

X-Ray Crystal Structure Determination of a New Alkaloid, Daphnilactone A, $C_{23}H_{35}O_2N$

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The structure of the title compound (I) has been determined from three-dimensional diffractometer data. The structure was determined directly by the symbolic addition procedure, and refined by least-squares methods to R 5.8% for 2014 observed non-zero reflections. The crystals are orthorhombic, space group $P2_12_12_1$ with $Z = 4$ in a unit cell of dimensions: $a = 14.258$, $b = 13.481$, $c = 10.090$ Å. The ring system of this alkaloid is novel but it has a 2-azabicyclo[3.3.1]nonane ring, a common moiety in daphniphyllum alkaloids.

A NEW alkaloid, daphnilactone A* was isolated from *Daphniphyllum macropodum* Miquel.¹ This alkaloid was a minor component of daphniphyllum alkaloids (yield: ca. 0.00001%), and so it could not be converted into a derivative containing a heavy atom and suitable for X-ray analysis. We therefore adopted the direct phase-determination method to establish the structure of daphnilactone A by single-crystal X-ray analysis.

* This alkaloid corresponds to alkaloid C in the preliminary communication.¹

EXPERIMENTAL

Daphnilactone A, m.p. 194.5—195.5 °C, was crystallized from benzene–n-hexane as colourless needles, elongated along the c -axis.

Crystal Data.— $C_{23}H_{35}O_2N$, $M = 357.52$. Orthorhombic, $a = 14.258 \pm 0.003$, $b = 13.481 \pm 0.003$, $c = 10.090 \pm 0.002$ Å, $U = 2095.09$ Å³, $D_m = 1.14$ g cm⁻³ (by flotation), $Z = 4$, $D_c = 1.13$, $F(000) = 784$. Systematic absences:

¹ M. Toda, H. Irikawa, S. Yamamura, and Y. Hirata, *Nippon Kagaku Zasshi*, 1970, **91**, 103.

$h00$ for h odd, $0k0$ for k odd, $00l$ for l odd; space group $P2_12_12_1(D_2^5, \text{No. } 19)$. Cu- K_α radiation, $\lambda = 1.5418 \text{ \AA}$; $\mu(\text{Cu-}K_\alpha) = 5.8 \text{ cm}^{-1}$.

Crystallographic Measurements.—Lattice constants and intensities were measured at 5°C , by use of a Hilger and Watts automatic four-circle diffractometer Y 290 equipped with a scintillation counter and pulse-height analyser.

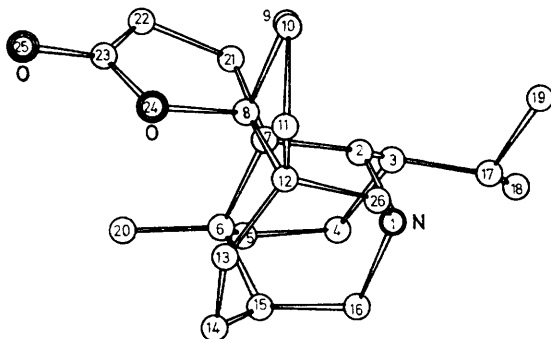


FIGURE 1 The configuration of daphnilactone A

Ni-filtered Cu- K_α radiation was employed. The standard deviation in cell parameters indicated by the precision of the least-squares fit were all $< 0.001 \text{ \AA}$.

The X-ray intensity data were collected by the θ — 2θ moving-crystal—moving-counter technique. The crystal used was so small (square cross-section $0.2 \times 0.2 \times 0.4 \text{ mm}$), that no absorption correction was made. A total of 2268 independent reflections were collected above background ($\theta \leq 70^\circ$), of which 2014 had intensities $> 3\sigma(F)$.² The intensities were corrected for Lorentz and polarization factors, scaled to the absolute level by means of Wilson's method, and normalized structure factor magnitudes ($|E|$ values) were derived.

Structure Analysis.—The structure was solved by a statistical method using the general principles laid down for noncentrosymmetric structures by Karle and Karle.³

In space group $P2_12_12_1$, there are eight possible origins and two enantiomorphs giving sixteen equivalent combinations of phases. It is possible to isolate an origin and enantiomorph by fixing the phases of four two-dimensional reflections. There were four two-dimensional reflections of suitable parity among the larger $|E|$ values, 0,5,6, 4,0,9, 11,0,9, and 17,4,0, and these were assigned the phases $\pi/2$. The phases of a two-dimensional reflection and another were assigned symbols (Table 1).

