Acta Crystallographica Section E
Structure Reports Online
ISSN 1600-5368

7-Ethenyl-1-[(*Z*)-hydroxymethylidene]-4b,7,10a-trimethylperhydrophenanthren-2-one

Hoong-Kun Fun,[a]* Charoen Pakhathirathien,[b] Suchada Chantrapromma,[b]‡ Chatchanok Karalai[b] and Kan Chantrapromma[c]

[a]X-ray Crystallography Unit, School of Physics, Universiti Sains Malaysia, 11800 USM, Penang, Malaysia, [b]Department of Chemistry, Faculty of Science, Prince of Songkla University, Hat-Yai, Songkhla 90112, Thailand, and [c]Research Unit of Natural Product Utilization, Walailak University, Thasala, Nakhon Si Thammarat 80160, Thailand

‡ Additional correspondence author, email: suchada.c@psu.ac.th

Correspondence e-mail: hkfun@usm.my

Key indicators

Single-crystal X-ray study
T = 100 K
Mean σ(C–C) = 0.002 Å
R factor = 0.041
wR factor = 0.106
Data-to-parameter ratio = 14.3

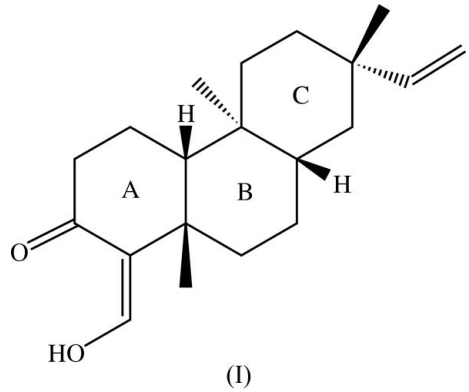
For details of how these key indicators were automatically derived from the article, see http://journals.iucr.org/e.

Received 1 November 2006
Accepted 2 November 2006

The title compound, $C_{20}H_{30}O_2$, also known as tagalsin F, is a diterpenoid which was isolated from *Ceriops tagal* (Perr.). One of the three fused cyclohexane rings adopts a half-chair conformation and the other two cyclohexane rings are in standard chair conformations. The hydroxymethylidene substituent is attached to the half-chair cyclohexane. The molecular structure is stabilized by intramolecular O—H···O hydrogen bonds and weak intermolecular C—H···O interactions. In the crystal structure, molecules are arranged alternately in a head-to-tail and tail-to-head fashion along the *b* axis.

Comment

Ceriops tagal (Perr.) C. B. Robinson is a mangrove plant belonging to the Rhizophoraceae family. The bark of this plant has been used for the treatment of infected wounds in Thailand and for obstetric and hemorrhagic conditions in the Philippines, and its decoction has been used as a substitute for quinine in the treatment of malaria (Bamroongrugsa, 1999). As part of our investigation of mangrove sources of bioactive compounds (Cheenpracha *et al.*, 2005; Pakhathirathien *et al.*, 2005; Chantrapromma *et al.*, 2006; Fun *et al.*, 2006), we have examined *C. Tagal* (Perr.), which was collected from Nakhon Si Thammarat province in the southern part of Thailand. The title compound, (I) was isolated from the roots of this plant and its X-ray crystal structure was undertaken in order to establish the relative stereochemistry. Compound (I) was not found to possess antimalarial activity compared with the artemisinin standard, which has an IC_{50} value of 3.3–3.9 n*M*.



(I)

The molecule of (I) contains a fused three-ring system, *A*/*B*/*C*. The *A*/*B* ring junction is *cis*-fused and the *B*/*C* ring junction is *trans*-fused. The cyclohexane ring *A* adopts a half-chair conformation, with puckering parameters (Cremer & Pople 1975) $Q = 0.464\ (2)$ Å, $\theta = 128.3\ (2)^\circ$ and $\varphi = 78.6\ (3)^\circ$. Rings *B*

