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Volatile Leaf Oils of Caribbean Myrtaceae. I. Three Varieties of *Pimenta racemosa* (Miller) J. Moore of the Dominican Republic and the Commercial Bay Oil

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ABSTRACT: The foliar essential oils of *Pimenta racemosa* (Miller) J. Moore var. *grisea* (Kiaerskov) Fosb. are dominated by geraniol (0-85.52%), methyl eugenol (0.30-92.60%), and/or trans-methyl isoeugenol (0-86.32%). The foliar essential oils of *Pimenta racemosa* var. *hispaniolensis* (Urban) Landrum are dominated by 1,8-cineole (0.05-37.96%), methyl chavicol (0-22.61%), methyl eugenol (0-63.88%), γ-terpinene (0-16.67%), terpinen-4-ol (0.08-28.98%), and/or thymol (0-44.02%). The foliar essential oils of *Pimenta racemosa* var. *ozua* (Urban & E. Ekman) Landrum are dominated by 1,8-cineole (47.24-55.93%), limonene (3.62-30.07%), and/or α-terpineol (6.68-15.12%). The commercial bay oil (*P. racemosa* var. *racemosa*) is dominated by chavicol (<0.01-15.51%), eugenol (44.41-68.93%), methyl eugenol (0-11.88%), and/or myrcene (0.10-16.17%).

KEY WORD INDEX: *Pimenta racemosa* var. *grisea*, *P. racemosa* var. *hispaniolensis*, *P. racemosa* var. *ozua*, *P. racemosa* var. *racemosa*, Myrtaceae, essential oil composition, 1,8-cineole, geraniol, methyl eugenol, trans-methyl isoeugenol, thymol.

INTRODUCTION: *Pimenta racemosa* (Miller) J. Moore is a Caribbean tree which grows to a height of about 15 m. It is known to exist in five varieties: var. *racemosa*, var. *grisea* (Kiaerskov) Fosb., var. *hispaniolensis* (Urban) Landrum, var. *ozua* (Urban & E. Ekman)

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Landrum, and var. *terebinthina* (Burret) Landrum. The var. *racemosa* is the most widespread of these varieties. It is probably native to the Lesser Antilles (from St. Thomas to Trinidad), Puerto Rico, and Cuba. The var. *grisea* is an understory tree in forests most commonly found at elevations below 800 m in Hispaniola, in Puerto Rico and Tortola. The var. *hispaniolensis*, which is endemic to Hispaniola, can be found at elevations of 950 to 1800 m. The var. *ozua* is endemic to north central Hispaniola, while the var. *terebinthina* is endemic to the Samaná Bay area in the Dominican Republic (1).

The var. *racemosa* is widely cultivated, especially on Dominica, for its leaf oil, known commercially as bay oil or West Indian bay oil. This oil is dominated by eugenol + isoeugenol (33.8-56.2%), myrcene (13.9-31.6%), and chavicol (8.9-21.6%) (2,3,4,5,6). Other reports of commercial bay oils indicated one oil that was rich in methyl eugenol (eugenol methyl ether), β-caryophyllene, and α-humulene (6); and another in which the hydrocarbon portion of the oil (25.7%) was dominated by myrcene (70.0%) and limonene (20.2%) (7).

Further chemical variation in *P. racemosa* is indicated by examination of non-commercial bay oils from the West Indies. Ames et al. (8) indicated leaf oil dominated by either methyl chavicol (estragole) and methyl eugenol or citral (i.e., geranial + neral). McHale et al. (3) examined two non-commercial bay oils: one dominated by methyl eugenol (43.1%), estragole (31.6%), and myrcene (12.0%), while the other oil was dominated by geranial (53.2%) and neral (32.6%).