TABLE 1

Phase assignments for implementing equation (1)

$ E $	h	k	l	ϕ	
3.29	0	5	6	$\pi/2$	Origin assignment (enantiomorph assignment)
4.03	4	0	9	$\pi/2$	
4.51	11	0	9	$\pi/2$	
3.57	17	4	0	$\pi/2$	
3.05	2	2	0	a	
2.61	10	5	2	p	

The formula [equation (1)] for the 199 reflections having ($|E| > 1.6$) was then applied. It was necessary that the

* Q is equivalent to R_K of ref. 3: $Q = \sum_h |E_h - t_h E_h| / \sum_h |E_h|$.

² K. Sasaki and Y. Hirata, *J. Chem. Soc. (B)*, 1971, 1565.

³ J. Karle and I. L. Karle, *Acta Cryst.*, 1966, **21**, 849.

value of the triple product $|E_h E_k E_{h-k}|$ be > 14.0 for a single

$$\phi_h \simeq \langle \phi_k + \phi_{h-k} \rangle_k \quad (1)$$

phase indication to be accepted. As new phases ϕ_h were calculated from combinations $(\phi_k + \phi_{h-k})$, they were added to the list of known phases and subsequently used to determine other phases. From these results, a was assigned 0 or π , and p $\pi/2$ or $-\pi/2$.

The tangent formula³ [equation (2)] was then applied

$$\tan \phi_h \simeq A/B \quad (2)$$

where

$$A = \sum_k |E_k E_{h-k}| \sin(\phi_k + \phi_{h-k})$$

and

$$B = \sum_k |E_k E_{h-k}| \cos(\phi_k + \phi_{h-k})$$

to the 199 reflections having $|E| > 1.6$. The consistency of a phase determined from the tangent formula can be judged from the value of the parameter t_h [equation (3)] which was

$$t_h = (A^2 + B^2)^{1/2} / \sum_k |E_k E_{h-k}| \quad (3)$$

required to be greater than an arbitrary value of 0.4. When this criterion was satisfied, then a phase determined by the tangent formula was subsequently used in next cycle. No changes were made to the phase of a reflection until the conclusion of a cycle, when the tangent formula had been applied to all reflections. After eight cycles, an index C (ref. 4) was printed for each set [equations (4) and (5)].

$$Z_h = A^2 + B^2 \quad (4)$$

$$C = \sum_h Z_h \quad (5)$$

Table 2 shows the values of C and Q ,* and a number of accepted phases for each starting set. An E -map was calcu-

TABLE 2

Comparison of four initial sets

Phases for symbols	$a = 0$ $p = \pi/2$	$a = 0$ $p = -\pi/2$	$a = \pi$ $p = \pi/2$	$a = \pi$ $p = -\pi/2$
C value ($\times 10^4$)	56.5	36.6	67.0	46.7
No. of accepted phases	137	137	157	150
Q value (%)	27	28	27	26

lated using 157 reflections with the phases in the most consistent set ($C = 67 \times 10^4$). Eight peaks separated by chemically reasonable distances and angles were chosen as a trial partial structure. This part of the structure is emphasized by heavy lines in Figure 2. Phases based on this partial structure were used as input for a recycling procedure,⁵ employing the tangent formula with $|E| > 1.2$, and the 19 atoms appeared in an E -map. A Fourier synthesis based on 19 atoms was carried out, and 26 carbon, nitrogen, and oxygen atoms were located.

The initial R value, based on the co-ordinates of the atoms as obtained from this map and overall isotropic thermal parameter of 4.5 \AA^2 , was 30.7%. Refinement proceeded to R 15% with individual isotropic motion, and 9% with 2 cycles of anisotropic refinement. At this stage, 35 hydrogen atom positions were clearly detectable in a

⁴ G. Germain and M. M. Woolfson, *Acta Cryst.*, 1968, **B24**, 91.

⁵ J. Karle, *Acta Cryst.*, 1968, **B34**, 182.