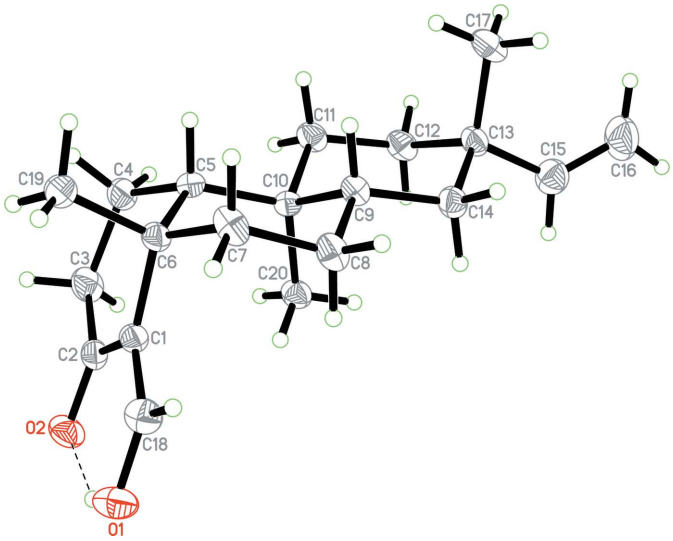


Figure 1
The molecular structure of (I), showing 60% probability displacement ellipsoids and the atomic numbering. The intramolecular hydrogen bond is shown as a dashed line.

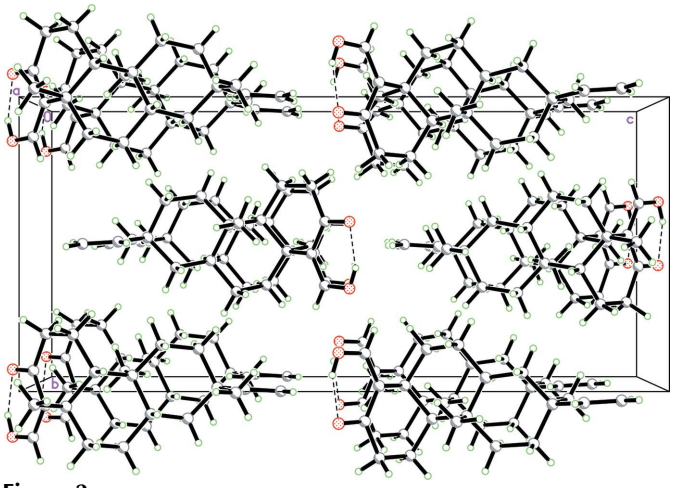


Figure 2
The crystal packing of (I), viewed down the *a* axis. Hydrogen bonds are shown as dashed lines.

and *C* have standard chair conformations. The hydroxymethylidine substituent is attached to cyclopentane ring *A* at atom C1 to form a planar unit that extends across atoms O1, O2, C1, C2, C3, C4 and C18, as indicated by the C2—C1—C18—O1 torsion angle of −0.4 (2)° and by the r.m.s. deviation of the atoms from the O1/O2/C1/C2/C3/C4/C18 plane of 0.023 (2) Å. The intramolecular O1—H1*A*···O2 hydrogen bond further contributes to the planarity of this section of the molecule. The bond angles around C1 and C18 are indicative of sp^2 hybridization for these atoms (Table 1). The C14—C13—C15—C16 torsion angle of −127.2 (2)° describes the orientation of the ethenyl group at C13. The bond distances and angles in (I) show normal values (Allen *et al.*, 1987).

The crystal packing of (I) is stabilized by a weak intermolecular C—H···O interaction (Fig. 2 and Table 2). The molecules are arranged in an alternating head-to-tail and tail-to-head fashion along the *b* axis (Fig. 2).

Experimental

The air-dried and crushed roots of *C. tagal* (3.6 kg) were extracted with dichloromethane and then concentrated *in vacuo* to give a residue (16.3 g). This residue was subjected to rapid column chromatography over silica gel using solvents of increasing polarity from hexane through to 50% acetone–hexane. The eluates were collected and combined, based on thin-layer chromatography, to give 14 fractions (F1–F14). Fraction F2 was further purified by repeated rapid column chromatography with 5% acetone in hexane, yielding compound (I) (41.2 mg). Colourless plates of (I) were obtained by recrystallization from hexane–dichloromethane (1:4 *v*/*v*) after several days (m.p. 374–375 K).

Crystal data

$C_{20}H_{30}O_2$
M_r = 302.44
Orthorhombic, $P2_12_12_1$
a = 6.6826 (2) Å
b = 10.8100 (3) Å
c = 23.8901 (6) Å
V = 1725.79 (8) Å^3
Z = 4
D_x = 1.164 Mg m^{-3}
Mo *Kα* radiation
μ = 0.07 mm^{-1}
T = 100.0 (1) K
Plate, colourless
0.45 × 0.22 × 0.05 mm

Data collection

Bruker SMART APEX2 CCD area-detector diffractometer
ω scans
Absorption correction: multi-scan (*SADABS*; Bruker, 2005)
T_{min} = 0.968, T_{max} = 0.996
26531 measured reflections
2881 independent reflections
2631 reflections with $I > 2\sigma(I)$
R_{int} = 0.047
θ_{max} = 30.0°

Refinement

Refinement on F^2
$R[F^2 > 2\sigma(F^2)]$ = 0.041
$wR(F^2)$ = 0.106
S = 1.05
2881 reflections
202 parameters
H-atom parameters constrained
$w = 1/[\sigma^2(F_o^2) + (0.0546P)^2 + 0.4474P]$ where $P = (F_o^2 + 2F_c^2)/3$
$(\Delta/\sigma)_{max} < 0.001$
$\Delta\rho_{max}$ = 0.35 e Å^{-3}
$\Delta\rho_{min}$ = −0.18 e Å^{-3}

Table 1
Selected geometric parameters (Å, °).

