squares techniques, for 116 parameters including non-H atoms anisotropic and H atoms isotropic with fixed temperature factors. Final $R = 0.033$, $wR = 0.040$ with the weighting scheme $w = 1/\sigma^2(F_o)$, $S = 1.44$. Maximum final shift to e.s.d. ratio was 0.06, maximum and minimum peak heights in the final difference Fourier map were 0.26 and -0.15 e Å^{-3}. Neutral atomic scattering factors and f' and f'' values for non-H atoms were taken from *International Tables for X-ray Crystallography* (1974, Vol. IV). Computer programs: *TEXSAN* (Molecular Structure Corporation, 1985) and *PLUTO* (Motherwell & Clegg, 1978). The molecule has a centre of symmetry. The atomic positions of non-H atoms are listed in Table 1.* Bond lengths, bond angles and selected torsion angles for the non-H atoms are given in Table 2. The molecular structure with the numbering scheme is shown in Fig. 1.

Related literature. Intramolecular symmetry is confirmed by previously reported IR and Raman spectroscopy studies (McQuillan & Oxton, 1978). The distortion of the tetrahedral configuration around the P atom, and the P—P$^{\rm i}$, P—S, P—C bond lengths and bond angles are in good agreement with the structure of tetraethyldiphosphine disulfide (Dutta & Woolfson, 1961), bis(cyclotetramethylene)diphosphine disulfide (Lee & Goodacre, 1969) and bis(cyclopentamethylene)diphosphine disulfide (Lee & Goodacre, 1970).

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Structure of Ethyl 2-(3-Methyl-1,2,4-oxadiazol-5-yl)-2-phenylhydrazonoethanoate

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Abstract. $C_{13}H_{14}N_4O_3$, $M_r = 274.3$, orthorhombic, $Pna2_1$, $a = 19.048$ (6), $b = 10.507$ (4), $c = 6.691$ (4) Å, $V = 1339.1$ Å^3, $Z = 4$, $D_m = 1.36$ (flotation), $D_x = 1.360$ Mg m^{-3}, λ(Mo $K\alpha$) = 0.7107 Å, $\mu = 0.933$ cm^{-1}, $F(000) = 576$, $T = 298$ K, $R = 0.0839$, $wR = 0.0643$ for 773 observed reflections. The whole molecule is approximately planar, which can be seen from the values of the torsion angles about the non-rigid bonds; the molecular dimensions are in agreement with those found in analogous compounds. An important feature of the molecule is an intramolecular N—H···N hydrogen bond, which is characterized by an N···N distance of 2.66 (1) Å.

Experimental. Single crystals were obtained by slow evaporation of a solution in absolute ethanol, under reduced constant pressure in a dry atmosphere, Approximate unit-cell parameters were determined from preliminary Weissenberg and precession photographs. A crystal of ~0.4 × 0.2 × 0.07 mm was used

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Table 1. *Fractional atomic coordinates* ($\times 10^4$) *and equivalent isotropic temperature factors* ($\text{Å}^2 \times 10^3$)

$$U_{eq} = (1/3)\textstyle\sum_i\sum_j U_{ij}a_i^*a_j^*\mathbf{a}_i.\mathbf{a}_j.$$

	x	y	z	U_{eq}
O(1)	3176 (2)	6941 (3)	5167 (25)	76 (2)
O(2)	5205 (2)	5380 (4)	4982 (23)	81 (2)
O(3)	4054 (2)	5112 (3)	5050 (27)	113 (3)
N(1)	4958 (2)	9156 (4)	4997 (24)	51 (2)
N(2)	5010 (2)	7918 (4)	5121 (25)	51 (2)
N(4)	3569 (2)	8913 (4)	5095 (21)	47 (2)
N(3)	2575 (2)	7757 (4)	5011 (33)	80 (3)
C(1)	5552 (3)	9946 (5)	5276 (20)	42 (3)
C(2)	5439 (3)	11242 (5)	5149 (29)	50 (3)
C(3)	6011 (3)	12069 (5)	5223 (25)	53 (3)
C(4)	6680 (3)	11590 (5)	5127 (31)	62 (3)
C(5)	6785 (3)	10297 (5)	5050 (26)	50 (2)
C(6)	6227 (3)	9453 (5)	5136 (30)	51 (2)
C(7)	4451 (3)	7201 (5)	5094 (30)	45 (3)
C(8)	3749 (3)	7703 (5)	5000	47 (2)
C(9)	2855 (3)	8875 (5)	5312 (17)	43 (3)
C(10)	2397 (3)	10065 (6)	5240 (26)	67 (4)
C(11)	4633 (3)	5799 (5)	4962 (26)	58 (3)
C(12)	4163 (5)	3736 (6)	4762 (40)	162 (10)
C(13)	3678 (4)	3108 (6)	5517 (39)	160 (12)