TABLE 3

Final co-ordinates ($\times 10^4$) and thermal parameters ($\times 10^4$)* for the carbon, nitrogen, and oxygen atoms in daphnilactone A

	<i>x</i>	<i>y</i>	<i>z</i>	β_{11}	β_{22}	β_{33}	β_{12}	β_{23}	β_{31}
N(1)	5866(3)	3120(3)	4351(4)	36(2)	41(2)	61(4)	17(4)	04(5)	-08(5)
C(2)	5500(3)	3912(3)	3471(4)	32(2)	45(3)	62(4)	06(4)	09(6)	02(5)
C(3)	5808(3)	3795(4)	2001(5)	40(3)	55(3)	71(5)	14(5)	05(7)	23(6)
C(4)	5268(4)	2994(4)	1229(5)	56(3)	69(4)	84(5)	03(6)	-32(8)	20(7)
C(5)	4231(4)	2898(4)	1636(5)	55(3)	66(4)	71(5)	-11(6)	-37(7)	00(7)
C(6)	4003(3)	3000(3)	3121(4)	37(2)	42(3)	65(4)	-07(4)	-07(6)	-08(6)
C(7)	4426(3)	4011(3)	3627(4)	26(2)	42(3)	52(4)	00(4)	06(6)	06(5)
C(8)	4247(3)	4287(3)	5089(4)	24(2)	44(3)	62(4)	03(4)	01(6)	-06(5)
C(9)	4633(3)	5325(3)	5537(5)	42(3)	44(3)	98(6)	-06(5)	-36(7)	-07(7)
C(10)	4659(4)	5280(5)	7048(5)	56(3)	85(4)	87(6)	-05(7)	-61(9)	-22(7)
C(11)	4653(4)	4165(5)	7444(5)	59(3)	88(4)	63(5)	38(7)	-26(8)	-22(7)
C(12)	4656(3)	3574(4)	6132(4)	42(3)	53(3)	56(4)	21(5)	10(6)	-08(6)
C(13)	4078(4)	2631(4)	6360(5)	58(3)	57(3)	95(6)	07(6)	58(8)	33(8)
C(14)	3975(4)	1862(4)	5214(6)	64(3)	53(3)	107(6)	-18(6)	52(8)	10(8)
C(15)	4433(4)	2099(3)	3883(5)	59(3)	35(3)	99(6)	-04(5)	04(7)	-06(7)
C(16)	5497(4)	2148(4)	4025(5)	49(3)	41(3)	98(6)	10(5)	-07(7)	-10(7)
C(17)	6884(4)	3682(4)	1884(6)	37(3)	76(3)	110(6)	20(6)	13(9)	48(7)
C(18)	7159(5)	3564(7)	0386(7)	73(5)	170(8)	142(9)	22(11)	-06(16)	131(11)
C(19)	7401(4)	4563(5)	2487(8)	35(3)	87(5)	218(11)	-17(6)	08(13)	21(10)
C(20)	2916(3)	2921(4)	3163(6)	36(3)	63(4)	117(6)	-21(5)	-09(8)	-05(7)
C(21)	4070(3)	4926(3)	2837(4)	36(3)	44(3)	71(4)	19(4)	24(6)	10(6)
C(22)	3097(3)	5306(4)	3278(5)	37(2)	54(3)	80(5)	20(5)	21(7)	10(6)
C(23)	2697(3)	4902(4)	4530(5)	32(2)	57(3)	74(5)	22(5)	03(7)	-12(5)
O(24)	3235(2)	4351(2)	5361(3)	27(1)	50(2)	57(3)	11(3)	13(4)	02(3)
O(25)	1886(2)	5036(3)	4858(4)	37(2)	105(3)	89(4)	49(4)	31(7)	18(5)
C(26)	5687(3)	3384(4)	5729(5)	40(3)	64(3)	68(5)	27(5)	03(7)	-14(6)

* Thermal parameters are in the form: $T = \exp[-\beta_{11}h^2 + \beta_{22}k^2 + \beta_{33}l^2 + \beta_{12}hk + \beta_{23}kl + \beta_{31}lh]$.

difference-Fourier map. All parameters were refined with unit weights, and with anisotropic thermal parameters (except hydrogen atoms which were allowed isotropic thermal parameters) by block-diagonal least-squares for three cycles. *R* was reduced to 5.8% for 2014 reflections.