O1—C18	1.311 (2)	C1—C18	1.382 (2)
O2—C2	1.2790 (19)	C15—C16	1.311 (3)
C18—C1—C2	117.22 (15)	O1—C18—C1	124.00 (16)
C18—C1—C6	123.22 (15)	O1—C18—H18*A*	118.0
C2—C1—C6	119.26 (14)	C1—C18—H18*A*	118.0
C12—C13—C15—C16	115.0 (2)	C2—C1—C18—O1	−0.4 (2)
C14—C13—C15—C16	−127.2 (2)	C6—C1—C18—O1	173.20 (15)

Table 2
Hydrogen-bond geometry (Å, °).

D—H···*A*	*D*—H	H···*A*	*D*···*A*	*D*—H···*A*
O1—H1*A*···O2	0.82	1.73	2.4571 (19)	147
C3—H3*A*···O2^{i}	0.97	2.56	3.331 (2)	137

Symmetry code: (i) $x-\frac{1}{2}, -y+\frac{1}{2}, -z+1$.

All H atoms were placed in calculated positions, with an O—H distance of 0.82 Å and C—H distances in the range 0.93–0.97 Å. The U_{iso}(H) values were constrained to be $1.5U_{eq}$(C,O) for hydroxyl and methyl H atoms and $1.2U_{eq}$(C) for the remaining H atoms. A rotating-group model was used for the methyl groups. In the absence of significant anomalous scattering effects, Friedel pairs were merged; the absolute configuration has been assigned arbitrarily.

Data collection: *APEX2* (Bruker, 2005); cell refinement: *APEX2*; data reduction: *SAINT* (Bruker, 2005); program(s) used to solve structure: *SHELXTL* (Sheldrick, 1998); program(s) used to refine structure: *SHELXTL*; molecular graphics: *SHELXTL*; software used to prepare material for publication: *SHELXTL* and *PLATON* (Spek, 2003).

The authors thank Prince of Songkla University, the Malaysian Government, and Universiti Sains Malaysia for Scientific Advancement Grant Allocation (SAGA) grant No. 304/PFIZIK/653003/A118. The assistance of the Bioassay Research Facility of BIOTEC, Thailand, in providing bioactivity assays is also appreciated.

References

Allen, F. H., Kennard, O., Watson, D. G., Brammer, L., Orpen, A. G. & Taylor, R. (1987). *J. Chem. Soc. Perkin Trans. 2*, pp. S1–19.
Bamroongrugsa, N. (1999). *Songklanakarin J. Sci. Technol.* **21**, 377–386.
Bruker (2005). *APEX2* (Version 1.27), *SAINT* and *SADABS*. Bruker AXS Inc., Madison, Wisconsin, USA.
Chantrapromma, S., Pakathirathien, C., Fun, H.-K., Razak, I. A. & Karalai, C. (2006). *Acta Cryst.* E**62**, o1742–o1744.
Cheenpracha, S., Srisuwan, R., Karalai, C., Ponglimanont, C., Chantrapromma, S., Chantrapromma, K., Fun, H.-K., Anjum, S. & Rahman, A. U. (2005). *Tetrahedron*, **61**, 8656–86.
Cremer, D. & Pople, J. A. (1975). *J. Am. Chem. Soc.* **97**, 1354–1358.
Flack, H. D. (1983). *Acta Cryst.* A**39**, 876–881.
Fun, H.-K., Chantrapromma, S., Boonnak, N., Chaiyadej, K., Chantrapromma, K. & Yu, X.-L. (2006). *Acta Cryst.* E**62**, o3725–o3727.
Pakhathirathien, C., Karalai, C., Ponglimanont, C., Subhadhirasakul, S. & Chantrapromma, K. (2005). *J. Nat. Prod.* **68**, 1787–1789.
Sheldrick, G. M. (1998). *SHELXTL*. Version 5.10. Bruker AXS Inc., Madison, Wisconsin, USA.
Spek, A. L. (2003). *J. Appl. Cryst.* **36**, 7–13.