

A New Hexaoxygenated Xanthone from the Roots of *Moutabea guianensis* Aubl.

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Received: 08 November 2016/Revised: 25 November 2016/Accepted: 23 December 2016

ABSTRACT- Plants of Polygalaceae family are source of several compounds such as xanthenes, coumarins, phenols, triterpenes, steroids, pyrones derivatives and alkaloids. These plants contain chemical compounds with a large spectrum of biological activities, including anti-depressant and anti-angiogenic. *Moutabea guianensis* is an Amazonian species belongs to the Polygalaceae family. In this work, from roots of *M. guianensis* were isolated a new xanthone, 3,8-dihydroxy-1,2,4,5-tetramethoxyxanthone, named moutabeone D, and one known xanthone, 1,3,5-trihydroxy-2-methoxyxanthone. Column chromatography on silica gel and semi-preparative HPLC led the isolation of these compounds. The structures were elucidated by spectroscopic data (HRESIMS, UV, IR, 1D, and 2D NMR).

Key-words- *Moutabea guianensis*, Polygalaceae, Xanthenes

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INTRODUCTION

Moutabea Aublet is a Neotropical genus with approximately 12 recognized species, including *Moutabea floribunda* recently discovery [1]. Several authors already mentioned the need of a taxonomic up-date and revision of this genus [2]. The genus *Moutabea* belongs to the Polygalaceae family which includes about 22 genera and 1300 species [3]. Several phytochemical investigations have revealed that the family is a rich source of xanthenes [4]. Although the chemistry of this family has been widely studied [5-10], only one species, *M. guianensis* has been investigated among the *Moutabea* genus. Studies with the roots of *M. guianensis* led to the identification of xanthenes [11,12]. Ongoing the chemical study on this plant has now resulted in the isolation and identification of one new xanthone identified as 3,8-dihydroxy-1,2,4,5-tetramethoxyxanthone (1), named moutabeone D and the

known xanthone 1,3,5-trihydroxy-2-methoxyxanthone (2) (Fig. 1). In this paper, we described the isolation and structure elucidation of compound 1 and 2.

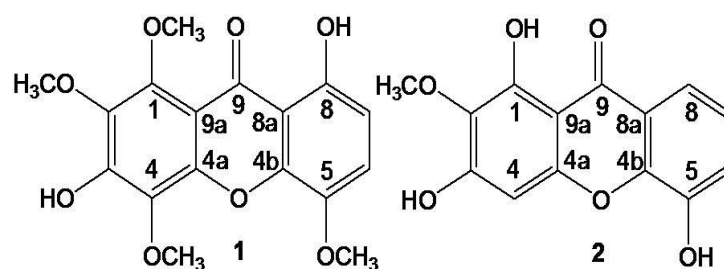


Fig. 1: Structures of compounds 1 and 2 isolated from *Moutabea guianensis*

MATERIALS AND METHODS

This work was carried out at the Laboratory of Chemistry of Natural Products at Federal University of Para, in Belem City, State of Para, Brazil, from March to October 2016.

Chemical reagents and Equipments

UV spectrum was obtained from LC equipped with DAD ProStar 335 (Varian, Palo Alto, CA, USA). IR was carried out on a Shimadzu Prestige 21 (Tokyo, Japan). NMR spectra were recorded on a Varian Mercury-300 NMR Spectrometer, operating at 300 MHz at ¹H and 75 MHz at

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	 DOI: 10.21276/ijlssr.2017.3.1.11

^{13}C , using *d*-chloroform as solvent. HRESIMS was carried out on a Waters Xevo G2-S QToF/Tof spectrometer (Milford, MA, USA). Column chromatography was performed on silica gel 60 (70–230 mesh, Macherey-Nagel, Düren, Germany). Precoated sheets of silica gel with UV254 indicator (thickness 200 μm) were used for TLC (Sorbent technologies, Norcross, GA, USA). Spots were visualized either with a UV lamp (254 nm) or by spraying with aqueous H_2SO_4 (50%) saturated with CeSO_4 solution, followed by heating. Semipreparative HPLC was carried out using a Varian Polaris with UV detector model ProStar 335 using a Phenomenex Gemini (Torrence, CA, USA) C18 column (250 mmx10 mm, 5 μm).