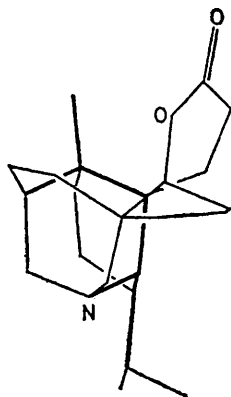


FIGURE 2 Outline of daphnilactone A as it appears projected down the *c*-axis. Atoms connected by heavily shaded lines constitute the initial portion of the structure found by the symbolic addition procedure

The final structure amplitudes are listed in Supplementary Publication No. SUP 20409 (5 pp., 1 microfiche).* The final co-ordinates and thermal parameters for carbon, nitrogen, and oxygen atoms in daphnilactone A are given in Table 3. Table 4 lists the final co-ordinates and thermal parameters for the hydrogen atoms. After the final cycle of refinement, the shifts of all parameters were $< 0.5\sigma$.

* See note about Supplementary Publications in Notice to Authors No. 7, *J. Chem. Soc. (A)*, 1970, Issue No. 20 (items less than 10 pp. are supplied as full size copies).

TABLE 4

Final co-ordinates ($\times 10^3$) and thermal parameters ($\times 10$) for the hydrogen atoms in daphnilactone A. The C-H distances (\AA) are also given

	<i>x</i>	<i>y</i>	<i>z</i>	<i>B</i>	C-H (\AA)
H(1)	591(4)	444(4)	387(6)	44(16)	1.01
H(2)	561(4)	447(5)	147(7)	56(18)	1.09
H(3)	554(5)	227(5)	136(7)	72(21)	1.05
H(4)	522(3)	315(4)	019(5)	19(11)	1.07
H(5)	400(5)	217(5)	127(7)	53(17)	1.10
H(6)	380(4)	347(4)	102(6)	31(14)	1.16
H(7)	528(4)	545(4)	511(6)	46(16)	1.03
H(8)	417(5)	593(5)	518(7)	71(21)	1.12
H(9)	522(5)	566(6)	741(9)	81(23)	1.02
H(10)	399(6)	563(6)	748(9)	96(26)	1.15
H(11)	521(4)	392(5)	806(6)	47(17)	1.06
H(12)	402(4)	399(5)	807(6)	50(17)	1.12
H(13)	444(5)	231(5)	718(7)	65(20)	1.07
H(14)	340(5)	290(5)	660(7)	60(20)	1.06
H(15)	428(5)	127(5)	549(7)	74(23)	0.95
H(16)	327(4)	173(4)	504(6)	42(16)	1.03
H(17)	429(5)	149(5)	325(7)	67(21)	1.06
H(18)	580(4)	194(4)	312(6)	43(16)	1.05
H(19)	572(4)	170(4)	474(6)	40(15)	0.99
H(20)	708(4)	309(4)	250(5)	46(16)	1.05
H(21)	457(4)	551(5)	294(7)	60(20)	1.07
H(22)	394(4)	484(4)	176(6)	37(15)	1.11
H(23)	311(3)	602(4)	341(5)	20(11)	0.97
H(24)	260(4)	517(4)	250(6)	29(13)	1.08
H(25)	616(4)	392(4)	599(6)	32(14)	1.02
H(26)	595(4)	281(4)	627(6)	49(17)	1.02
H(27)	698(5)	282(5)	015(8)	72(22)	1.07
H(28)	789(5)	360(5)	028(8)	78(23)	1.04
H(29)	682(5)	417(6)	-011(8)	95(25)	1.08
H(30)	810(5)	455(5)	217(7)	61(20)	1.04
H(31)	750(5)	443(5)	360(7)	63(20)	1.14
H(32)	729(5)	519(6)	182(8)	82(25)	1.10
H(33)	263(5)	350(5)	255(7)	64(20)	1.08
H(34)	283(5)	213(5)	283(7)	63(20)	1.13
H(35)	252(5)	305(5)	418(7)	59(20)	1.18

