33 %), spathulenol (20%), bicyclogermacrene (16%), β-caryophyllene (1-10%) and β-elemene (3-8%).

The essential oils of these both species are poor in monoterpenes except one sample of Z. *hyemale* which contains about 6% linalool.

The samples of essential oils from Z. *hyemale* are richer in oxygenated sesquiterpenes. Unfortunately, these results are not sufficient to establish easy rules for the identification of Z. *hyemale* and Z. *naranjillo* species based on essential oil study. It would be necessary to collect more samples at different seasons, location and vegetative life of plants.

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