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New Myrsinol Diterpenes from *Euphorbia prolifera*

ZHANG, Wan-Jin(张万金) CHEN, Dao-Feng*(陈道峰) HOU, Ai-Jun(侯爱君)

Department of Pharmacognosy, School of Pharmacy, Fudan University, Shanghai 200032, China

Four new myrsinol diterpenes, euphorprolitherins A—D (**1**—**3**, **5**), were isolated from the ethanolic extract of the root of *Euphorbia prolifera*, along with a known one, SPr5 (**4**). The structures were elucidated on the basis of their spectroscopic evidences. The structure and relative stereochemistry of **1** were confirmed by X-ray crystallography.

Keywords *Euphorbia prolifera*, Euphorbiaceae, myrsinol diterpene, euphorprolitherins A—D

Introduction

Euphorbia plants of Euphorbiaceae are well known to contain a large number of diterpene esters, which are the derivatives of tigliane, myrsinol, ingenane, daphane, jatrophane, lathyrane, and abietane diterpenes.[1] The antineoplastic, pro-inflammatory and tumor-promoting activities of *Euphorbia* plants are largely due to the presence of these diterpenoid esters. *Euphorbia prolifera* Buch.-Ham. ex D. Don was used in Chinese folk medicine for the treatment of inflammation and tumors.[2] Previous phytochemical investigations of this plant have led to the identification of five tigliane diterpenes,[2] three myrsinol diterpenes[3-5] and one ergostanol compound.[4] The bioactivity of this plant was focused on the irritant activity.[2] The ethanolic extract of the root exhibits significant inhibitory effect on some cancer cell lines in our study. For the search of antitumor constituents, further chemical research on this plant was carried out by our group. Fractionation of the petrol soluble part of the ethanolic extract resulted in the isolation of four new myrsinol diterpenes (**1**—**3, 5**), namely euphorprolitherins A—D as well as one known myrsinol diterpene SPr5 (**4**).[3] **1** and **2** are diterpenes with a new parent alcohol of 14-desoxo-10,18-dihydromyrsinol.[6] Herein the isolation and structure elucidation of compounds **1**—**3** and **5** were described**.**

Results and discussion

Euphorprolitherin A (**1**) was obtained as colorless cubic crystals. IR absorption at 1714 cm^{-1} indicated the presence of carbonyl groups. The ESIMS of **1** exhibited a quasi-molecular ion $[M+Na]^+$ at *m/z* 773.4. Combining with the analysis of the NMR spectra, the molecular formula was assigned as $C_{38}H_{54}O_{15}$, from which twelve degrees of unsaturation was deduced. The signals at δ 2.10, 2.08, 2.06, 1.98 and 1.96 in the ^1H NMR spectrum showed the occurrence of five methyls protons assignable to five acetyls. The vicinal coupling signals of a quartet at δ 2.40 (J=7.0 Hz, 2H) and a triplet at δ 1.17 (J=7.0 Hz, 3H) indicated one propionyl group. The existence of one 2-methylbutyryl group [δ 2.40 (m,

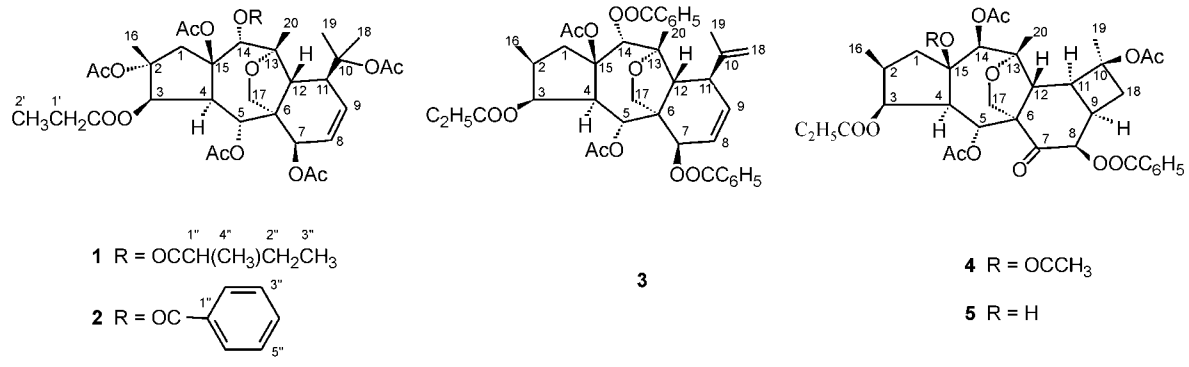

Figure 1 Structures of myrsinol diterpenes isolated from *Euphorbia prolifera*.