Table 2. *Final interatomic distances* (Å), *bond angles* (°) *and torsion angles* (°)

C(1)—C(2)	1.381 (8)	C(1)—C(2)—C(3)	119.4 (5)
C(2)—C(3)	1.394 (8)	C(2)—C(3)—C(4)	119.7 (5)
C(3)—C(4)	1.372 (8)	C(3)—C(4)—C(5)	120.0 (5)
C(4)—C(5)	1.374 (7)	C(4)—C(5)—C(6)	121.3 (5)
C(5)—C(6)	1.385 (8)	C(5)—C(6)—C(1)	118.3 (5)
C(6)—C(1)	1.389 (8)	C(6)—C(1)—C(2)	120.5 (5)
N(1)—C(1)	1.416 (7)	C(2)—C(1)—N(1)	116.4 (5)
N(1)—N(2)	1.307 (6)	C(6)—C(1)—N(1)	120.8 (5)
N(2)—C(7)	1.304 (7)	C(1)—N(1)—N(2)	121.0 (4)
C(7)—C(8)	1.439 (8)	N(1)—N(2)—C(7)	120.8 (4)
C(8)—O(1)	1.358 (7)	N(2)—C(7)—C(8)	123.2 (5)
O(1)—N(3)	1.434 (6)	C(7)—C(8)—O(1)	121.8 (3)
N(3)—C(9)	1.306 (8)	C(7)—C(8)—N(4)	126.4 (3)
C(9)—C(10)	1.525 (8)	C(8)—O(1)—N(3)	106.4 (3)
C(9)—N(4)	1.368 (7)	O(1)—N(3)—C(9)	101.6 (4)
N(4)—C(8)	1.318 (7)	N(3)—C(9)—C(10)	119.9 (5)
C(7)—C(11)	1.516 (8)	N(3)—C(9)—N(4)	114.6 (5)
C(11)—O(2)	1.175 (7)	N(4)—C(9)—C(10)	122.8 (5)
C(11)—O(3)	1.319 (7)	C(9)—N(4)—C(8)	103.6 (4)
O(3)—C(12)	1.473 (8)	N(4)—C(8)—O(1)	110.8 (2)
C(12)—C(13)	1.243 (18)	C(8)—C(7)—C(11)	124.5 (4)
		N(2)—C(7)—C(11)	112.0 (5)
		C(7)—C(11)—O(2)	125.1 (5)
		C(7)—C(11)—O(3)	109.7 (5)
		C(11)—O(3)—C(12)	114.4 (5)
		O(3)—C(12)—C(13)	111.3 (6)
		O(3)—C(11)—O(2)	124.7 (5)

