Fig. 1. Perspective view of the molecule with atom labelling.

respectively *versus* the benzo ring (plane 1). The regioisomer (3) is clearly revealed. The cyclohexenic ring has a half-boat conformation [C(4) 1·31 Å above the benzo ring], the C(5) atom being slightly displaced (0·16 Å) in the same way as in a similar previously studied compound (Mercier, Kitane, Tshiamala, Vebrel, Laude & Sheldrick, 1986).

The C—C bond common to the five- and six-

membered rings (1·566 Å) is comparable to analogous bonds observed in fused pentagonal rings (Mercier, Ramah, Laude, Faure & Loiseleur, 1984). The C=N double bond is not conjugated with the α-phenyl group (plane 2) (angle 22·3° with plane 2) but is exactly coplanar with the β-phenyl group (plane 3) as is the pyrazolinic plane.

The regioisomer (3) is clearly revealed by the typical bond lengths of the C=N—N group [C=N = 1·293, N(2)—N(3) = 1·367 Å] as well as the C(3a)—N(3) = 1·473 Å and C(9b)—C(1) = 1·516 Å bonds of the pyrazolinic ring.

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Quinol–1,8-Epoxymenthane (1/2)*

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Abstract. $C_6H_6O_2.2C_{10}H_{18}O$, $M_r = 418·62$, monoclinic, $P2_1/n$, $a = 7·6793$ (3), $b = 15·1204$ (8), $c = 10·8318$ (13) Å, $\beta = 102·260$ (7)°, $V = 1229·04$ Å³, $Z = 2$, $D_x = 1·13$ Mg m⁻³, $\lambda(Cu\,K\alpha) = 1·5418$ Å, $\mu = 0·51$ cm⁻¹, $F(000) = 460$, $T = 293$ K, $R = 0·046$ for 1870 reflexions. The complex occurs as isolated trimolecular units with O⋯H—O bonding [2·724 (2) Å]. The torsion angle about the O⋯H link [32·3 (5)°] implies partial involvement of both lone pairs of electrons of the ether O atom. Other bond lengths and angles have typical values.

Introduction. 1,8-Epoxymenthane (1,8-cineole), CIN, is a cyclic ether where, in spite of the bulk of the molecule, the lone pairs of electrons on the oxygen atom are well

exposed to the approach of other species. Thus CIN can act as a monodentate ligand in complexes such as $SnCl_4.2CIN$ (Sumarova & Nevskaya, 1956).

CIN
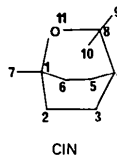
Attempts in our laboratory to investigate the crystal structures of the complexes of CIN with iodine, silver perchlorate, tin(II) bromide and ferric chloride have been abandoned because of inadequate crystal quality and low thermal stability. An alternative role for CIN is in the formation of hydrogen-bonded complexes similar to those formed by the bridging cyclic diether 1,4-

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* Quinol is 1,4-benzenediol.

dioxan (dx). Among many others, structures have been reported for dioxan–sulfuric acid (Hassel & Hvoslev, 1954) and for $NiBr_2.4H_2O.2dx$ (Barnes & Weakley, 1976) and $SmBr_3.9H_2O.2dx$ (Barnes & Nicoll, 1985) where the metal complexes $NiBr_2.4H_2O$ and $[Sm(H_2O)_9]^{3+}$ are interlinked by dioxan molecules hydrogen-bonded to coordinated water. The analogy between the dioxan and cineole complexes has been demonstrated in a series of $[Sn_2X_6(OH)_2(OH_2)_2].n(ether)$ (X = Cl, Br) species where the coordinated hydroxyl and water moieties are both hydrogen-bonded to the ether molecules (Barnes, Sampson & Weakley, 1980). The best characterized hydrogen-bonded complexes of CIN not involving metal ions are those of the phenols (Baeyer & Villiger, 1902). Although these are highly crystalline no structures have been reported until the present determination of the title compound.

Experimental. The complex was prepared by recrystallizing quinol from CIN. The equidimensional multifaceted crystals ($0.30 \times 0.30 \times 0.30$ mm) lose CIN rapidly in air and so were stored in the mother liquor before mounting in Lindemann-glass capillaries (m.p. 379.7 K with extensive decomposition). Cell dimensions were measured from 25 accurately centred reflexions with $\theta \simeq 34°$ before data collection on an Enraf–Nonus CAD-4 diffractometer (SERC service at Queen Mary College, London). 2503 reflexions were collected in the range $2 \le 70°$: $0 \le h \le 9$, $0 \le k \le 18$, $-12 \le l \le 12$. These reduced to 2333 unique reflexions with $R_{int} = 0.034$, of which 1870 with $F_o > 3\sigma(F_o)$ were used in the refinement.

