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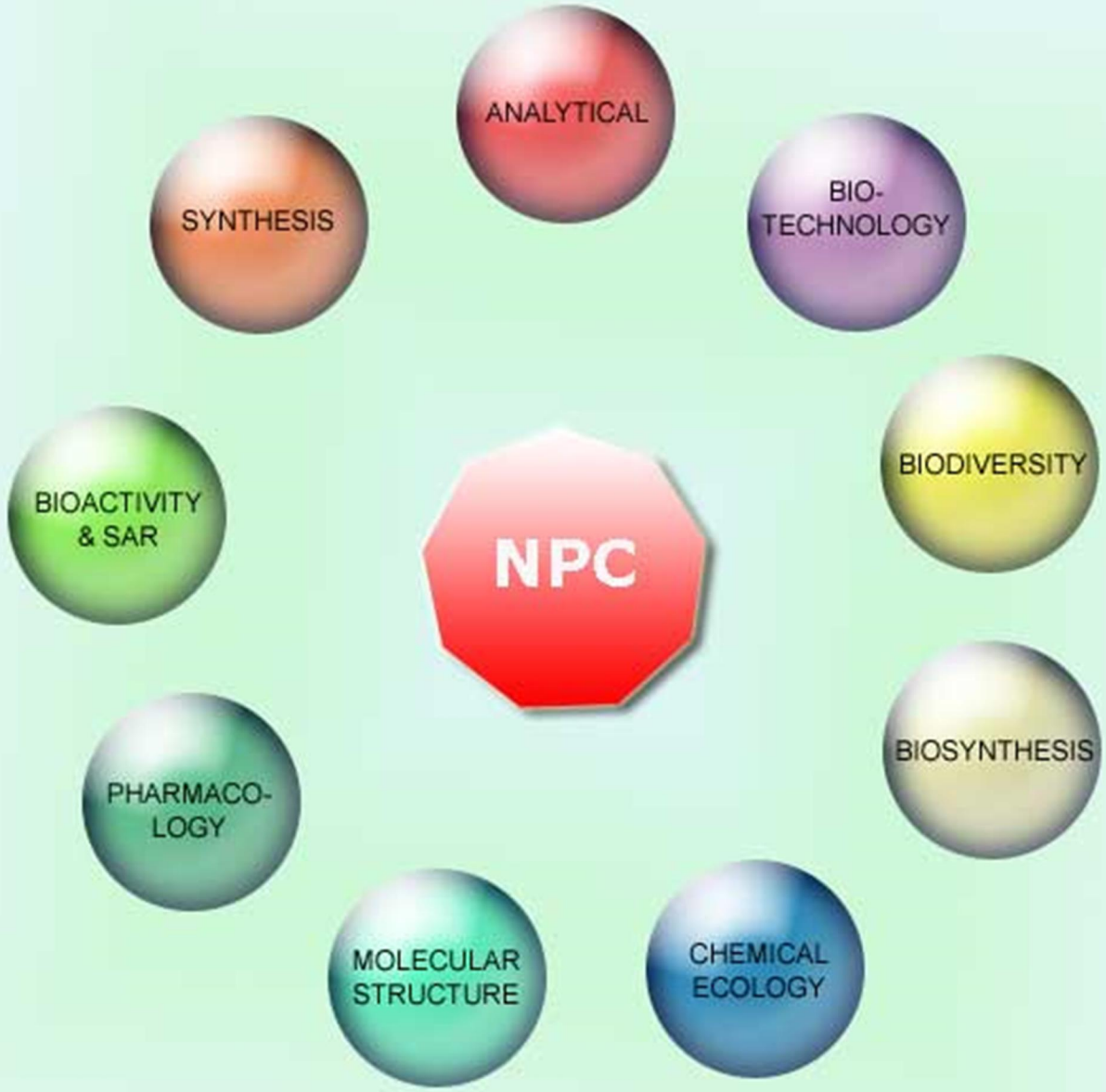

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Pyranocoumarin and Triterpene from *Millettia richardiana*

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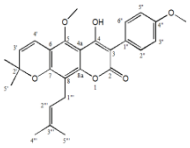
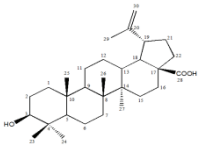
From the stem bark of a Madagascan endemic plant, *Millettia richardiana* Baill., lonchocarpenin and betulinic acid were isolated and their structures established by spectroscopic methods. The analysis of dichloromethane fractions suggested the presence of β-amyrin, lupeol, palmitic acid, linoleic acid and stearic acid. Except for β-amyrin and lupeol, these compounds are described for the first time for the *Millettia* genus.