* E-mail: dfchen@shmu.edu.cn; Tel.: 86-21-54237453; Fax: 86-21-64042268
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1H), 1.83 (m, 1H), 1.50 (m, 1H), 1.21 (d, J=6.9 Hz, 3H) and 0.96 (t, J=7.5 Hz, 3H)] was also evident. Accordingly, **1** was presumably substituted by seven ester groups. Four oxymethine protons geminal to ester functions [δ 5.89 (dd, J=11.0, 1.4 Hz), 5.57 (s), 5.37 (d, J= 4.0 Hz) and 4.83 (d, J=6.5 Hz)] suggested that the other three ester groups were located at quaternary carbons. Additionally, the signals of four tertiary methyl groups [δ 1.63, 1.52, 1.39 and 1.15 (each 3H, s)], two vicinal olefinic protons [δ 6.16 (ddd, J=10, 6.5, 1.4 Hz) and 5.90 (dd, J=10, 6.0 Hz)] and an oxygenated methylene group [δ 4.09 (d, J=8.8 Hz) and 3.47 (dd, J= 8.8, 1.4 Hz)] were also observed in the ^{1}H NMR spectrum. In the ^{13}C NMR and DEPT spectra of **1**, excluding the signals of seven ester moieties, twenty carbon signals appeared, which supported the result of ^{1}H NMR spectrum. With the consideration of 12 degrees of unsaturation, of which 7 resulted from ester residues, 5 degrees of unsaturation for a tetracyclic skeleton with one double bond were revealed. By comparison of the above facts with those of myrsinol derivatives[7], it is proposed that **1** was different from them by the absence of the typical double bond between C-10 and C-18. This was verified by the chemical shifts of C-10 (δ 85.8) and C-18 (δ 25.3). Thus, **1** was a diterpene heptaester of a new parent alcohol of 14-desoxo-10,18-dihydromyrsinol.

On the basis of 2D NMR spectra (^{1}H-^{1}H COSY, HMQC and HMBC) of **1**, the unambiguous assignment of all protons and carbons were achieved, which also clarified the positions of the ester groups. The following HMBC cross peaks: H-3 (δ 5.37, d, J=4.0 Hz) with the signal of propionyl group (δ 173.6, s); H-5 (δ 5.89, dd, J =11.0, 1.4 Hz) with the acetyl signal at δ 169.2 (s); H-7 (δ 4.83, d, J=6.5 Hz) with the acetyl signal at δ 170.4 (s); and H-14 (δ 5.57, s) with the carbonyl carbon of 2-methylbutyryl (δ 175.4, s) disclosed that the propionate, two acetate, and the 2-methylbutyrate groups were located at C-3, C-5, C-7 and C-14, respectively. Three quaternary carbon signals at δ 87.0 (C-2), 85.8 (C-10), and 90.2 (C-15) directed the other three acetate groups at C-2, C-10, and C-15, respectively.

The relative stereochemistry of **1** was established by the NOESY experiment. The NOE correlations: H-4α with H-3 and H-7 with H-17a, b supported the α-orientations for H-3 and H-7. The NOE correlations: H-1β with Me-16; H-12β with H-5; H-12β with H-14 and H-11α with Me-20 assigned the β-orientations for Me-16, H-5, H-14 and Me-20. Consequently, compound **1** was identified as 14-desoxo-2α,5α,7β,10,15β-*O*-pentaacetyl-3β-*O*-propionyl-14α-*O*-(2-methylbutyryl)10,18-dihydromyrsinol, which was further confirmed by X-ray crystallographic analysis.