All calculations were performed on the Dundee University DEC10 computer using SHELX76 (Sheldrick, 1976), XANADU (Roberts & Sheldrick, 1975) and PLUTO (Motherwell & Clegg, 1978) program packages. Atomic scattering factors as in SHELX76.

The molecule lies about a centre of symmetry, which must be coincident with the centre of the quinol ring. The direct-methods routine EEES revealed all of the non-hydrogen atoms. Conventional least-squares refinements gave R 0.115 with all of the non-hydrogen atoms anisotropic. A difference synthesis showed all H atoms,
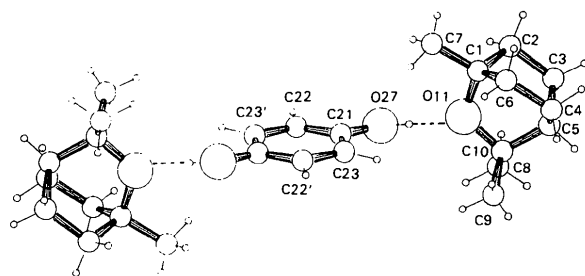

Fig. 1. The quinol–1,8-cineole (1/2) complex with atomic numbering.

Table 1. *Coordinates* $(\times 10^4)$ *for non-hydrogen atoms with e.s.d.'s in parentheses*

$$U_{eq} = \tfrac{1}{3}\Sigma_i \Sigma_j U_{ij} a_i^* a_j^* \mathbf{a}_i.\mathbf{a}_j.$$

	x	y	z	$U_{eq}(\text{Å}^2 \times 10^3)$
C1	3266 (2)	2601 (1)	2935 (1)	50 (1)
C2	5078 (2)	3037 (1)	3389 (2)	66 (1)
C3	4822 (3)	3994 (1)	3762 (2)	68 (1)
C4	2822 (2)	4194 (1)	3535 (2)	57 (1)
C5	2071 (3)	4077 (1)	2124 (2)	67 (1)
C6	2218 (3)	3102 (1)	1799 (2)	62 (1)
C7	3387 (3)	1620 (1)	2677 (2)	80 (1)
C8	1915 (2)	3558 (1)	4302 (1)	45 (1)
C9	−106 (3)	3656 (1)	4025 (2)	70 (1)
C10	2636 (3)	3643 (1)	5719 (2)	65 (1)
C11	2307 (2)	2659 (1)	3951 (1)	49 (1)
C21	1224 (2)	684 (1)	5311 (1)	47 (1)
C22	1489 (2)	−115 (1)	5946 (1)	50 (1)
C23	−269 (2)	796 (1)	4363 (1)	49 (1)
O27	2466 (2)	1339 (1)	5682 (1)	72 (1)

Table 2. *Interatomic distances* (Å) *and angles* (°)

C2–C1	1.523 (2)	C6–C5	1.525 (3)
C6–C1	1.522 (2)	C9–C8	1.525 (2)
C7–C1	1.516 (3)	C10–C8	1.523 (2)
O11–C1	1.451 (2)	O11–C8	1.460 (2)
C3–C2	1.527 (3)	C22–C21	1.383 (2)
C4–C3	1.533 (3)	C23–C21	1.378 (2)
C5–C4	1.524 (2)	O27–C21	1.374 (2)
C8–C4	1.532 (2)	C22–C23'	1.384 (2)
C6–C1–C2	109.6 (1)	C9–C8–C4	113.2 (1)
C7–C1–C2	113.3 (2)	C10–C8–C4	112.6 (1)
C7–C1–C6	112.4 (1)	C10–C8–C9	109.1 (2)
O11–C1–C2	107.8 (1)	O11–C8–C4	107.5 (1)
O11–C1–C6	108.5 (1)	O11–C8–C9	107.2 (1)
O11–C1–C7	105.0 (1)	O11–C8–C10	106.8 (1)
C3–C2–C1	109.6 (1)	C8–O11–C1	114.8 (1)
C4–C3–C2	109.0 (1)	C23–C21–C22	119.3 (1)
C5–C4–C3	106.9 (2)	O27–C21–C22	117.6 (1)
C8–C4–C3	110.1 (1)	O27–C21–C23	123.1 (1)
C8–C4–C5	110.6 (1)	C21–C22–C23'	120.5 (1)
C6–C5–C4	108.3 (1)	C21–C23–C22'	120.1 (1)
C5–C6–C1	110.3 (1)		