Keywords: *Millettia richardiana,* Pyranocoumarin, Triterpene, Lonchocarpenin, Betulinic acid.

Millettia is a large genus of Fabaceae (about 100 species). Eight species are represented in Madagascar, of which *M. richardiana* Baill. is used for making furniture [1]. However, the African species showed antiparasitic [2a], anti-inflammatory [2b], molluscicidal [2c], leishmanicidal [2d] and antiplasmodial activities [2e]. Chemical investigation of *Millettia* species has led to the isolation of isoflavonoids [3a], steroids [3b], triterpenes [3c], coumarins [3d], and alkaloids [3e]. Pyridine metabolism in leaves of *M. pinnata* has also been studied [4]. In this paper we report the isolation and identification of a pyranocoumarin, lonchocarpenin (**1**), betulinic acid (**2**), β-amyrin, lupeol, palmitic acid, linoleic acid and stearic acid from the methanolic extract of the stem bark of *M. richardiana*.



Lonchocarpenin (**1**)



Betulinic acid (**2**)

Table 1: NMR spectroscopic data of compound **1** (^{1}H, ^{13}C 400 MHz, $CDCl_3$).

Position	δ_C	δ_H (H, m, *J*)	COSY	HMBC (H => C)
2	162.68			
3	103.93			
4	160.64			
4a	101.71			
5	150.18			
6	110.58			
7	154.67			
8	114.95			
8a	151.52			
2'	77.46			
3'	131.39	5.76, 1H, d (10)	4'	2', 5', 6
4'	115.65	6.50, 1H, d (10)	3'	2', 5, 6, 7
5'	28.10	1.46, 6H, s		2', 3', 5'
1"	123.82			
2", 6"	132.01	7.47, 2H, d (8.8)	3"	3, 4"
3", 5"	113.72	6.95, 2H, d (8.8)	2"	1", 5", 4"
4"	158.84			
1'"	22.06	3.47, 2H, d (7.4)	2'"	7, 8, 8a, 2'", 3'"
2'"	121.37	5.21, 1H, t (7.4)	1'"	
3'"	132.53			
4'"	25.98	1.66, s		2'", 3'", 5'"
5'"	18.27	1.83, s		2'", 3'", 4'"
4-OH		10.03, s		3, 4, 4a
5-OMe	64.55	3.93, s		5
4"-OMe	55.47	3.81, s		4"

[a] Carbon number to which the proton is correlated.

Compound **1**, a yellow powder, showed a molecular pseudo ion at *m/z* 449 suggesting an atomic mass of 448 and the molecular formula $C_{27}H_{28}O_6$. The ^{1}H NMR spectrum showed typical dimethylpyran ring signals at δ_H 6.50 (H-4', d, *J*= 10 Hz), 5.76 (H-3', d, *J*= 10 Hz), and 1.46 (2'-Me_2, s) and 1,4-disubstitued aromatic ring signals at δ_H 7.47 (H-6" and H-2", d, *J*= 8.8 Hz) and 6.95 (H-5" and H-3", d, *J*= 8.8 Hz). The ^{1}H NMR spectrum also revealed the presence of a C-prenyl group from the resonances at δ_H 3.47 (H_2-1"', d, *J*= 7.4 Hz), 5.21 (H-2"', t, *J*= 7.3 Hz), 1.83 (H_3-4"', s) and 1.66 (H_3-5"', s), two methoxyl groups (δ_H 3.93 and 3.81) and the characteristic OH-4 resonance at δ_H 10.03 [5]. The HMBC correlations of H_2-1"' to C-7, C-8, C-8a, C-2"', C-3"' and the proton of a hydroxyl group to C-3, C-4 and C-4a indicated the location of the prenyl group at C-8 on the A ring and confirmed the location of the OH at C-4 on ring B. The correlations of H_3 at δ_H 3.93 to C-5 and H_3 at δ_H 3.81 to C-4" permitted the location of the two methoxyl groups at C-5 and C-4", respectively. The correlations of H-4' to C-5, C-6, C-7, C-2' and H-2"/H-6" to C-3 and C-4" suggested that the chromene ring was connected to C-6 and C-7 on ring A and the aromatic ring to C-3 on the B ring. This compound is lonchocarpenin (**1**) isolated for the first time in 1969 [6]. The proton (δ_H) and carbon (δ_C) chemical shifts of **1** are

reported here for the first time. By concerted use of one and two-dimensional NMR spectroscopy, compound **2** was identified as betulinic acid [7a,b]. However, the assignments of carbons 15 and 21 were not similar to those previously reported [7b]. Therefore, we revised the chemical shifts for C-15 (δ_C 30.26) and C-21 (δ_C 31.03) of betulinic acid. Further components identified were β-amyrin (**4**) [7c], lupeol (**5**) [8], palmitic acid (**6**), linoleic acid (**7**) and stearic acid (**8**) [9].