Euphorprolitherin B (**2**) was obtained as colorless needles from the mixture of petroleum ether and acetone. The ESIMS of **2** produced a quasi-molecular ion [M+ Na]$^+$ at *m/z* 793.4, suggesting the molecular weight of 770. Combining with the analysis of the NMR spectra, the molecular formula was deduced to be $C_{40}H_{50}O_{15}$.

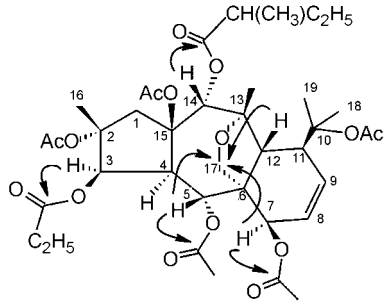

Figure 2 The key HMBC correlations of **1**.

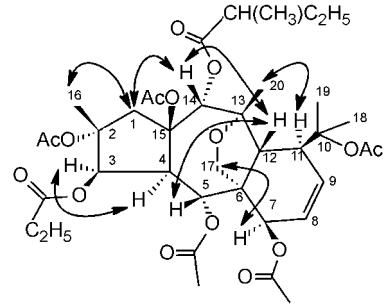

Figure 3 The key NOESY correlations of **1**.

The comparison of the NMR spectra of **1** and **2** disclosed that the 2-methylbutyrate group of **1** was replaced by a benzoate group in **2.** Thus, **2** was characterized as 14-desoxo-2α,5α,7β,10,15β-*O*-pentaacetyl-3β-*O*-propionyl-14α-*O*-benzoyl-10,18-dihydromyrsinol.

Euphorprolitherin C (**3**) was isolated as colorless cubic crystals. The molecular formula of **3** was determined to be $C_{41}H_{46}O_{11}$ on the basis of a quasi-molecular ion [M+Na]$^+$ at *m/z* 737.4 in the ESIMS and NMR spectra. The ^{1}H and ^{13}C NMR spectra of **3** were similar to those of 14-desoxo-3β-*O*-propionyl-5α,14β,15β-*O*-triacetyl-7β-*O*-benzoylmyrsinol (**6**) isolated from *Euphorbia seguieriana*,[7] except for the substitution of a benzoate for an acetate group. A comparison of both ^{1}H NMR spectra indicated that the chemical shift of H-14 moved downfield from δ 5.03 in **6** to δ 5.29 in **3** due to the deshielding effect of a diamagnetic ring current. Consequently, it was deduced that the benzoate group was located at C-14, which was further supported by the comparison of ^{1}H NMR spectrum of **3** with that of 14-desoxo-3β-*O*-propinyl-5α,15β-*O*-diacetyl-7β-*O*-benzoyl-14β-*O*-nicotinoylmyrsinol.[7] For both compounds, the chemical shifts of H-14 showed little difference because of similar deshielding effect of a diamagnetic ring current at C-14, which were δ 5.29 and 5.31, respectively. The relative stereochemistry of **3** was established by the coupling constant values for the derivatives of a parent alcohol of 14-desoxomyrsinol.[7] The values of $^3J_{1\alpha,2}$, $^2J_{1\alpha,1\beta}$, $^3J_{1\beta,2}$, $^3J_{2,3}$, $^3J_{3,4}$, $^3J_{4,5}$, $^3J_{7,8}$, $^3J_{11,12}$ and $^2J_{17a,17b}$ were consistent with the corresponding values of the derivatives of 14-desoxomyrsinol. Therefore, **3** was

deduced to be 14-desoxo-3β-*O*-propionyl-5α,15β-*O*-diacetyl-7β,14β-*O*-dibenzoylmyrsinol, named euphorprolitherin C.