RESULTS AND DISCUSSION

The present X-ray analysis has established the molecular structure of daphnilactone A as (I). The skeleton

is novel except for a 2-azabicyclo[3,3,1]nonane and a cyclopentane ring, which are common in daphniphyllum

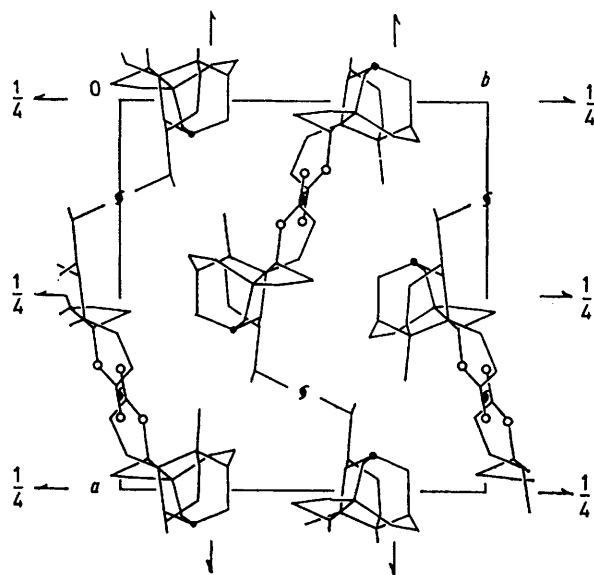
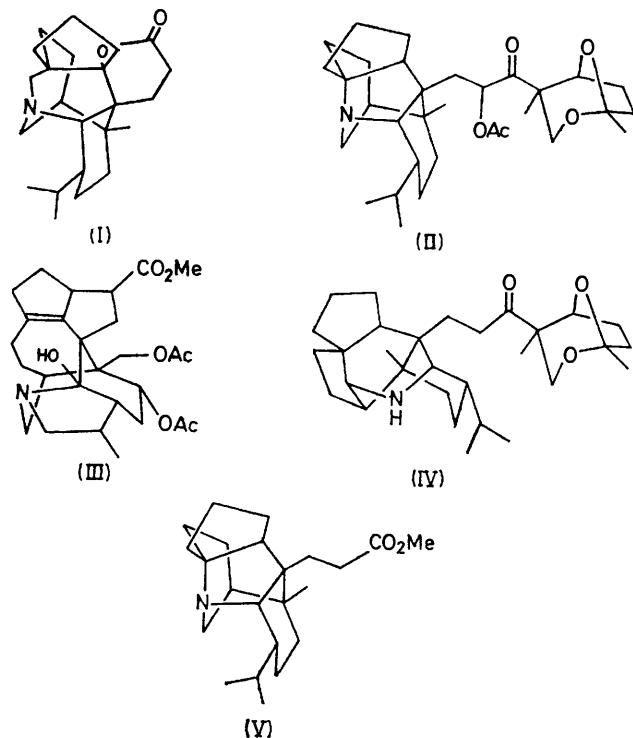


FIGURE 3 Contents of the unit cell viewed down the *c*-axis

alkaloids. Generally from a structural point of view, daphniphyllum alkaloids can be classified into three



groups: the daphniphylline (II),⁶ yuzurimine (III),⁷ and secodaphniphylline (IV)² groups. This alkaloid belongs to a fourth group and is characterized by the new C(26) carbon and the spiro-lactone by comparison

⁶ N. Sakabe, H. Irikawa, H. Sakurai, and Y. Hirata, *Tetrahedron Letters*, 1966, 963.

with the daphniphylline group [*e.g.* methyl homo-daphniphyllate (V)⁸].