Plant material

Roots and stem of *M. guianensis* were collected on March 2012 in the experimental field of Embrapa Amazônia Oriental in the city Belém, state of Pará, Brazil. A voucher specimen (No. 195862) was deposited in the Herbarium MG of Museu Paraense Emílio Goeldi (Belém-PA, Brazil).

Extraction and isolation

Dry roots of *M. guianensis* (928 g) were ground and successively percolated with hexane (3 L), ethyl acetate (3 L), methanol (3 L) at room temperature for five days each solvent. Solutions were evaporated to dryness under vacuum giving hexane, ethyl acetate and methanol extracts. The ethyl acetate extract (2.0 g) was submitted to CC using silica gel and mixtures of hexane, EtOAc, and MeOH as eluents leading to 20 fractions of 125 mL each. The fractions were combined according to TLC in seven groups (G1-G7). Group G6 (982.3 mg) was purified by semi-preparative HPLC using an isocratic system of $\text{CH}_3\text{CN}-\text{H}_2\text{O}$ 62:38, with a flow rate of 4.7 mL/min yielding **compound 1** (8.1 mg, t_{R} 57.77 min). The stem of *M. guianensis* (2200 g) was subjected to the same process of roots. The ethyl acetate extract (4.9 g) was submitted to CC using mixtures of hexane, EtOAc and MeOH as eluents leading to 26 fractions of 125 mL each. The fractions were combined in eight groups (A1-A8). Group A5 (298.0 mg) was submitted to CC using mixtures of hexane and EtOAc as eluents in increasing order of polarity, collecting 123 fractions of 12 mL. The **compound 2** (6.0 mg) was obtained from fraction 87, eluted with hexane/EtOAc (3:7).

RESULTS AND DISCUSSION

These chemical investigations led to the identification of two compounds. **Compound 2** identified as 1,3,5-trihydroxy-2-methoxyxanthone had already been isolated from other species [13,14] and the compound 1 had reported here as a new compound.

Compound 1 (Fig. 1) was obtained as yellow solid. Its molecular formula was determined to be $\text{C}_{17}\text{H}_{17}\text{O}_8^+$ by HRESIMS exhibiting the quasimolecular ion at m/z 349.0907 $[\text{M}+\text{H}]^+$ (calcd. for $\text{C}_{17}\text{H}_{17}\text{O}_8^+$, 349.0923), which indicated ten degrees of unsaturation. The ^1H NMR showed seven signals, including four singlets attributed to the methoxy hydrogens (δ_{H} 3.95, 3.98, 4.02 and 4.13); two

doublets *ortho*-coupled due the aromatic hydrogens at δ_{H} 6.70 ($J = 9.0$ Hz) and 7.21 ($J = 9.0$ Hz) and one singlet of a bonded hydroxyl group at δ_{H} 12.46. The ^{13}C NMR and DEPT spectra showed sixteen signals, including a conjugated ketone carbonyl signal at δ_{C} 181.4 characteristic for a monochelated carbonyl carbon [15], twelve aromatic carbons signals, eight of them oxygen-substituted (δ_{C} 131.5–154.9) and four oxygen-nonsubstituted (δ_{C} 108.7–120.2). In the same spectrum three methoxyl signals (δ_{C} 57.6, 62.1 and 61.7) were observed and the most intense of them (δ_{C} 61.7) was attributed to two methoxyl groups. The structure of compound 1 was also further deduced from HMBC spectrum. This spectrum showed that the *ortho*-coupled aromatic hydrogen at δ_{H} 7.21 (H-6) was correlated to the carbons of δ_{C} 154.9 (C-8), 139.7 (C-5) and 144.9 (C-4b). In addition, correlation of the methoxyl group (δ_{H} 3.95) with δ_{C} 139.5 (C-5) suggested the methoxyl group was linked to C-5. These correlations revealed that the ring A of 1 was similar to ring A of Angustin A [16]. The spectroscopic data above suggested that ring B had three methoxyl groups, one hydroxyl group as substituent. To confirm the methoxyl groups positions a NOE-diff experiment was carried out, which revealed spacial interactions between δ_{H} 3.98 (1-OCH₃) and δ_{H} 4.13 (2-OCH₃). Irradiation at δ_{H} 4.02 didn't enhance the signals δ_{H} 3.98 and δ_{H} 4.13, which confirm that one methoxyl group is at C-4. NMR data are shown in Table 1. On the basis of these results, the structure of moutabeone D (**1**), was determined as 3,8-dihydroxy-1,2,4,5-tetramethoxyxanthone.