The structure of known compound **4** was identified by comparison of NMR spectra with those of the myrsinol diterpene *Euphorbia* substance SPr5 isolated from the same plant, structure of which was confirmed by single-crystal X-ray analysis.[3]

Euphorprolitherin D (**5**), colorless cubic crystals, was assigned a molecular formula of $C_{36}H_{44}O_{13}$ from its ESIMS and the analysis of the NMR spectra. The ^1H and ^{13}C NMR spectra of **5** exhibited similarities to those of derivatives of cyclomyrsinol[3-5,7-9]. The NMR spectra of **5** lacked the signals of one acetate group compared with those of **4**.[3] The location of ester groups of **5** were deduced by the cross peaks in the HMBC spectrum: propionate carbonyl at δ 173.2 with the proton at δ 5.36 (t, J=4.0 Hz, H-3); benzoate carbonyl at δ 164.1 with the proton at δ 5.54 (d, J=5.8 Hz, H-8); one acetate carbonyl at δ 170.1 with the proton at δ 5.78 (dd, J= 11.2, 1.0 Hz, H-5) and the another one at δ 170.5 with the proton at δ 4.97 (s, H-14).

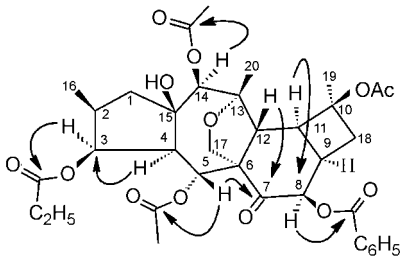

Figure 4 The key HMBC correlations of **5**.

To confirm the relative stereochemistry of **5**, a NOESY experiment was carried out. The cross peaks of H-3 with H-2, H-4α and H-4α with H-14 indicated that H-2, H-3 and H-14 were α-orientated. The cross peak between H-5 and H-12β, together with the coupling constant between H-4 and H-5 (J=11.1 Hz), showed the β-orientation of H-5. Therefore, euphorprolitherin D (**5**) was identified as 3β-*O*-propionyl-5α,10β,14β-*O*-triacetyl-8β-*O*-benzoylcyclomyrsinol.

Experimental

General

Melting points were determined on an X4 micro melting point apparatus and uncorrected. Optical rotations were measured on a JASCO P-1020 polarimeter. IR spectra were measured on a JASCO FT/IR-230 spectrometer with KBr pellets. EIMS data were recorded on a HP5989A spectrometer. ESIMS data were taken on a PE Mariner spectrometer. HRESIMS data were obtained on an AB QSTAR pulsar spectrometer. 1D- and 2D-NMR were run on a Bruker AM-400 instrument with TMS as internal standard. Column chromatography was performed on silica gel (200—300 meshes, Qingdao). Analytical TLC was performed on precoated GF_{254} plates (Yantai).

Plant material

The roots of *Euphorbia prolifera* Buch.-Ham. ex D. Don were collected in Xuanwei，Yunnan Province，People's Republic of China, in October of 1997, and identified by Dr. Chen Dao-Feng, Department of Pharmacognosy, School of Pharmacy, Fudan University, where a voucher specimen is deposited (Chen-971001).

Extraction and isolation

The air-dried roots (6.6 kg) were powered and soaked in 95% ethanol at room temperature for six times. After concentration *in vacuo*, a crude extract (1097 g) was yielded, which was suspended by 800 mL of H_2O. The suspension was extracted with petroleum ether and ethyl acetate, successively. The petroleum ether soluble part afforded 435 g of extract, of which 240 g was subjected to silica gel column chromatography eluted with petroleum ether-ethyl acetate gradient system of increasing polarity (99∶1, 98∶2, 95∶5, 90∶10, 80∶20, and 50∶50) to give 960 fractions. Fractions 877—896 were subjected to silica gel column chromatography with petroleum ether-acetone (8∶2, 7∶3, and 6∶4) yielding **1** (23 mg). Fractions 897—915 were chromatographed over silica gel (petroleum ether-acetone, 8∶2) to afford **2** (17 mg) which were recrystallized from petrol-acetone (2∶1). Fractions 760—814 were repeatedly fractionated by column chromatography on silica gel developed by a gradient mixture of petroleum ether-ethyl acetate (9∶1, 8.5∶1.5, 8∶2, 7.5∶2.5, and 1∶1), followed by petroleum ether-acetone (86∶14) to provide **3**. Recrystallization from MeOH yielded **3** (15 mg). Purification of fractions 871—876 over silica gel (petroleum ether-ethyl acetate 75∶25), followed by the recrystallization from MeOH, resulted in the isolation of **4** (23 mg). Separation of fractions 916—922 by chromatography on silica gel (petroleum ether-ethyl acetate 7∶3) yielded **5** (24 mg).

