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Research Article

Monoacyl Monogalactosyl Glycerol from *Cycas lacrimans*

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ABSTRACT
Chemical investigation of the sarcotesta of *Cycas lacrimans*, a plant endemic to the Philippines led to the isolation of monoacylmonogalactosylglycerol (**1**) and 2α,18-dihydroxy-isopimara-7,15-diene (**2**). The structure of **1** was elucidated by extensive 1D and 2D NMR spectroscopy and confirmed by mass spectrometry. The structure of **2** was identified by comparison of its NMR data with those reported in the literature.

Keywords: *Cycas lacrimans*, Cycadaceae, monoacylmonogalactosylglycerol, 2α,18-dihydroxy-isopimara-7,15-diene

INTRODUCTION

Cycas, the only currently known genus of the Family Cycadaceae, are considered as fossil plants though they may have evolved only about 12 million years ago[1]. Their long existence and persistence through time have sparked special interest in their biology and evolution. The cycads resemble palms in morphology thus are commonly called sago palm. They bear naked seeds and are dioecious (male and female as separate individuals). These are widely distributed in the Tropics, with species found in Asia, Africa, Southeast Asia, Pacific, and Australia[2]. They also grow on volcanic, limestone, ultramafic, sandy, or even water-logged soils in grassland and forest habitats[3].

In the Philippines, there are eleven (11) cycad species namely, *C. aenigma* K.D. Hil & Lindstrom, *C. curranii* (J. Schust.) K.D Hill, *C. edentate* de Laub., *C. lacrimans* Lindstrom & K.D. Hill, *C. nitida* K.D. Hill & Lindstrom, *C. riuminiana* Porte ex Regel, *C. saxatilis* K.D. Hill & Lindstrom, *C. sancti-lasallei* Agoo & Madulid, *C. wadei* Merr., *C. vespertilio* Lindstrom & K.D. Hill, and *C. zambalensis* Madulid & Agoo[3-5]. All species, except for *C. edentata*, are endemic to the country[4]. *C. revoluta*, a widely cultivated species, is an introduced species from Japan and Taiwan.

The demand of *Cycas* species for domestic and international horticultural trade, grassland and forest fires of anthropogenic origin, and conversion of their natural habitats to settlements and other land uses have threatened to varying degrees the wild populations of the genus[6]. Some of these species which have been assessed and evaluated in the 2010 IUCN Red List of Threatened Species are *C. curranii*[7], *C. wadei*[8] and *C. zambalensis*as Critically Endangered (CR)[7], *C. riuminiana*as Endangered (E)[7], and *C. saxatilis*as Vulnerable (V)[9].

This study was conducted as part of our research on the chemical constituents of the genus *Cycas* that are endemic and native to the Philippines. We previously reported the isolation of 2-[2-hydroxy-5-(3-hydroxypropyl)-3-methoxyphenyl]-1-(4-hydroxy-3-methoxyphenyl) propane-1,3-diol, phytyl fatty acid ester, lutein, chlorophyll a, and a mixture of stigmasterol and β-sitosterol from the leaflets[10]; squalene, β-sitosterol, stigmasterol, and triglycerides from the sarcotesta[11]; β-sitosterol, stigmasterol, triglycerides, and phytyl fatty acid esters from the endotesta[11]; β-sitosterol, stigmasterol, triglycerides, and β-sitosteryl fatty acid esters from the sclerotesta[11]; and triglycerides and β-sitosteryl fatty acid esters from the bark[11]; and sesamin, 4-hydroxymethyl-3,5-dimethyldihydro-2-furanone, squalene, and phytyl fatty acid ester from the petiole and rachis; and 8-hydroxypinoresinol from the sclerotesta[12] of *C. sancti-lasallei*. Another *Cycas* species we investigated was *C. vespertilio* which yielded pinoresinol (1), sesamin (2), paulownin (3), a mixture of β-sitosterol and stigmasterol, and triacylglycerols from the cone base[13]; pinoresinol, paulownin, β-sitosterol, stigmasterol, triacylglycerols, and lariciresinol from the cataphylls[13]; β-sitosterol from the megasporophyll lamina[13]; β-sitosterol and a mixture oftrans-4-hydroxycinnamate fatty acid esters and cis-4-hydroxycinnamate fatty acid esters from the unripe sarcotesta[13]; β-sitosterol and triacylglycerols from the ripe sarcotesta[13]; pinoresinol, lariciresinol, mixtures of α-amyrin acetate and lupeol acetate in a 2.5:1 ratio and β-sitosterol and stigmasterol in a 2:1 ratio, triglycerides, and

