the *trans*-3,6-dialkyl derivatives (*n*-pentyl and *n*-nonyl) prepared by this approach exhibit melting points and ¹H NMR data different from those recorded in the literature (Miura & Nojima, 1980; Miura, Ikegami, Nojima, Kusabayashi, McCullough & Walkinshaw, 1983). Consequently we undertook an X-ray study of the *n*-nonyl derivative obtained by our method in order to confirm its identity. Apart from two disubstituted derivatives, namely the 3,6-diphenyl- (Groth, 1967*a*) and 3,6-dimethoxy-1,2,4,5-tetroxanes (Chiang, Butler & Kuczkowski, 1988), most tetroxanes studied by X-ray crystallography are symmetrically tetrasubstituted (Groth, 1967*b,c,d*; Shulz, Kirsche & Höhne, 1967).

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Structure of 7-Hydroxy-1,6,12(*S*)-triacetoxyneoclerodane-4(18),13(14)-dien-15,16-olide (Grandifolide A): a Diterpene

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Abstract. $C_{26}H_{36}O_9$, $M_r = 492.5$, orthorhombic, $P2_12_12_1$, $a = 17.467$ (3), $b = 17.492$ (3), $c = 8.568$ (2) Å, $V = 2616$ (1) Å^3, $Z = 4$, $D_x = 1.25$ Mg m^{-3}, $\lambda(\text{Cu }K\alpha) = 1.54178$ Å, $\mu = 0.741$ mm^{-1}, $F(000) = 1056$, $T = 293$ K, $R = 0.047$ for 1711 observed reflections. The X-ray study confirms that in the solid state the structure of the title compound is similar to that inferred from chemical and spectroscopic evidence. The Cremer & Pople [*J. Am. Chem. Soc.* (1975), **97**, 1354–1358] ring-puckering parameters for the six-membered rings are $\theta = 7.8$ (4), $\varphi = -127$ (3)°, $Q = 0.598$ (4) Å (ring *A*); $\theta = 6.6$ (4), $\varphi = 79$ (4)°, $Q = 0.548$ (3) Å (ring *B*), indicating a distorted chair conformation in each case. The *A/B* junction is *cis*. The acetoxy, hydroxyl and C(8) methyl groups are equatorial whereas the methyl groups at C(5) and C(9) are axial. The crystal structure is stabilized by an inter-

molecular hydrogen bond between the O(3)—H hydroxyl group and the O(7) carbonyl group, O(3)—H⋯O(7) (*x, y*, 1 + *z*) 2.956 (6) Å.

Experimental. Grandifolide *A* is a naturally occurring diterpene which was isolated from the aerial parts of the plant *Cormutia grandifolia* (Schl. et Cham) Schauer, Verbenacea family. The sample was collected in San Andrés Tuxtla (Estado de Veracruz, Mexico).

The title compound was crystallized from acetone–hexane and gave colourless crystals. Size of crystal 0·16 × 0·26 × 0·36 mm. Nicolet *R*3 four-circle diffractometer, Ni-filtered Cu *Kα* radiation. Lattice parameters from 25 machine-centred reflections with $10.1 < 2\theta < 21.1°$. 1902 reflections with $3 < 2\theta < 110°$ for one octant, 1711 independent with $I > 2.8\sigma(I)$, index range h 0→18, k 0→18, l 0→9, 2θ/θ scan mode, variable scan speed. Two standard reflections (111, 102) monitored every 50 measurements; no significant variation. Lp correction. Data

Table 1. *Fractional atomic coordinates and equivalent isotropic temperature factors*