Euphorprolitherin A (1)

$C_{38}H_{54}O_{15}$, colorless cubic crystals (EtOH); m.p. 192—195 ℃; $[\alpha]_D^{25}$ −27.3 (c 0.11, $CHCl_3$); IR (KBr) ν: 1741, 1370, 1246, 1131, 1098, 1019 cm^{-1}; EIMS *m/z* (%): 649 (8), 630 (4), 589 (2), 570 (7), 529 (3), 510 (4), 497 (2), 451 (2), 437 (3), 377 (5), 293 (27), 43 (100); ESIMS *m/z* (%): 773.4 (100) $[M+Na]^+$; HRMS calcd for $C_{38}H_{54}O_{15}Na$ 773.3360, found 773.3361; ^1H NMR data, see Table 2; ^{13}C NMR data, see Table 1.

Euphorprolitherin B (2)

$C_{40}H_{50}O_{15}$, colorless needles (petroleum ether-acetone); m.p. 184—186 ℃; $[\alpha]_D^{25}$ –100.0 (c 0.09, $CHCl_3$); IR (KBr) ν: 1740, 1480, 1370, 1245, 1100, 1018, 714 cm^{-1}; EIMS *m/z* (%): 710 (1), 669 (6), 650 (5), 609 (2), 590 (9), 530 (4), 470 (3), 456 (5), 105 (85), 43 (100); ESIMS *m/z* (%): 793.4 (98) $[M+Na]^+$; HRMS

Table 1 ^{13}C NMR spectral data of compounds **1**—**3** and **5** (in $CDCl_3$, 100 MHz, δ)

C	**1**	**2**	**3**	**5**	C	**1**	**2**	**3**	**5**
1	46.6 t	47.1 t	43.7 t	50.6 t	OAc				
2	87.0 s	87.0 s	36.7 d	34.7 d		169.8 s	169.4 s	168.8 s	170.5 s
3	78.3 d	78.1 d	76.7 d	78.7 d		169.2 s	169.3 s	167.9 s	170.1 s
4	47.6 d	47.5 d	51.8 d	50.6 d		170.4 s	170.4 s	22.5 q	169.1 s
5	68.6 d	68.6 d	69.1 d	69.3 d		170.8 s	170.7 s	20.7 q	21.6 q
6	53.3 s	53.6 s	54.6 s	62.8 s		168.5 s	168.5 s		21.3 q
7	62.8 d	63.0 d	64.7 d	205.3 d		22.4 q	22.4 q		20.6 q
8	125.9 d	125.9 d	123.3 d	72.2 d		20.9 q	20.9 q		
9	129.9 d	130.0 d	133.6 d	30.5 d		21.0 q	21.0 q		
10	85.8 s	85.9 s	147.0 s	77.9 s		22.5 q	22.5 q		
11	44.7 d	44.7 d	41.8 d	41.3 d		22.3 q	22.3 q		
12	37.1 d	37.1 d	40.7 d	41.7 d	OMBu				
13	90.1 s	89.9 s	89.4 s	88.9 s		175.4 s			
14	72.5 d	73.2 d	81.8 d	82.1 d	1″	40.6 d			
15	90.2 s	90.1 s	90.0 s	81.1 s	2″	26.9 t			
16	18.9 q	18.8 q	14.1 q	14.5 q	3″	11.7 q			
17	70.0 t	69.8 t	69.2 t	66.9 t	4″	15.7 q			
18	25.3 q	25.2 q	112.3 t	35.5 t	OBz				
19	21.4 q	21.3 q	20.7 q	25.1 q			165.8 s	165.7 s	164.1 s
20	24.2 q	24.4 q	24.6 q	21.9 q	1″		130.0 s	129.7 s	130.0 s
OPr					2″, 6″		130.1 d	130.7 d	130.1 d
	173.6 s	173.5 s	174.0 s	173.2 s	3″, 5″		128.4 d	128.3 d	128.5 d
1′	28.0 t	28.0 t	27.7 t	26.3 t	4″		133.4 d	133.2 d	133.3 d
2′	8.8 q	8.8 q	8.8 q	8.8 q				165.6 s	
					1‴			129.6 s	
					2‴, 6‴			129.9 d	
					4‴			127.9 d	
					3‴, 5‴			132.6 d	