EXPERIMENTAL: Leaf conditions (fresh or air-dried), origins, and herbarium collector number for vouchers at Delaware State College (DOV), Jardín Botánico Nacional (JBSD),

Table I. Collector numbers, origins and leaf conditions for three varieties of *P. racemosa*

Variety	Collector No.	Origin	Leaf Condition
grisea	Landrum 4734	Bayaguana, Rio Comatillo San Cristobal, D.R.	dried
grisea	Landrum 4736	Bayaguana, Rio Comatillo San Cristobal, D.R.	dried
grisea	Landrum 4737	Bayaguana, Rio Comatillo San Cristobal, D.R.	dried
grisea	Zanoni et al. 38395	Sierra Martin Garcia Azua, D.R.	fresh
hispaniolensis	Landrum 4701	La Vega, Las Aguas Blancas Constanza, D.R.	dried
hispaniolensis	Landrum 4709	Aceitillar, Las Abejas Alcoa Concession, Pedernales, D.R.	dried
hispaniolensis	Zanoni 35309	Arroyo Miguel Martin Azua, D.R.	dried
hispaniolensis	Zanoni 40827A	Arroyo Miguel Martin Azua, D.R.	fresh
ozua	Landrum 4725	Inoa near San Jose de las Matas Santiago, D.R.	dried
ozua	Landrum 4726	Moncion Santiago Rodriguez, D.R.	dried
ozua	Landrum 4733.5	Moncion Santiago Rodriguez, D.R.	dried
ozua	Zanoni et al. 38839	Meseta Abajo Santiago Rodriguez, D.R.	fresh

and/or the New York Botanical Garden (NY) are listed in Table I. Leaves were dried (29-43°C for 1-2 days) and steam distilled, and the oils examined by GC/MS at Delaware State College as previously reported (9) with a modification of the temperature program of 60°C held for one min, to 115°C at 2.5°C per min, then to 210°C at 1.0°C per min and held for 10 min. At Jardín Botánico Nacional, fresh leaves were water distilled for two hours using a modified Clevenger Apparatus. GC/ITMS was performed at Baylor University as previously reported (10) except that the injector temperature was modified to 200°C, and additional library searches were performed with the EPA/NIH mass spectral data base (11).

RESULTS AND DISCUSSION: The essential oils of the three varieties of *P. racemosa* and two commercial bay oils are listed in Tables II-V.

Besides the high concentration of eugenol, myrcene and chavicol expected in the commercial oils, the previous reports of high concentrations of methyl eugenol, β-caryophyllene, α-humulene, limonene, methyl chavicol, geranial and neral in *P. racemosa* (3, 6, 7, 8) were not backed by herbarium vouchers. McHale et al. (3) gave the name of *P. acris* var. *citrifolia*. As a result, the botanical identity of these plants cannot be ascertained, and these reports become meaningless except as an indication of what chemical constituents to expect in foliar oils of the Myrtaceae. Nevertheless, we were able to ascertain that foliar oils of varieties of *P. racemosa* may be dominated by chavicol, 1,8-cineole, methyl chavicol, eugenol, geraniol, limonene, methyl eugenol, myrcene, trans-methyl isoeugenol, γ-terpinene, terpinen-4-ol, α-terpineol and/or thymol.

Table II. Chemical composition of the leaf oils of *P. racemosa* var. *grisea*

Constituent	Landrum 4734	Landrum 4736	Landrum 4737	Zanoni et al. 38395
2-hexenal	-	-	-	<0.01%
benzaldehyde	0.35%	0.19%	0.03%	<0.01
1-octen-3-ol	0.86	0.14	0.31	0.30
3-octanone	2.15	3.02	1.23	0.92
myrcene	-	-	0.71	-
3-octanol	1.92	2.64	2.13	1.50
octanal	0.36	0.23	-	0.31
linalool	-	-	1.68	-
nonanal	-	-	-	0.07
1-octen-3-yl acetate	-	-	-	<0.01
3-octanyl acetate	-	-	-	1.31
methyl chavicol	0.09	0.08	-	0.21
decanal	2.04	1.67	0.33	1.48
neral	-	-	1.06	-
geraniol	-	-	85.52	-
geranial	-	-	1.69	-
trans-anethole	0.38	0.24	-	-
eugenol	-	-	1.47	0.08
geranyl acetate	-	-	1.23	-
methyl eugenol	2.46	2.47	0.30	92.60
β-caryophyllene	0.64	0.35	-	0.37
cis-isoeugenol	0.58	0.36	-	-
cis-methyl isoeugenol	1.18	0.86	-	-
germacrene D	0.16	-	0.46	-
trans-methyl isoeugenol	85.08	86.32	-	0.16
elemicin	-	-	-	<0.01
caryophyllene oxide	-	-	-	0.06

Table III. Chemical composition of the leaf oils of *P. racemosa* var. *hispaniolensis*