U_{eq} is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x	y	z	$U_{eq}(\text{Å}^2)$
O(1)	0·3265 (2)	0·1775 (2)	−0·0374 (3)	0·058 (1)
O(2)	0·5191 (1)	0·0170 (1)	0·4764 (3)	0·052 (1)
O(3)	0·5400 (2)	0·1624 (2)	0·6035 (4)	0·069 (1)
O(4)	0·2512 (1)	0·2476 (2)	0·2211 (3)	0·057 (1)
O(5)	0·2037 (5)	0·4736 (4)	0·2444 (14)	0·143 (4)
O(6)	0·2669 (11)	0·5594 (8)	0·4086 (31)	0·233 (10)
O(7)	0·3911 (3)	0·2006 (3)	−0·2527 (5)	0·120 (2)
O(8)	0·4595 (2)	0·0146 (2)	0·7078 (4)	0·081 (1)
O(9)	0·2208 (3)	0·1893 (3)	0·4403 (5)	0·125 (2)
C(1)	0·3799 (2)	0·1194 (2)	−0·0187 (4)	0·052 (1)
C(2)	0·3360 (3)	0·0443 (2)	0·0028 (5)	0·067 (2)
C(3)	0·3768 (3)	−0·0229 (2)	0·0716 (5)	0·065 (2)
C(4)	0·3980 (2)	−0·0038 (2)	0·2369 (5)	0·050 (1)
C(5)	0·4510 (2)	0·0655 (2)	0·2515 (4)	0·045 (1)
C(6)	0·4731 (2)	0·0809 (2)	0·4211 (4)	0·045 (1)
C(7)	0·5179 (2)	0·1546 (2)	0·4452 (5)	0·055 (1)
C(8)	0·4701 (2)	0·2227 (2)	0·3896 (5)	0·051 (1)
C(9)	0·4419 (2)	0·2171 (2)	0·2177 (4)	0·045 (1)
C(10)	0·4041 (2)	0·1367 (2)	0·1898 (4)	0·039 (1)
C(11)	0·3852 (2)	0·2844 (2)	0·1929 (4)	0·047 (1)
C(12)	0·3129 (2)	0·2895 (2)	0·2942 (5)	0·047 (1)
C(13)	0·2853 (2)	0·3704 (3)	0·3115 (6)	0·066 (2)
C(14)	0·3096 (8)	0·4234 (6)	0·0048 (21)	0·108 (6)
C(15)	0·2668 (4)	0·4968 (4)	0·3256 (17)	0·139 (4)
C(16)	0·2099 (8)	0·3921 (6)	0·2056 (18)	0·115 (6)
C(17)	0·5122 (3)	0·2968 (3)	0·4289 (6)	0·085 (2)
C(18)	0·3668 (3)	−0·0409 (2)	0·3554 (6)	0·067 (2)
C(19)	0·5246 (2)	0·0467 (2)	0·1555 (5)	0·059 (1)
C(20)	0·5077 (2)	0·2329 (2)	0·1018 (6)	0·061 (1)
C(21)	0·3361 (3)	0·2089 (3)	−0·1748 (5)	0·070 (2)
C(22)	0·2709 (3)	0·2604 (3)	−0·2165 (6)	0·093 (2)
C(23)	0·5068 (2)	−0·0096 (2)	0·6208 (5)	0·055 (1)
C(24)	0·5578 (3)	−0·0748 (2)	0·6565 (6)	0·071 (2)
C(25)	0·2085 (2)	0·2005 (3)	0·3056 (6)	0·063 (2)
C(26)	0·1468 (3)	0·1660 (3)	0·2155 (6)	0·085 (2)

Table 2 (*cont.*)