The spectral data indicate that (I) has an isopropyl group [$\delta(\text{CDCl}_3)$ 0.91, 0.93 p.p.m., each 3H], a tertiary methyl group (δ 1.05, 3H), and a lactone ring [ν_{max} (KBr) 1735 cm^{-1} (C=O)]; these results show that atoms (24) and (25) are oxygen. The interatomic distances and valency angles are listed in Table 5. The mean estimated standard deviation of bond distances was 0.006 Å, and of the valency angle was 0.6°. The mean value of

TABLE 5
Bond lengths and angles in daphnilactone A

(a) Distances (Å)			
N(1)–C(2)	1.483	C(9)–C(10)	1.526
N(1)–C(16)	1.450	C(10)–C(11)	1.555
N(1)–C(26)	1.458	C(11)–C(12)	1.545
C(2)–C(3)	1.555	C(12)–C(13)	1.532
C(2)–C(7)	1.546	C(12)–C(26)	1.546
C(3)–C(4)	1.537	C(13)–C(14)	1.559
C(3)–C(17)	1.546	C(14)–C(15)	1.527
C(4)–C(5)	1.540	C(15)–C(16)	1.526
C(5)–C(6)	1.539	C(17)–C(18)	1.570
C(6)–C(7)	1.576	C(17)–C(19)	1.526
C(6)–C(15)	1.563	C(21)–C(22)	1.543
C(6)–C(20)	1.553	C(22)–C(23)	1.490
C(7)–C(8)	1.542	C(23)–O(24)	1.358
C(7)–C(21)	1.554	C(23)–O(25)	1.216
C(8)–C(9)	1.570		
C(8)–C(12)	1.540		
C(8)–O(24)	1.472		

(b) Angles (deg.)			
C(2)–N(1)–C(16)	112.8	C(9)–C(8)–O(24)	103.7
C(2)–N(1)–C(26)	109.5	C(12)–C(8)–O(24)	106.2
C(16)–N(1)–C(26)	111.9	C(8)–C(9)–C(10)	105.1
N(1)–C(2)–C(3)	113.5	C(9)–C(10)–C(11)	107.2
N(1)–C(2)–C(7)	110.5	C(10)–C(11)–C(12)	110.6
C(3)–C(2)–C(7)	112.6	C(8)–C(12)–C(11)	105.2
C(2)–C(3)–C(4)	114.1	C(8)–C(12)–C(13)	114.6
C(2)–C(3)–C(17)	111.2	C(8)–C(12)–C(26)	106.5
C(4)–C(3)–C(17)	112.9	C(11)–C(12)–C(13)	107.3
C(3)–C(4)–C(5)	103.7	C(11)–C(12)–C(26)	108.3
C(4)–C(5)–C(6)	108.1	C(13)–C(12)–C(26)	114.5
C(5)–C(6)–C(7)	108.1	C(12)–C(13)–C(14)	119.4
C(5)–C(6)–C(15)	109.1	C(13)–C(14)–C(15)	107.6
C(5)–C(6)–C(20)	103.4	C(6)–C(15)–C(14)	115.3
C(7)–C(6)–C(15)	111.2	C(6)–C(15)–C(16)	113.8
C(7)–C(6)–C(20)	115.6	C(14)–C(15)–C(16)	110.6
C(15)–C(6)–C(20)	109.0	N(1)–C(16)–C(15)	114.9
C(2)–C(7)–C(6)	105.8	C(3)–C(17)–C(18)	109.4
C(2)–C(7)–C(8)	106.4	C(18)–C(17)–C(19)	109.9
C(2)–C(7)–C(21)	109.9	C(3)–C(17)–C(19)	110.9
C(6)–C(7)–C(8)	117.1	C(7)–C(21)–C(22)	114.2
C(6)–C(7)–C(21)	113.3	C(21)–C(22)–C(23)	110.6
C(8)–C(7)–C(21)	104.1	C(22)–C(23)–O(24)	120.5
C(7)–C(8)–C(9)	115.7	O(24)–C(23)–O(25)	122.7
C(7)–C(8)–C(12)	116.1	C(22)–C(23)–O(25)	122.7
C(7)–C(8)–O(24)	110.8	C(8)–O(24)–C(23)	118.1
C(9)–C(8)–C(12)	103.1	N(1)–C(26)–C(12)	117.2

the bond distances for various types of bond are calculated as 1.547 for C–C single bonds, 1.464 for C–N single bonds, 1.358 Å for an aromatic C–O single bond, and 1.216 Å for a C=O double bond.

The structure is heptacyclic and four rings (δ -lactone,

⁷ H. Sakurai, N. Sakabe, and Y. Hirata, *Tetrahedron Letters*, 1966, 6309.