Experimental

General: NMR, BRÜKER Avance AC400; MS, Waters 2995/2996-Micromass Q-Tof micro spectrometer (ES^+-MS) and Agilent 5975 spectrometer (EI-MS).

Plant materials: *Millettia richardiana* Baill. (Fabaceae) air-dried stem bark, collected in Sahafary, DIANA's Region, Madagascar, was identified by botanists at the Centre National d'Application des Recherches Pharmaceutiques, Antananarivo, Madagascar, where a voucher specimen has been deposited in the Herbarium.

Extraction and isolation: Dried stem bark (1 kg) was extracted with MeOH (3 x 4 L) at room temperature for 3 days each time. After concentration under reduced pressure, the MeOH extract (80 g) was suspended in H_2O (500 mL) and partitioned sequentially using CH_2Cl_2, AcOEt and *n*-BuOH (500 mL x 3) furnishing dichloromethane (21.8 g), ethyl acetate (3.3g), *n*-butanol (18.7 g) and aqueous (30.0 g) extracts, respectively. The CH_2Cl_2 extract (18.0 g) was chromatographed over a silica gel column (160 g), eluting successively with CH_2Cl_2 and mixtures of CH_2Cl_2-CH_3OH (97:3, 95:5 and 90:10), to give 6 fractions, E2A (CH_2Cl_2, 3.8 g), E2B (CH_2Cl_2, 1.1 g), E2C (CH_2Cl_2, 1.6 g), E2DE [CH_2Cl_2-CH_3OH (97:3) 8.5 g], E2F [CH_2Cl_2-CH_3OH (95:5),1.6 g] and E2G [CH_2Cl_2-CH_3OH (90:10), 1.5 g]. Fraction E2B (1.1 g) was submitted to CC over silica gel (25 g), eluted successively with a gradient solvent system of *n*-hexane-CH_2Cl_2 (100:0 →0:100) and a mixture of CH_2Cl_2-CH_3OH (90:10), to give fractions E2B1- E2B6. Fraction E2B4 [*n*-hexane-CH_2Cl_2 (50:50), 207 mg] was subjected to a silica gel column (20 g), eluting with a gradient solvent system of cyclohexane-EtOAc (100:0 →85:15), to obtain compound **1** (85:15, 23.8 mg).

Fraction E2C (1.4g) was chromatographed over a silica gel column (33g), eluting successively with a gradient solvent system of *n*-hexane-CH_2Cl_2 (75:25 →0:100) and mixtures of CH_2Cl_2-CH_3OH (94:6 and 80:20), to provide fractions E2C1-E2C6. Fraction E2C1 [*n*-hexane-CH_2Cl_2 (75:25), (310 mg)] was submitted to flash CC (Easy VarioFlashD17, 10 g) and eluted with a gradient solvent system of cyclohexane-EtOAc (99:1 →95:5) to afford a white solid (98:2, 70.3mg) identified as a mixture of compounds **3** and **4**. Fraction E2C5 [CH_2Cl_2-CH_3OH (94:6), 490 mg] was subjected to flash CC (RediSep, 12g) and eluted with a gradient solvent system of cyclohexane-EtOAc (89:11 →50:50) to give compound **2** (81:19, 79.7 mg).

Fraction E2DE (9 g) was submitted to CC over silica gel (120 g) and eluted successively with a gradient solvent system of *n*-hexane-CH_2Cl_2 (100:0 →0:100) and mixtures of CH_2Cl_2-CH_3OH (94:16 and 80:20) to obtain fractions E2DE1-E2DE7. Fraction E2DE7 [CH_2Cl_2-CH_3OH (80:20), 3 g] was chromatographed over silica gel (90 g), eluting with mixtures of CH_2Cl_2-CH_3OH (100:0, 90:10 and 80:20) to give fractions E2DE7A-E2DE7C. Fraction E2DE7A (100:0, 760mg) was subjected to flash CC (Chromabond, 15 g) and eluted with a gradient solvent system of cyclohexane-EtOAc (100:0 →15:85) to yield a yellow oil (97:3, 40.0 mg) identified as a mixture of compounds 5-7.

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