Constituent	Landrum 4701	Landrum 4709	Zanoni 35309	Zanoni 40827A
2-hexenal	-	-	-	0.11%
α-thujene	-	-	-	4.04
α-pinene	1.17%	0.46%	0.50%	1.11
camphene	-	-	-	0.12
benzaldehyde	-	0.04	-	-
sabinene	0.19	-	-	1.08
β-pinene	0.35	0.06	0.14	0.55
3-octanone	0.06	0.37	-	0.12
myrcene	0.68	0.26	0.38	4.72
3-octanol	-	0.34	-	-
α-phellandrene	0.43	0.07	0.23	0.44
δ-3-carene	-	-	-	0.23
α-terpinene	1.18	0.04	0.57	3.66
p-cymene	6.81	1.44	2.09	8.59
limonene	4.55	2.24	2.58	0.65
β-phellandrene	-	-	-	0.70
1,8-cineole	37.96	17.57	27.50	0.05
cis-ocimene	-	-	-	0.51
trans-ocimene	-	1.54	-	0.23
γ-terpinene	-	0.20	1.48	16.67
terpinolene	0.48	0.77	0.48	<0.01
α-p-dimethylstyrene	-	-	-	1.06
linalool	0.89	0.30	0.43	0.26
p-mentha-1,3,8-triene	-	-	-	0.08
β-thujone	-	-	-	0.16
cis-pinene hydrate	-	-	-	0.03
cis-sabinene hydrate	-	-	-	0.50
trans-sabinene hydrate	-	-	-	2.55
borneol	-	-	-	0.40
umbellulone	-	-	-	0.17
terpinen-4-ol	28.98	1.73	16.21	0.88
p-cymen-8-ol	0.19	0.13	0.10	0.27
α-terpineol	7.54	0.57	4.22	0.16
methyl chavicol	0.23	5.13	22.61	-
methyl thymol	-	-	-	0.32
chavicol	-	-	0.25	-
trans-anethole	-	-	3.33	-
thymol	-	-	-	44.02
carvacrol	-	-	-	0.95
eugenol	-	0.23	-	0.02
α-copaene	-	-	-	0.10
trans-methyl cinnamate	-	0.73	-	-
methyl eugenol	3.15	63.88	7.08	-
β-caryophyllene	-	-	-	1.54
(Z)-α-trans-bergamotene	-	-	-	0.09
α-humulene	-	-	-	0.19
cis-methyl isoeugenol	-	-	0.15	-
germacrene D	-	-	-	0.35
β-selinene	-	-	-	1.22
RT1627, bp 180, 165?	-	-	-	0.27
α-selinene	-	-	-	0.50
trans-methyl isoeugenol	-	0.75	8.11	-
δ-cadinene	-	-	-	0.02
caryophyllene oxide	-	-	-	0.14

Table IV. Chemical composition of the leaf oils of *P. racemosa* var. *ozua*

Constituent	Landrum 4725	Landrum 4726	Landrum 4733.5	Zanoni et al. 38839
α-thujene	-	-	-	0.10%
α-pinene	2.09%	1.72%	2.17%	1.36
sabinene	0.56	0.40	-	0.83
β-pinene	1.50	1.01	1.13	0.65
3-octanone	-	-	-	<0.01
myrcene	0.86	0.51	0.57	0.45
3-octanol	0.20	0.10	0.09	-
α-phellandrene	0.48	0.12	0.05	0.08
α-terpinene	0.58	0.19	-	0.09
p-cymene	1.80	3.39	2.51	0.71
limonene	9.32	3.62	10.83	30.07
1,8-cineole	55.93	47.24	54.08	50.84
trans-ocimene	-	-	-	<0.01
γ-terpinene	0.94	1.08	0.71	0.74
cis-linalool oxide (furanoid)	-	0.10	-	<0.01
trans-linalool oxide (furanoid)	-	0.14	-	-
terpinolene	0.58	0.11	0.06	0.06
α-p-dimethylstyrene	0.19	-	-	-
linalool	1.81	1.44	0.72	0.51
nonanal	-	-	-	0.06
cis-rose oxide	-	-	-	0.04
cis-pinene hydrate	-	-	-	0.14
cis-sabinene hydrate	-	-	-	<0.01
trans-rose oxide	-	0.14	-	<0.01
trans-pinene hydrate	-	-	-	0.10
trans-sabinene hydrate	-	-	-	<0.01
nonanol	-	-	-	0.05
terpinen-4-ol	5.05	15.67	7.58	4.00
p-cymen-8-ol	0.15	0.32	0.13	0.09
3-decanone	1.90	0.92	1.72	0.53
α-terpineol	14.36	15.12	13.56	6.68
cis-piperitol	-	-	-	<0.01
methyl chavicol	-	0.24	-	-
trans-piperitol	-	-	-	0.06
citronellol	-	0.29	-	0.19
neral	-	-	-	0.06
carvone	-	-	-	<0.01
geranial	-	-	-	0.08
2-undecanone	-	-	0.38	-
9-hydroxy-2-nonanone*	-	-	-	0.14
3-dodecanone	-	-	0.14	-
methyl eugenol	-	0.52	-	0.07
β-caryophyllene	-	-	-	0.12
trans-methyl isoeugenol	-	-	0.56	<0.01
caryophyllene oxide	-	-	-	0.09