⁸ M. Toda, S. Yamamura, and Y. Hirata, *Tetrahedron Letters*, 1969, 2585.

cyclopentane, piperidine, and cycloheptane) have the C(8)-atom in common giving a complex spiro-system. The piperidine and cycloheptane ring have distorted chair conformations and the cyclopentane ring adopts a deformed α -envelope. The displacements of the atoms from various planes are shown in Table 8. Intramolecular non-bonded interactions form at least one reasonable explanation for bond lengthening in daphnilactone A. For example, there are such interactions between the methyl group C(20) and atoms C(8), C(21), C(22), C(23), and O(24) which could be used to explain the length (1.576 Å) of the C(6)–C(7) bond. Other intramolecular non-bonded contacts are listed in Table 6.

TABLE 6
Some intramolecular non-bonded contacts (Å)

C(17) ... N(1)	2.978	C(20) ... C(14)	2.932
C(18) ... C(2)	3.258	C(20) ... C(21)	3.181
C(19) ... C(2)	3.017	C(20) ... C(22)	3.288
C(17) ... C(4)	3.198	C(9) ... C(23)	2.996
C(18) ... C(4)	2.930	C(20) ... C(23)	3.022
C(20) ... C(8)	3.282	C(20) ... O(24)	2.973

The molecular packing arrangement along the *c*-axis is illustrated in Figure 3. All intermolecular contacts were calculated, and the most significant are given in

TABLE 7
Some intermolecular distances < 3.8 Å

Carbon-carbon		Carbon-oxygen	
C(14) ... C(19 ^I)	3.76	C(5) ... O(25 ^{II})	3.68
C(16) ... C(21 ^{III})	3.59	C(21) ... O(25 ^{II})	3.30
		C(22) ... O(25 ^{II})	3.48
		O(25) ... C(11 ^{II})	3.45
		O(25) ... C(16 ^I)	3.72

Roman numerals as superscripts refer to the following equivalent positions:

$$\begin{array}{ll} \text{I} & \frac{1}{2} + x, \frac{1}{2} - y, 1 - z \\ \text{II} & \frac{1}{2} - x, 1 - y, -\frac{1}{2} + z \\ \text{III} & 1 - x, -\frac{1}{2} + y, \frac{1}{2} - z \end{array}$$

TABLE 8

Equations of best least-squares planes in the form $lx + mY + nZ = p$ where *X*, *Y*, and *Z* are orthogonal co-ordinates in Å. Distances (Å) of relevant atoms from the planes are given in square brackets

	<i>l</i>	<i>m</i>	<i>n</i>	<i>p</i>
Plane (1):				
C(9)—(11)	0.9999	0.0053	−0.0128	6.5777
[C(9) −0.007, C(10) 0.011, C(11) −0.010, C(12) −0.557]				
Plane 2:				
C(22), C(23), O(24), O(25)	0.2718	0.8317	0.4842	8.7512
[C(22) −0.001, C(23) 0.002, O(24) −0.001, O(25) −0.001, C(7) 0.768, C(8) 0.187, C(21) −0.265]				
Plane (3):				
C(2), C(3), C(5), C(6)	−0.4505	0.8698	−0.2013	0.3295
[C(2) 0.019, C(3) −0.017, C(5) 0.018, C(6) −0.018, C(4) −0.453, C(7) 0.793]				
Plane (4):				
N(1), C(7), C(8), C(26)	0.5330	0.8378	−0.1182	7.4618
[N(1) −0.001, C(7) 0.001, C(8) 0.001, C(26) −0.001, C(2) 0.722, C(12) −0.618]				

Table 7. The shortest C ... O contact (3.30 Å) is between C(21) and O(25) in a neighbouring molecule.

From a biogenetic point of view, we suggest that (I) shows the absolute configuration of daphnilactone A by comparison with daphniphyllum alkaloids² (II)—(V).

All computations were performed on a FACOM 230 = 60 at Nagoya University Computation Centre: data reduction, DRPS by K. Sasaki; Σ -2 list, tangent formula, SGMA by K. Sasaki; structure factors, diagonal least-squares, and Fourier with isotropic temperature factors, DLSF by C. Katayama and K. Sasaki; structure factors, block-diagonal least-squares, and Fourier synthesis with anisotropic temperature factors, ATLS by C. Katayama and K. Sasaki.

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