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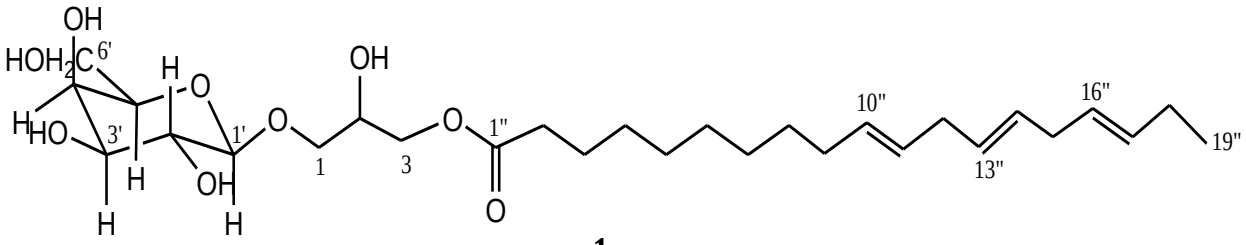

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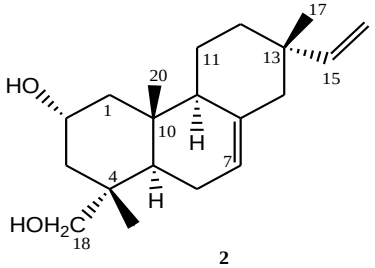

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fatty alcohols from the male cone[14]; 9αH-isopimara-7,15-diene, squalene, β-sitosterol and stigmasterol from the bark[15]; squalene, β-sitosterol and phytyl fatty acid ester from the petiole and rachis[15]; squalene, β-sitosterol, stigmasterol, phytyl fatty acid ester and triglycerides from the endotesta[15]; 9αH-isopimara-7,15-diene, squalene, β-sitosterol, triglycerides and adianenone from the roots[15]; β-sitosterol, stigmasterol, triglycerides and chlorophyll a from the leaflets[15]; and β-sitosterol and triglycerides from the sclerotesta[15]. Recently, we reported the isolation of dihydrodehydrodiconiferyl alcohol, squalene, β-carotene, chlorophyllide a and lutein from the leaflets; squalene, lutein, balanophonin, and β-sitosterol, from the petiole and rachis; β-sitosterol, isopimaran-19-ol, and 3-oxoisopimara-7,15-diene from the bark; dihydrodehydrodiconiferyl alcohol, squalene, β-sitosterol and stigmasterol from the roots; squalene, β-carotene, lutein and β-sitosterol from the sarcotesta; and β-sitosterol from the endotesta of *C. zambalensis*[16]. Another *Cycas* species, *C. edentata* yielded β-sitosteryl-3β-glucopyranoside-6'-*O*-palmitate (**1**) from the sarcotesta; β-sitosteryl fatty acid ester (**2**), unsaturated fatty acid methyl esters (**3**), and a mixture of β-sitosterol (**4a**) and stigmasterol (**4b**) in about 5:1 ratio from the endotesta[17]; chlorophyll a (**5**) from the leaflets; and triacylglycerols (**6**) from the male cone[17]; and 9αH-isopimara-7,15-diene, β-sitosteryl fatty acid ester, and a mixture of β-sitosterol and stigmasterol from the bark[18]; and β-sitosteryl fatty acid ester, and a mixture of β-sitosterol and stigmasterol from the sclerotesta[18]. Recently, we reported the isolation of 2-[2-hydroxy-5-(3-hydroxypropyl)-3-methoxyphenyl]-1-(4-hydroxy-3-methoxy phenyl)propane-1,3-diol, pinoresinol, and fatty alcohols from the leaflets[19]; and triglycerols, and a mixture of β-sitosterol and stigmasterol from the petiole and rachis[19]; squalene, a mixture of β-sitosterol and stigmasterol from the microsporophyll lamina[20]; a mixture of β-sitosterol and stigmasterol**,** triacylglycerols, and a mixture of phytyl fatty acid ester and β-sitosteryl fatty acid ester from the roots[20] of *C. aenigma*.