C(6)—C(5)—C(19)	109·4 (3)	C(10)—C(5)—C(19)	114·6 (3)
C(5)—C(6)—C(7)	114·1 (3)	C(5)—C(6)—O(2)	108·2 (3)
C(7)—C(6)—O(2)	108·6 (3)	C(6)—C(7)—C(8)	109·5 (3)
C(6)—C(7)—O(3)	110·7 (3)	C(8)—C(7)—O(3)	111·8 (3)
C(7)—C(8)—C(9)	114·8 (3)	C(7)—C(8)—C(17)	109·1 (3)
C(9)—C(8)—C(17)	114·5 (3)	C(8)—C(9)—C(10)	109·5 (3)
C(8)—C(9)—C(11)	106·5 (3)	C(10)—C(9)—C(11)	112·9 (3)
C(8)—C(9)—C(20)	111·2 (3)	C(10)—C(9)—C(20)	112·0 (3)
C(11)—C(9)—C(20)	104·6 (3)	C(1)—C(10)—C(5)	107·7 (3)
C(1)—C(10)—C(9)	115·6 (3)	C(5)—C(10)—C(9)	115·7 (3)
C(9)—C(11)—C(12)	119·5 (3)	C(11)—C(12)—C(13)	112·1 (3)
C(11)—C(12)—O(4)	109·8 (3)	C(13)—C(12)—O(4)	106·3 (3)
C(12)—C(13)—C(14A)	129·0 (7)	C(12)—C(13)—C(14B)	131·3 (6)
C(12)—C(13)—C(16A)	114·8 (5)	C(12)—C(13)—C(16B)	122·0 (5)
C(14A)—C(13)—C(14B)	58·1 (10)	C(14A)—C(13)—C(16A)	116·0 (8)
C(14A)—C(13)—C(16B)	74·7 (8)	C(14B)—C(13)—C(16A)	84·8 (7)
C(14B)—C(13)—C(16B)	106·5 (7)	C(16A)—C(13)—C(16B)	68·4 (8)
C(13)—C(14A)—C(14B)	63·4 (9)	C(13)—C(14A)—C(15)	98·9 (11)
C(13)—C(14A)—O(5B)	92·7 (9)	C(13)—C(14A)—C(16B)	57·3 (7)
C(14B)—C(14A)—C(15)	58·0 (9)	C(14B)—C(14A)—O(5B)	86·8 (9)
C(14B)—C(14A)—C(16B)	98·4 (12)	C(15)—C(14A)—O(5B)	38·0 (5)
C(15)—C(14A)—C(16B)	81·5 (8)	O(5B)—C(14A)—C(16B)	49·3 (6)
C(13)—C(14B)—C(14A)	58·5 (8)	C(13)—C(14B)—C(15)	105·6 (9)
C(14A)—C(14B)—C(15)	72·9 (10)	C(14A)—C(15)—C(16B)	49·0 (9)
C(14A)—C(15)—O(5A)	110·4 (7)	C(14A)—C(15)—O(5B)	81·9 (9)
C(14A)—C(15)—O(6A)	115·6 (15)	C(14A)—C(15)—O(6B)	145·9 (9)
C(14B)—C(15)—O(5A)	92·4 (9)	C(14B)—C(15)—O(5B)	115·1 (8)
C(14B)—C(15)—O(6A)	145·0 (11)	C(14B)—C(15)—O(6B)	121·7 (11)
O(5A)—C(15)—O(5B)	64·6 (8)	O(5A)—C(15)—O(6A)	121·9 (11)
O(5A)—C(15)—O(6B)	102·3 (10)	O(5B)—C(15)—O(6A)	88·5 (13)
O(5B)—C(15)—O(6B)	122·3 (9)	O(6A)—C(15)—O(6B)	49·3 (12)
C(15)—O(5A)—O(5B)	50·4 (8)	C(15)—O(5A)—C(16A)	110·1 (8)
O(5B)—O(5A)—C(16A)	103·2 (9)	C(14A)—O(5B)—C(15)	60·1 (8)
C(14A)—O(5B)—O(5A)	98·6 (9)	C(14A)—O(5B)—O(6A)	87·9 (8)
C(14A)—O(5B)—C(16B)	58·8 (6)	C(15)—O(5B)—O(5A)	65·1 (9)
C(15)—O(5B)—O(6A)	49·2 (10)	C(15)—O(5B)—C(16B)	109·1 (9)
O(5A)—O(5B)—O(6A)	97·8 (11)	O(5A)—O(5B)—C(16B)	92·1 (9)
O(6A)—O(5B)—C(16B)	146·5 (10)	C(15)—O(6A)—O(5B)	42·3 (7)
C(15)—O(6A)—O(16B)	64·4 (15)	O(5B)—O(6A)—O(6B)	97·0 (15)
C(15)—O(6B)—O(6A)	66·3 (12)	C(13)—C(16A)—O(5A)	99·1 (9)
C(13)—C(16A)—C(16B)	50·9 (6)	O(5A)—C(16A)—C(16B)	78·5 (9)
C(13)—C(16B)—C(14A)	48·1 (7)	C(13)—C(16B)—O(5B)	103·7 (9)
C(13)—C(16B)—C(16A)	60·7 (8)	C(14A)—C(16B)—O(5B)	71·9 (7)
C(14A)—C(16B)—C(16A)	93·4 (10)	O(5B)—C(16B)—C(16A)	85·8 (9)
C(1)—O(1)—C(21)	120·3 (3)	O(1)—C(21)—O(7)	124·1 (5)
O(1)—C(21)—C(22)	111· 7 (4)	O(7)—C(21)—C(22)	124·1 (5)
C(6)—O(2)—C(23)	118·5 (3)	O(2)—C(23)—O(8)	124·6 (4)
O(2)—C(23)—C(24)	111·1 (4)	O(8)—C(23)—C(24)	124·3 (4)
C(12)—O(4)—C(25)	119·9 (3)	O(4)—C(25)—O(9)	122·0 (4)
O(4)—C(25)—C(26)	112·6 (4)	O(9)—C(25)—C(26)	124·4 (5)