*tentative identification

Table V. Chemical composition of two commercial bay oils
(*P. racemosa* **var.** *racemosa*)

Constituent	Sample A by GC/MS	Sample B by GC/ITD
α-thujene	-	<0.01%
α-pinene	0.47%	0.46
camphene	-	0.14
sabinene	-	<0.01
1-octen-3-ol	1.74	-
β-pinene	-	0.09
3-octanone	1.10	-
myrcene	16.17	0.10
3-octanol	0.84	-
α-phellandrene	0.45	0.07
α-terpinene	0.19	0.02
α-p-dimethylstyrene	-	0.30
limonene	3.90	2.07
1,8-cineole	1.38	1.39
cis-ocimene	0.10	0.08
trans-ocimene	1.42	0.08
γ-terpinene	0.16	<0.01
terpinolene	0.24	0.09
p-cymene	0.97	<0.01
linalool	3.63	0.13
camphor	-	<0.01
terpinen-4-ol	0.90	0.05
p-cymen-8-ol	-	<0.01
α-terpineol	0.52	0.14
methyl chavicol	0.03	0.05
chavicol	15.51	<0.01
3-phenyl-2-propenal*	-	0.11
geraniol	0.12	-
geranial	0.22	-
α-cubebene	-	<0.01
eugenol	44.41	68.93
α-copaene	0.41	0.32
β-elemene	-	0.20
methyl eugenol	-	11.88
α-gurjunene	-	0.13
β-caryophyllene	0.64	7.24
α-amorphene	0.39	-
aromadendrene	-	0.09
α-humulene	0.15	1.31
allo-aromadendrene	-	0.28
β-cadinene	-	<0.01
β-chamigrene	-	0.17
β-selinene	-	0.33
α-selinene	-	0.63
α-muurolene	-	0.05
α-farnesene	0.59	-
γ-cadinene	0.15	0.05

Table V (cont.).

Constituent	Sample A by GC/MS	Sample B by GC/ITD
cis-calamenene	0.07	<0.01
δ-cadinene	1.03	0.28
eugenyl acetate	-	0.51
spathulenol	-	0.12
globulol	-	0.36
γ-cadinol	-	<0.01
γ-muurolol	-	<0.01
α-cadinol	-	0.30

*tentative identification

Pimenta racemosa var. *grisea* can be characterized as having foliar oils dominated by geraniol (0-85.52%), methyl eugenol (0.30-92.60%), and/or trans-methyl isoeugenol (0-86.32%). *Pimenta racemosa* var. *hispaniolensis* can be characterized as having foliar essential oils dominated by 1,8-cineole (0.05-37.96%), methyl chavicol (0-22.61%), methyl eugenol (0-63.88%), γ-terpinene (0-16.67%), terpinen-4-ol (0.08-28.98%), and/or thymol (0-44.02%). *Pimenta racemosa* var. *ozua* can be characterized as having foliar oils dominated by 1,8-cineole (47.24-55.93%), limonene (3.62-30.07%), and/or α-terpineol (6.68-15.12%). These contrast with the commercial bay oil (*P. racemosa* var. *racemosa*), which is dominated by chavicol (<0.01-15.51%), eugenol (44.41-68.93%), methyl eugenol (0-11.88%), and/or myrcene (0.10-16.17%).

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