We earlier reported the isolation of β-sitosteryl-3β-glucopyranoside-6'-*O*-palmitate, a mixture of alkyl 4-hydroxy-*trans*-cinnamate esters and alkyl 4-hydroxy-*cis*cinnamate esters, β-sitosterol, unsaturated triacylglycerols, and methyl fatty acid esters from the sarcotesta[21]; isopimaran-19-ol from the megasporophyll lamina[21]; 9αH-isopimara-7,15-diene and triacylglycerols from the bark[22]; triacylglycerols, oleic acid, and 1,2-dioleylglycerol from the leaflets[19]; triacylglycerols, β-sitosterol, and stigmasterol from the petiole and rachis[19]; β-sitosterol from the roots[22]; and triacylglycerols and β-sitosterol from the endotesta and sclerotesta[22] of *C. lacrimans.* We report herein the isolation of a monoacylmonogalactosyl glycerol (**1**) from the sarcotesta and 2α,18-dihydroxy-isopimara-7,15-diene (**2**) from the megasporophyll lamina of *Cycas lacrimans*. To the best of our knowledge, this is the first report on the isolation of **1** and **2** from *C. lacrimans.*

MATERIALS AND METHODS

General Experimental Procedure

NMR spectra were recorded on a Varian VNMRS spectrometer in $CDCl_3$ at 600 MHz for ^1H NMR and 150 MHz for ^{13}C NMR spectra. Column chromatography was performed with silica gel 60 (70-230 mesh). Thin layer chromatography was performed with plastic backed plates coated with silica gel F_{254} and the plates were visualized by spraying with vanillin/H_2SO_4 solution followed by warming.

Sample Collection

Cycas lacrimans was collected in 2013. Voucher specimens were collected and authenticated by one of the authors (EMGA) and deposited in the De La Salle University-Manila Herbarium (DLSU-M3113).

Isolation

A glass column 18 inches in height and 1.0 inch internal diameter was used for the fractionation of the crude extracts. Ten milliliter fractions were collected. Fractions with spots of the same R_f values were combined and rechromatographed in appropriate solvent systems until TLC pure isolates were obtained. A glass column 12 inches in height and 0.5 inch internal diameter was used for the rechromatography. Five milliliter fractions were collected. Final purifications were conducted using Pasteur pipettes as columns. One milliliter fractions were collected.

Isolation of the chemical constituents of the sarcotesta

The freeze-dried sarcotesta of *C. lacrimans* (140 g) were ground in a blender, soaked in CH_2Cl_2 for 3 days and then filtered. The filtrate was concentrated under vacuum to afford a crude extract (1.2 g) which was chromatographed using increasing proportions of acetone in CH_2Cl_2 at 10% increment. The acetone fraction was rechromatographed

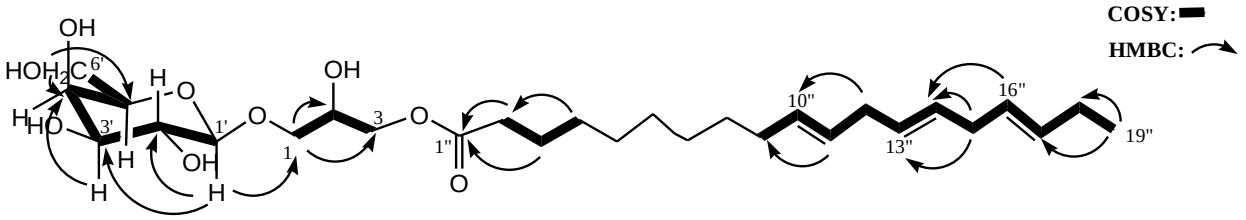

Figure 1: ^{1}H-^{1}H COSY and key HMBC correlations of **1**.