N(3)—O(1)—C(8)—N(4)	10.5 (1.0)	O(1)—N(3)—C(9)—N(4)	17.1 (1.2)
N(3)—O(1)—C(8)—C(7)	179.9 (0.9)	O(1)—N(3)—C(9)—C(10)	178.9 (0.9)
C(8)—O(1)—N(3)—C(9)	− 16.1 (1.1)	N(1)—C(1)—C(6)—C(5)	168.2 (1.0)
C(12)—O(3)—C(11)—O(2)	− 12.0 (1.8)	N(1)—C(1)—C(2)—C(3)	− 174.1 (1.0)
C(11)—O(3)—C(12)—C(13)	156.8 (1.4)	C(2)—C(1)—C(6)—C(5)	5.8 (1.6)
C(12)—O(3)—C(11)—C(7)	175.0 (1.1)	C(6)—C(1)—C(2)—C(3)	− 10.9 (1.6)
N(2)—N(1)—C(1)—C(2)	178.9 (1.0)	C(1)—C(2)—C(3)—C(4)	9.5 (1.7)
N(2)—N(1)—C(1)—C(6)	18.0 (1.5)	C(2)—C(3)—C(4)—C(5)	− 2.9 (1.8)
C(1)—N(1)—N(2)—C(7)	172.0 (1.0)	C(3)—C(4)—C(5)—C(6)	− 2.2 (1.8)
N(1)—N(2)—C(7)—C(8)	− 2.2 (1.8)	C(4)—C(5)—C(6)—C(1)	0.9 (1.7)
N(1)—N(2)—C(7)—C(11)	172.0 (1.0)	N(2)—C(7)—C(8)—N(4)	− 6.0 (1.6)
C(9)—N(4)—C(8)—O(1)	− 0.5 (0.9)	N(2)—C(7)—C(8)—O(1)	− 173.6 (1.0)
C(8)—N(4)—C(9)—C(10)	− 172.5 (0.8)	N(2)—C(7)—C(11)—O(3)	176.6 (1.0)
C(8)—N(4)—C(9)—N(3)	− 11.3 (1.1)	N(2)—C(7)—C(11)—O(2)	3.6 (1.7)
C(9)—N(4)—C(8)—C(7)	− 169.2 (0.8)	C(8)—C(7)—C(11)—O(2)	177.8 (1.1)

for data collection on a Philips PW 1100 four-circle diffractometer with graphite monochromator. Accurate unit-cell parameters and crystal-orientation matrices (with e.s.d.'s) were determined from least-squares refinement of the 2θ, ω, χ and φ values of 20 carefully centred reflections ($9 < \theta < 15°$). Data were collected using θ–2θ scans of speed 0.03° s^{-1} and width 1.20°, 2θ range 4–50° and for $0 \le h \le 22$, $0 \le k \le 12$, $0 \le l \le 7$. Two standard reflections (311, 220) were monitored every 180 min; no significant variation occurred. 1258 data were measured, of which 773 had $I \ge 3\sigma(I)$. Lorentz and polarization corrections were applied. Intensities were placed on an absolute scale by Wilson's method, and a trial structure obtained by direct methods using *MULTAN*80 (Main, Fiske, Hull, Lessinger, Germain, Declercq & Woolfson, 1980). The structure was refined by blocked-matrix least squares, minimizing $\sum w(|F_o| - |F_c|)^2$, with $w = 2.667[\sigma^2(F_o) + 0.00011F_o^2]^{-1}$; the adequacy of the weighting scheme was confirmed by analysis of the variation of the mean $w(|F_o| - |F_c|)^2$ with $|F_o|$ and $\sin\theta/\lambda$. Scattering factors were taken from *International Tables for X-ray Crystallography* (1974, Vol. IV, pp. 71–103). The refinement was carried out allowing all non-H atoms to vibrate anisotropically; all the H atoms were located from a difference Fourier map and isotropically refined in the last least-squares cycle. Final conventional $R = 0.0839$ ($wR = 0.0643$) for the 773 observed reflections. Calculations were carried out on the CYBER 76 computer of 'CINECA' with *SHELX*76 (Sheldrick, 1976). $(\Delta/\sigma)_{\max}$ in the final refinement cycle for positional parameters of the non-H atoms was 0.09; maximum and minimum heights in final difference Fourier synthesis were ± 0.3 e Å^{-3}. This structure determination is on the borderline of acceptable precision mainly owing to the poor quality of the crystals. Nevertheless, since the aim of this work was the determination of the molecular conformation, we believe that our results do warrant presentation.