Table 1: ^1H -NMR (300 MHz) and ^{13}C -NMR (75 MHz) spectral data for compound 1 in CDCl_3

Positions	δ_{H} (J in Hz)	δ_{C}	DEPT	HMBC (H→C)
1	-	148.9	C	
2	-	131.5	C	
3	-	148.4 ^a	C	
4	-	137.9	C	
4a	-	147.1 ^a	C	
4b	-	144.9	C	
5	-	139.7	C	
6	7.21 (d, 9.0)	120.2	CH	4b, 5, 8
7	6.70 (d, 9.0)	109.0	CH	5, 8, 8a, 9
8	-	154.9	C	
8a	-	109.2	C	
8b	-	108.7	C	
9	-	181.4	C	
1-OCH ₃	3.98 (s)	62.1	CH ₃	1
2-OCH ₃	4.13 (s)	61.7	CH ₃	2
4-OCH ₃	4.02 (s)	61.7	CH ₃	4
5-OCH ₃	3.95 (s)	57.6	CH ₃	5
3-OH	No observed	-	-	
8-OH	12.46 (s)	-	-	

^a Signals can be interchanged

Spectroscopy data

Compound 1- UV λ_{\max} /nm (acetonitrile-water): 204, 233, 278, 306, 373 (sh). IR (KBr) 3757, 3273, 2935, 2844, 2350, 1957, 1591, 1477, 1350, 1237, 1056, 966, 890, 804, 727 cm^{-1} . ^1H -NMR (300 MHz, CDCl_3) and ^{13}C -NMR (75 MHz, CDCl_3) data, see Table 1. HRESIMS: m/z 349.0907 ($[\text{M}+\text{H}]^+$, $\text{C}_{17}\text{H}_{17}\text{O}_8^+$; calcd. 349.0923).

Compound 2- ^1H -NMR (300 MHz, CDCl_3): δ_{H} 6.54 (s, H-1), 7.35 (dd, $J=7.5$ e 1.8 Hz, H-6), 7.28 (dd, $J=8.0$ e 7.5 Hz, H-7), 7.67 (dd, $J=8.0$ e 1.8 Hz, H-8), 3.88 (s, 2-OCH₃); ^{13}C -NMR (75 MHz, CDCl_3): δ_{C} 155.5 (C-1), 131.7 (C-2), 153.9 (C-3), 94.8 (C-4), 159.3 (C-4a), 146.2 (C-4b), 147.0 (C-5), 121.4 (C-6), 124.9 (C-7), 116.3 (C-8), 121.9 (C-8a), 182.3 (C-9), 104.1 (C-9a), 60.9 (2-OCH₃).

CONCLUSIONS

M. guianensis belonging to the Polygalaceae family is a species with few chemical studies. The chemical study of roots and stem from *M. guianensis* led to isolation of the two more xanthenes for this species. The compounds were isolated using High Performance Liquid Chromatography (HPLC) and Column Chromatography (CC), and structural characterization of compounds was established on the basis of spectroscopic methods, mainly 1D and 2D nuclear magnetic resonance (NMR). One of them 3,8-dihydroxy-1,2,4,5-tetramethoxyxanthone, named moutabeone D, it is a new natural product. The other compound, 1,3,5-trihydroxy-2-methoxyxanthone, was isolated for the first time in this genus. The results presented in this paper showed that *M.* genus is a great natural source of xanthenes and needs further studies.

ACKNOWLEDGEMENT

The authors are grateful to the National Council of Technological and Scientific Development (CNPq), Coordination for the Improvement of Higher Education Personnel (CAPES), Pro-Rector of Research and Post-graduation at the UFPA (PROPESP) and Amazon Foundation for Support at Studies and Research of Pará (FAPESPA) for financial support.

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**How to cite this article:**

Ripardo Filho HS, Pacheco LC, Da Silva EA, Guilhon GMSP, Santos LS: A New Hexaoxygenated Xanthone from the Roots of *Moutabea guianensis* Aubl. *Int. J. Life Sci. Scienti. Res.*, 2017; 3(1): 817-819. DOI:10.21276/ijlssr.2017.3.1.11

Source of Financial Support: CNPq- FAPESPA, **Conflict of interest:** Nil