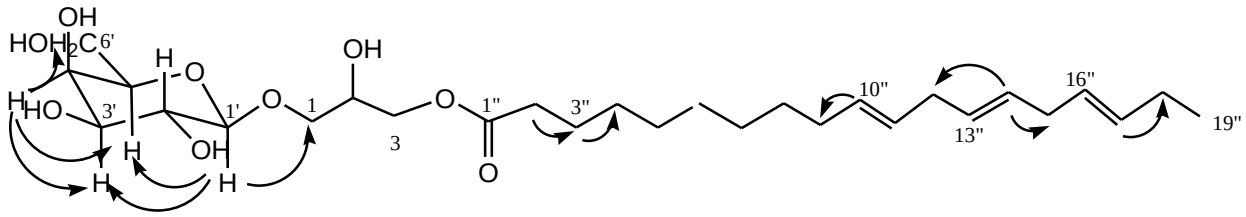

Figure 2: NOESY correlations of **1a**.

in (6 ×) in CH_3CN:Et_2O:CH_2Cl_2 (2.5:2.5:5 by volume ratio) to afford **1** (10 mg) after trituration with petroleum ether.

Isolation of the chemical constituents of the megasporophyll lamina

The air-dried megasporophyll lamina of *C. lacrimans* (26 g) were ground in a blender, soaked in CH_2Cl_2 for 3 days and then filtered. The solvent was evaporated under vacuum to afford a crude extract (0.2 g) which was chromatographed using increasing proportions of acetone in CH_2Cl_2 at 10% increment. The 20% acetone in CH_2Cl_2 fraction was rechromatographed (3 ×) in 15% EtOAc in petroleum ether to afford **2** (4 mg).

Monoacylmonogalactosylglycerol **(1):** ^{1}H NMR ($CDCl_3$, 600 MHz): δ 3.88 (dd, *J* = 11.6, 5.4 Hz, H_a-1), 3.73 (dd, *J* = 11.6, 6.6 Hz, H_b-1), 5.30 (m, H-2), 4.37 (dd, *J* = 12, 1.8 Hz, H_a-3), 4.19 (dd, *J* = 12, 6.6 Hz, H_b-3), 4.26 (d, *J* = 7.2 Hz, H-1'), 3.62 (dd, *J* = 7.2, 9.6 Hz, H-2'), 3.58 (br d, *J* = 9.6 Hz, H-3'), 4.00 (br s, H-4'), 3.53 (t, *J* = 4.8 Hz, H-5'), 3.96 (m, H_a-6'), 3.85 (m, H_b-6'), 2.31 (t, *J* = 7.2 Hz, H_a-2"), 1.56–1.60 (m, H_2-3"), 1.23–1.35 (m, H_2-4"–H_2-8"), 2.05-1.98 (m, H-9"), 5.33–5.39 (m, H-10", H-11", H-13", H-14", H-16", H-17"), 2.79 (t, *J* = 6.6 Hz, H_2-12" or H_2-15"), 2.76 (t, *J* = 6.6 Hz, H_2-12" or H_2-15"), 2.05 (m, H_2-18"), 0.96 (t, *J* = 7.8 Hz, H_3-19"); ^{13}C NMR ($CDCl_3$, 150 MHz): δ 68.46 (C-1), 70.18 (C-2), 62.67 (C-3), 103.95 (C-1'), 71.74 (C-2'), 73.42 (C-3'), 69.55 (C-4'), 74.49 (C-5'), 63.02 (C-6'), 173.49 (C-1"), 34.28 (C-2"), 24.86 (C-3"), 29.76–29.03 (C-4"–8"), 27.19 (C-9"), 131.96–127.10 (C-10", C-11", C-13", C-14", C-16", C-17"), 25.62, 25.52 (C-12", C-15"), 20.54 (C-18"), 14.27 (C-19").