* OPr=$OOCC_2H_5$; OMBu=$OOCCH(CH_3)C_2H_5$; OBz=$OOCC_6H_5$

calcd for $C_{40}H_{50}O_{15}Na$ 793.3047, found 793.3036; ^{1}H NMR data, see Table 2; ^{13}C NMR data, see Table 1.

Euphorprolitherin C (3)

$C_{41}H_{46}O_{11}$, colorless cubic crystals (MeOH); m.p. 233—235 ℃; $[\alpha]_D^{25}$ –15.0 (*c* 0.12, $CHCl_3$); IR (KBr) ν: 1740, 1716, 1450, 1369, 1318, 1265, 1223, 1108, 1070, 1021, 908, 714 cm^{-1}; EIMS *m/z* (%): 654 (4), 593 (3), 532 (4), 472 (3), 398 (5), 277 (1), 105 (100); ESIMS *m/z* (%): 737.4 (100) $[M+Na]^+$; HRMS calcd for $C_{41}H_{46}O_{11}Na$ 737.2937, found 737.2930; ^{1}H NMR data, see Table 2; ^{13}C NMR data, see Table 1.

Euphorprolitherin D (5)

$C_{36}H_{44}O_{13}$, colorless cubic crystals (EtOH): m.p. 275—277 ℃; IR (KBr) ν: 2965, 1739, 1458, 1371, 1229, 1177, 1088, 1025, 715 cm^{-1}; EIMS *m/z* (%): 625 (1), 564 (6), 551 (5), 490 (2), 430 (3), 105 (85); ESIMS *m/z* (%): 707.3 (100) $[M+Na]^+$; ^{1}H NMR data, see Table 2; ^{13}C NMR data, see Table 1.

X-ray crystallographic analysis of Euphorprolitherin A (1)

The compound crystallized from ethanol in the orthorhombic group, $C_{38}H_{54}O_{15}$, *P*212121, a=0.8449(4) nm, b=1.0685(4) nm, c=4.2568(9) nm, α=90.00(2)°, β=90.00(3)°, γ=90.00(3)°, V=3.843(2) nm^3, Z=4 and with the calculated density of 1.298 g/cm^3. Crystal size: 0.40 mm×0.40 mm×0.20 mm. Final R indices [$I>2\sigma(I)$]: R_1=0.0478, wR_2=0.1227. R indices (all data): R_1=0.1315, wR_2=0.1615. The structure was solved by direct methods refined by full-matrix least squares techniques. The crystallographic data have been

Table 2 ^{1}H NMR spectral data of compounds **1**—**3** and **5** (in $CDCl_3$, 400 MHz, δ)