Table 2. *Bond lengths (Å) and bond angles (°), e.s.d.'s are given in parentheses*

C(1)—C(2)	1·527 (5)	C(1)—C(10)	1·555 (5)
C(1)—O(1)	1·460 (5)	C(2)—C(3)	1·495 (6)
C(3)—C(4)	1·501 (6)	C(4)—C(5)	1·531 (5)
C(4)—C(18)	1·322 (6)	C(5)—C(6)	1·527 (5)
C(5)—C(10)	1·582 (5)	C(5)—C(19)	1·561 (5)
C(6)—C(7)	1·521 (5)	C(6)—O(2)	1·456 (4)
C(7)—C(8)	1·530 (5)	C(7)—O(3)	1·417 (5)
C(8)—C(9)	1·555 (6)	C(8)—C(17)	1·528 (6)
C(9)—C(10)	1·572 (5)	C(9)—C(11)	1·552 (5)
C(9)—C(20)	1·545 (5)	C(11)—C(12)	1·536 (5)
C(12)—C(13)	1·501 (6)	C(12)—O(4)	1·447 (4)
C(13)—C(14A)	1·294 (15)	C(13)—C(14B)	1·357 (11)
C(13)—C(16A)	1·644 (15)	C(13)—C(16B)	1·464 (14)
C(14A)—C(14B)	1·290 (23)	C(14A)—C(15)	1·633 (16)
C(14A)—O(5B)	1·864 (17)	C(14A)—C(16B)	1·678 (17)
C(14B)—C(15)	1·449 (14)	C(15)—O(5A)	1·365 (14)
C(15)—O(5B)	1·159 (15)	C(15)—O(6A)	1·305 (21)
C(15)—O(6B)	1·284 (13)	O(5A)—O(5B)	1·359 (17)
O(5A)—C(16A)	1·466 (14)	O(5B)—O(6A)	1·722 (18)
O(5B)—C(16B)	1·486 (13)	O(6A)—O(6B)	1·080 (30)
C(16A)—C(16B)	1·754 (22)	O(1)—C(21)	1·310 (5)
C(21)—O(7)	1·179 (7)	C(21)—C(22)	1·496 (7)
O(2)—C(23)	1·339 (5)	C(23)—O(8)	1·190 (5)
C(23)—C(24)	1·480 (6)	O(4)—C(25)	1·325 (5)
C(25)—O(9)	1·190 (6)	C(25)—C(26)	1·456 (7)
C(2)—C(1)—C(10)	112·8 (3)	C(2)—C(1)—O(1)	104·3 (3)
C(10)—C(1)—O(1)	110·4 (3)	C(1)—C(2)—C(3)	113·6 (4)
C(2)—C(3)—C(4)	108·4 (3)	C(3)—C(4)—C(5)	113·8 (3)
C(3)—C(4)—C(18)	120·9 (4)	C(5)—C(4)—C(18)	125·0 (4)
C(4)—C(5)—C(6)	111·8 (3)	C(4)—C(5)—C(10)	106·3 (3)
C(6)—C(5)—C(10)	108·1 (3)	C(4)—C(5)—C(19)	106·8 (3)