***2α,18-Dihydroxy-isopimara-7,15-diene*(2)**: ^{1}H NMR (600 MHz, $CDCl_3$): δ 0.96, 2.15 (H_2-1), 3.90 (m, H-2), 1.36, 1.66 (H_2-3), 1.40 (H-5), 1.85, 1.88 (H_2-6), 5.34 (m, H-7), 1.76 (H-9), 1.38, 1.58 (H_2-11), 1.38, 1.48 (H_2-12), 1.90, 1.95 (H_2-14), 5.78 (dd, *J*=10.8, 17.4 Hz, H-15), 4.85 (dd, *J*=1.8, 17.4 Hz, H-16), 4.91 (dd, *J*=1.8, 10.8 Hz, H-16), 0.85 (s, H_3-17), 0.91 (s, H_3-19), 3.15 (d, *J*=10.8 Hz, H-18), 3.37 (d, *J*=10.8 Hz, H-18), 0.93 (s, H_3-20). ^{13}CNMR (150 MHz, $CDCl_3$): δ 48.65 (C-1), 65.03 (C-2), 44.82 (C-3), 39.23 (C-4), 42.89 (C-5), 23.20 (C-6), 121.68 (C-7), 135.52 (C-8), 51.76 (C-9), 37.15 (C-10), 20.34 (C-11), 36.01 (C-12), 36.82 (C-13), 45.96 (C-14), 150.51 (C-15), 109.12 (C-16), 21.48 (C-17), 71.69 (C-18), 19.04 (C-19), 16.50 (C-20).

RESULTS AND DISCUSSION

Silica gel chromatography of the dichloromethane extract of *Cycas lacrimans* afforded a monoacylmonogalactosylglycerol (**1**). Its structure was elucidated by extensive 1D and 2D NMR spectroscopy and confirmed by mass spectrometry as follows.

The ^{1}H NMR (see experimental part) and COSY (Fig. 1) spectra of **1** indicated a disubstituted glycerol with oxymethylene proton resonances at δ 3.88 (dd, *J* = 11.6, 5.4 Hz) and δ 3.73 (dd, *J* = 11.6, 6.6 Hz), coupled to the oxymethine proton at δ 5.30 (m), which was in turn coupled to the oxymethylene protons at δ 4.37 (dd, *J* = 12, 1.8 Hz) and 4.19 (dd, *J* = 12, 6.6 Hz). A glycoside was deduced from the resonances at δ 4.26 (d, *J* = 7.2 Hz) for the anomeric proton, coupled to the oxymethine proton at δ 3.62 (dd, *J* = 7.2, 9.6 Hz), which was in turn coupled to another oxymethine proton at δ 3.58 (br d, *J* = 9.6 Hz). The latter proton was also coupled to the oxymethine proton at δ 4.00 (br s), which was further coupled to another oxymethine proton at δ 3.53 (t, *J* = 4.8 Hz), which was finally coupled to the oxymethylene protons at δ 3.96 (m) and 3.85 (m). The large *J* values for the oxymethine protons in the glycoside (H-1', H-2', H-3', H-5') indicated that they are in the axial positions, while the H-4' proton which is a broad singlet is in the equatorial position. Thus, the hydroxyl is in the axial position and **1** contains a galactose. Long chain of unsaturated fatty acid esterified to the glycerol was deduced from the resonances at δ 2.31 (t, *J* = 7.2 Hz) for the α-methylene protons, coupled to the methylene protons at δ 1.56-1.60, which were further coupled to a number of methylene protons at δ 1.23-1.35. These methylene protons were further coupled to allylic methylene protons at δ 2.05-1.98, which were in turn coupled to olefinic protons at δ 5.33-5.39, which were also coupled to the double allylic methylene protons at δ 2.79 (t, *J*= 6.6 Hz) and 2.76 (t, *J*= 6.6 Hz). Integrations of these protons indicated two sets of double allylic methylene protons. Thus, there were three non-conjugated double

bonds in **1**. The latter olefinic proton was further coupled to the allylic methylene protons at δ 2.05, which were in turn coupled to the terminal methyl group at δ 0.96 (t, *J* = 7.8 Hz). Compound **1** is an omega-3 fatty acid which is characterized by a deshielded terminal methyl at δ 0.96 (t, *J* = 7.8 Hz) [19]. The chain length of the major fatty acid esterified to **1** was confirmed by ESI-MS which gave pseudomolecular ion at *m/z* = 551.21 $[M+Na]^+$ corresponding to a molecular formula of$C_{28}H_{48}O_9Na$.