H	**1**	**2**	**3**	**5**
1α	3.21 d (17.3)	3.31 d (17.4)	2.86 dd (15.9, 10.9)	2.47 dd (14.8, 9.7)
1β	2.40 d (17.3)	2.37 d (17.4)	2.70 dd (15.9, 9.0)	1.51 dd (15.1, 9.7)
2			2.20 m	2.20 m
3	5.37 d (4.0)	5.41 d (4.0)	5.30 t (3.6)	5.36 t (4.0)
4	3.70 dd (11.0, 4.0)	3.75 dd (11.0, 4.0)	3.22 dd (11.1, 3.6)	2.73 dd (11.2, 3.7)
5	5.89 dd (11.0, 1.4)	5.96 d (11.0)	6.11 dd (11.1, 1.5)	5.78 dd (11.2, 1.0)
7	4.83 d (6.5)	4.85 d (6.5)	5.07 d (6.5)	
8	6.16 ddd (10, 6.5, 1.4)	6.19 ddd (10, 6.5, 1.4)	6.28 ddd (9.7, 6.5, 1.5)	5.54 d (5.8)
9	5.90 dd (10, 6.0)	5.91 dd (10, 6.0)	5.86 dd (9.7, 5.5)	2.76 m
11	3.18 m	3.20 m	3.30 m	2.50 m
12	3.07 d (3.0)	3.20 m	3.49 d (3.8)	4.93 d (12.1)
14	5.57 s	5.83 s	5.29 s	4.97 s
16	1.39 s	1.32 s	0.82 d (6.7)	0.85 d (6.8)
17a	4.09 d (8.8)	4.17 d (8.8)	4.14 d (8.6)	4.20 d (9.5)
17b	3.47 dd (8.8, 1.4)	3.53 dd (8.8, 0.8)	3.62 dd (8.6, 1.5)	3.65 dd (9.5, 1.2)
18	1.63 s	1.64 s	4.86 brs	2.52 m
18			4.79 brs	2.43 m
19	1.52 s	1.55 s	1.89 s	1.67 s
20	1.15 s	1.23 s	1.33 s	1.22 s
OAc				
	2.10 s	2.14 s	2.20 s	2.20 s
	2.08 s	2.10 s	1.96 s	2.20 s
	2.06 s	2.00 s		1.85 s
	1.98 s	1.98 s		
	1.96 s	1.70 s		
OPr				
1′	2.40 q (7.0)	2.37 q (7.5)	2.15 q (7.5)	1.43 dq (16.8, 7.5)
1′				1.14 dq (16.8, 7.5)
2′	1.17 t (7.0)	1.16 t (7.5)	0.96 t (7.5)	0.65 t (7.5)
OMBu				
1″	2.40 m			
2″	1.83 m			
2″	1.50 m			
3″	0.96 t (7.4)			
4″	1.21 d (6.9)			
OBz				
2″ , 6″		8.09 d (7.7)	8.03 d (7.6)	8.34 dd (7.5, 1.3)
4″		7.59 t (7.7)	7.56 t (7.6)	7.62 t (7.5)
3″ , 5″		7.45 t (7.7)	7.38 t (7.6)	7.52 t (7.5)
2‴, 6‴			7.97 d (7.5)	
4‴			7.59 t (7.5)	
3‴, 5‴			7.44 t (7.5)	

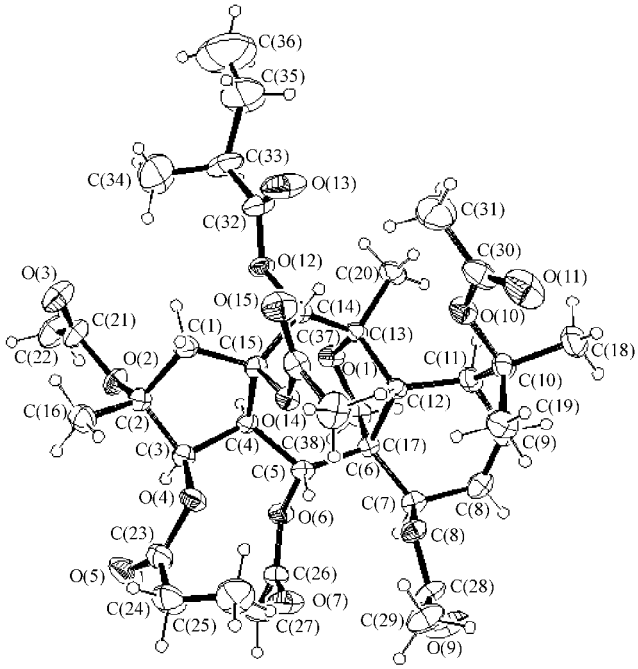

Figure 5 Perspective structure of **1** established by single crystal X-ray analysis.

deposited at the Cambridge Crystallographic Data Center. The coordinates can be obtained, upon request, from the Director, CCDC, 12 Union Road, Cambridge CB2 1EZ, UK. (Fax: +44-(0) 1223-336033 or E-mail: deposit@ccdc.cam.ac.uk).

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