We refined the structure in the space group *Pnam*, by using both full-matrix and blocked-matrix least-squares techniques. The results were worse than those obtained in the space group $Pna2_1$. The R value was significantly higher ($R = 0.1075$) and the thermal parameters of C(6) were non-positive-

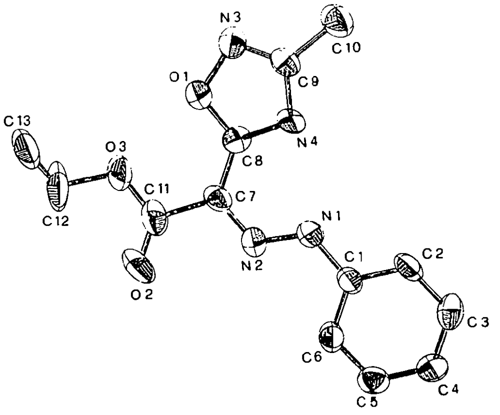

Fig. 1. The molecular structure of the title compound with the atom-numbering scheme.

definite. Therefore we concluded that the structure determination in $Pna2_1$ appears to be more sensible.

The final atomic parameters are given in Table 1 and interatomic distances and interbond angles with e.s.d.'s are listed in Table 2, along with torsion angles.* A perspective view of the molecule with the atom-numbering scheme is presented in Fig. 1.

Related literature. The title compound belongs to a series of substituted hydrazones synthesized and tested from a pharmacological point of view by the late Professor Silvano Rossi (Rossi, 1985). The hydrazone portion of the molecule, *i.e.* from C(1) to C(8) through N(1), N(2) and C(7), is of special interest. The geometry here as it pertains to the hydrazone–azo question has been discussed in detail by Pendergrass, Paul & Curtin (1972); they list literature values of bond lengths expected for the two tautomers: N—N 1.33–1.38, N=N 1.23–1.28; N—C(amide) 1.30–1.41; N=C 1.27–1.29 Å. The important point is that our distances fit into the pattern for the azo tautomer. Our N—C distance is on the low end of the range quoted and our N=N is slightly larger than observed in some related compounds, suggesting perhaps that there might be some small contribution present from the hydrazone tautomer.

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* Lists of structure factors, anisotropic thermal parameters, least-squares-planes data, H-atom coordinates and bond distances involving H atoms have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 55676 (10 pp.). Copies may be obtained through The Technical Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England. [CIF reference: HU1003]

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Structure of a 2′-Oxo-8.1′-neolignan

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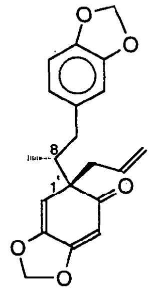
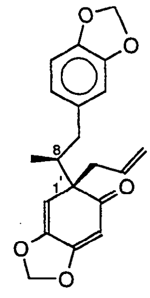
Abstract. Δ^8-1′,2′-Dihydro-3,4,3′,4′-bismethylenedioxy-2′-oxo-8.1-neolignan [6-allyl-6-(1,3-benzodioxol-5-ylpropylene)-1,3-benzodioxol-5(6*H*)-one], $C_{20}H_{20}O_5$, $M_r = 340.38$, orthorhombic, $P2_12_12_1$, $a = 6.628\ (4)$, $b = 12.604\ (6)$, $c = 20.774\ (11)$ Å, $V = 1735\ (3)$ Å^3, $Z = 4$, $D_x = 1.308$ g cm^{-3}, λ(Mo $K\alpha$) = 0.7107 Å, $\mu = 0.876$, $F(000) = 720$, $T = 295$ K, $R = 0.032$, $wR = 0.041$ for 972 unique reflections. The analysis confirmed the structure of this new 2′-oxo-8.1′-neolignan, originally proposed on the basis of NMR data, and established the relative stereochemistry. No hydrogen bonding was observed; the closest intermolecular contact was between O2 and O4 (O atoms in the 1-position on each 1,3-benzodioxole) at 2.99 Å. The molecule has a bent conformation with a dihedral angle of 51.2 (1)° between the two planes defined by the aromatic rings.

Experimental. The title compound (1) and a diastereomer (2) were isolated from separate fractions of an extract of *Piper capense* roots (Green & Wiemer, 1991). A single crystal of the title compound was obtained from methanol by slow evaporation. The crystal, with approximate dimensions 0.6 × 0.5 × 0.4 mm, was mounted on an Enraf–Nonius



(1)



(2)

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