which were included in the refinement with individual isotropic thermal parameters. Final refinement (minimizing $\Sigma w|F_o - |F_c||^2$), 223 refined parameters in two blocks, $R = 0.046$, $wR = 0.077$, $w = 1.000/[\sigma^2(F) + 0.002711F^2]$, mean $\Delta/\sigma = 0.071$, max. $\Delta/\sigma = 0.551$, max. and min. $\Delta\rho = +0.20, -0.14$ e Å^{-3}.

Discussion. Atomic coordinates are given in Table 1, with bond lengths and angles in Table 2.* Fig. 1 shows that the complex is a molecular species, being neither a quinol clathrate (Chan & Mak, 1983) nor a polymer, as in quinol–acetone (Lee & Wallwork, 1959) or quinol–urea (Mahmoud & Wallwork, 1975). The angle at the

oxygen atom in CIN, C1—O11—C8, is 114·8 (1)°, very close to the mean of the values found in the Sn complexes (Barnes, Sampson & Weakley, 1980). A librational calculation on the CIN molecule alone confirms its rigidity: $RG = 0.046$ compared with 0·28 for the whole complex.

All three methyl groups are well defined, with no suggestion of disorder. The quinol molecule is identical with those found in quinol itself (Wallwork & Powell, 1980) and in quinol complexes. The angle C23—C21—O27 has the typical value of 123·1 (3)° to minimize the C23···H271 interaction. O27 is 0·03 (2) Å above the ring plane while H271 is 0·24 (2) Å below the plane, with C21—O27—H271 = 109·9 (12)°.

The hydrogen-bond length O11···O27 is 2·724 (2) Å, with O11···H271 1·700 (26) Å and O27—H271···O11 173·9 (10)°. In the Sn complexes the average hydrogen-bond distances are 2·71 Å to hydroxyl and 2·59 Å to water.

C4, C8, O11 and C1 are constrained to be approximately coplanar by the ring system. Thus the limiting possibilities are that the hydrogen bond to H(271) uses only one lone pair of electrons from O11, in which case the torsion angle C10—C8—O11—H271 would be close to 0 (or 120)° or that the hydrogen bond uses the combined lone pairs contributing equally, with the torsion angle about 60°. The experimental value is 32·3 (12)°, implying that both lone pairs contribute, but unequally.

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Structure of 4-(N-Benzyl-N-methylamino)-1,7,8,9-tetrachloro-10,10-dimethoxy-3-oxa-4-azatricyclo[5.2.1.0²,⁶]deca-4,8-diene

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Abstract. $C_{18}H_{18}Cl_4N_2O_3$, $M_r = 452.16$, monoclinic, $P2_1/n$, $a = 9.692$ (5), $b = 24.204$ (4), $c = 8.664$ (5) Å, $\beta = 94.45$ (5)°, $V = 2026$ (2) Å^3, $Z = 4$, $D_x = 1.48$ g cm^{-3}, Cu $K\alpha$, $\lambda = 1.5418$ Å, $\mu = 56.168$ cm^{-1}, $F(000) = 928$, room temperature, $R = 0.070$ for 3192 symmetry-independent observed reflections [$I > 3\sigma(I)$]. The five-membered ring fused to the norbornene moiety is in an *endo* configuration. The dihedral angle between the least-squares planes through the benzene and five-membered heterocyclic rings is 80·7 (2)°.

Introduction. In order to establish the stereochemistry of the product in the (4+2)-cycloaddition reaction of 2-(N-benzyl-N-methylamino)oxazole with 1,1-dimethoxytetrachlorocyclopentadiene (Dondoni, Fogagnolo, Mastellari, Pedrini & Ugozzoli, 1986) we have determined the crystal and molecular structure of the title compound.

Experimental. Colourless prismatic crystal, 0·29 × 0·30 × 0·60 mm approximately, Siemens AED single-