The ^{13}C NMR spectrum (see experimental part) of **1** indicated resonances for the disubstituted glycerol at δ 62.67, 68.48 and 70.18; glycoside at δ 69.55, 71.74, 73.42, 74. 49 for the oxymethine carbons, δ 63.02 for the oxymethylene carbon and δ 109.95 for the anomeric carbon; and fatty acid ester at δ 173.49 for the carboxylate, δ 131.96–127.10 for the olefinic carbons, δ 34.28, 29.76–29.03, 27.19, 25.62, 25.52, 24.86, and 20.58 for the methylene protons, and 14.27 for the methyl carbon of **1**.

Protons attached to carbons were assigned (see experimental part) from HSQC 2D NMR data and the structure of **1** was elucidated by analysis of the HMBC 2D NMR data: key HMBC correlations are shown in Fig. 1. Thus, the presence of the glycerol in **1** was confirmed by the correlations between the oxymethylene protons (H_2-1) and the oxymethine carbon (C-2) and oxymethylene carbon (C-3). The glycoside was attached to the glycerol at C-1 on the basis of long-range correlation between H_2-1 and the anomeric carbon (C-1'). The assignments of the glycoside protons and carbons were confirmed by the HMBC spectrum as follows. Long-range correlations were observed between H-1' and C-2', C-3' and the glycerol (C-1). Further correlations were detected between H-3' and C-1', C-2', C-4', andC-5'. Additional correlations were observed between H_2-6' and C-5 and C-4. The long chain unsaturated fatty acid was esterified to the glycerol at C-3 based on long-range correlations between H-3 and the carbonyl carbon (C-1") of the fatty acid. The assignments of the fatty acid ester protons and carbons in **1** were confirmed by the HMBC spectrum as follows. From the carboxylate end, long-range correlations were observed between H_2-2", H_2-3" and C-1"; and H_2-3", H_2-4" and C-2". From the terminal methyl, H_3-19" correlated with the allylic (C-18") and olefinic (C-17") carbons. The olefinic proton (H-17") correlated with the terminal methyl (C-19"), allylic methylene (C-18"), olefinic (C-16") and double allylic(C-15") carbons. The double allylic protons (H_2-15") correlated with the olefinic carbons (C-17", C-16", C-14" and C-13). The secondset of double allylic protons (H_2-12") correlated with the olefinic carbons (C-14", C-13", C-11" and C-10"). The allylic methylene protons (H_2-9") correlated with olefinic (C-11", C-10"), and methylene (C-8" and C-7") carbons. All long-range correlations observed were consistent with the structure of **1**.

The relative stereochemistry of **1** was deduced from the NOESY spectrum as follows. In the glycoside of **1**, the anomeric proton (H-1') was close in space to the oxymethine protons (H-3', and H-5') and the methylene protons (H_2-1) of the glycerol. Thus, the glycoside was attached to C-1 of the glycerol. The oxymethine proton (H-4') was close in space to two other oxymethine protons (H-3' and H-5') and oxymethylene protons (H_2-6'). Thus, H-4 is in the equatorial position and the hydroxyl is in the axial position, confirming that the monosaccharide in **1** is galactose.In the glycerol of **1**,the oxymethine proton (H-2) was close to the oxymethylene protons (H_2-1 and H_2-3). In the long chain fatty acid of **1**, the α-methylene protons (H_2-2") were close to the methylene protons (H_2-3"), which were in turn close to another set of methylene (H_2-4"). The olefinic protons (H-10", H-11", H-13", H-14", H-16" and H-17") were close to the double allylic (H_2-12" and H_2-15") and allylic (H_2-9" and H_2-18") methylene protons. All NOESY correlations observed are consistent with the relative stereochemistry of **1** as shown in Fig. 2.

Literature search revealed that **1** has similar structure to (2*S*)-1-*O*-linolenoyl-3-*O*-β-D-galactopyranosylglycerol which was previously isolated from *Clinacanthus nutans*[23]. The only difference was the chain length of the attached fatty acid. Whereas, the attached fatty acid in the literature was linolenic acid, **1** has a nineteen carbon fatty acid with double bonds at C-10, C-13 and C-16.

The structure of **2** was identified by comparison of its ^1H NMR and ^{13}C NMR data with those of 2α,18-dihydroxy-isopimara-7,15-diene[24] reported in the literature.

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