adjusted to an approximately absolute scale and an overall U value of 0·053 Å², absorption ignored and $R_{int} = 0·026$. Structure solved by direct methods and partial structure expansion by an iterative E Fourier procedure using *SHELXTL* (Sheldrick, 1985). Least-squares refinement of all non-H atoms treated aniso-tropically; O(7), O(9), C(15) show some disorder [see thermal parameters in Table 1; O(5A), O(5B), O(6A), O(6B), C(14A), C(14B), C(16A) and C(16B) have an occupancy factor of 0·50 (2)]; H atoms of CH, CH$_2$ and CH$_3$ [except those on C(14), C(16)] were allowed to ride on bonded C with a fixed isotropic $U = 0·06$ Å². The H atom bonded to an O atom was located in a difference Fourier map at an advanced stage of anisotropic refinement and its coordinate refined. $\sum w(\Delta F)^2$ minimized, $w = [\sigma^2(F_o) + 0·002(F_o)^2]^{-1}$, where σ is standard deviation of observed amplitudes, based on counting statistics; isotropic extinction parameter $X = 0·004$. In the last cycle $(\Delta/\sigma)_{max} = 0·174$; $\Delta\rho$ from $-0·17$ to $0·20$ e Å^{-3}, $S = 1·25$; final $R = 0·047$, $wR = 0·065$;
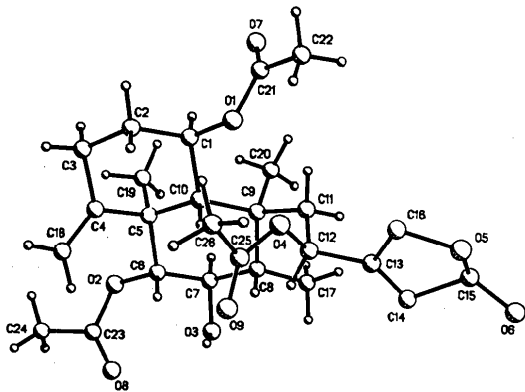
Fig. 1. The molecular structure of the title compound with numbering.

on Tektronix plotter with the *SHELXTL* system of programs.

Atomic coordinates are in Table 1.* A perspective molecular drawing and the atom labelling are displayed in Fig. 1. Bond distances and angles are listed in Table 2.

We thank Mrs Cynthia Lesh de Soriano for technical assistance.

scattering factors from *International Tables for X-ray Crystallography* (1974, Vol. IV). All computations performed on a Nova 4S computer and plots drawn

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(±)-*tert*-Butyl [(Methyl)(1-naphthyl)(phenyl)silyl]acetate

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Abstract. $C_{23}H_{26}O_2Si$, $M_r = 362\cdot6$, triclinic, $P\bar{1}$, $a = 11\cdot275$ (1), $b = 11\cdot585$ (1), $c = 9\cdot682$ (1) Å, $\alpha = 101\cdot91$ (1), $\beta = 99\cdot81$ (1), $\gamma = 119\cdot09$ (1)°, $V = 1025\cdot7$ (6) Å^3, $Z = 2$, $D_x = 1\cdot174$ g cm^{-3}, λ(Cu $K\alpha$) $= 1\cdot54184$ Å, $\mu = 10\cdot89$ cm^{-1}, $F(000) = 388$, $T = 301$ K, $R = 0\cdot038$ for 3157 observations (of 3884 unique data). Si—C distances are 1·859 (1) (methyl), 1·871 (1) (phneyl), 1·879 (1) (naphthyl) and 1·896 (1) Å (*tert*-butyl acetate). No evidence for an interaction between the Si atom and the carbonyl O atom was found.

Experimental. Colorless crystal of (1), dimensions 0·48 × 0·48 × 0·64 mm, mounted in capillary, space group from successful refinement of centrosymmetric model, cell dimensions from setting angles of 25

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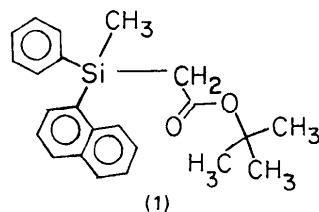
reflections having $32 > \theta > 30°$. Data collection on Enraf–Nonius CAD-4 diffractometer, Cu $K\alpha$ radiation, graphite monochromator, θ–2θ scans designed for $I = 50\sigma(I)$, subject to maximum scan time 120 s, scan rates varied 0·53–5·0° min^{-1}. Data with $2 < \theta < 70°$, $0 \le h \le 13$, $-14 \le k \le 14$, $-11 \le l \le 11$ measured, corrected for background, Lorentz, polarization, absorption by ψ scans, minimum relative transmission coefficient 97·46%.

Standard reflections 500, 030, 003, ±2·4% maximum variation; no decay correction. 3884 unique

(1)