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3-Methyl-9-phenyl-1,3,8,9-tetraazabicyclo[5.3.0]dec-7-en-10-one

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Abstract. $C_{13}H_{16}N_4O$, $M_r = 244.30$, orthorhombic, $P2_12_12_1$, $a = 8.6562\,(3)$, $b = 11.0875\,(4)$, $c = 12.8259\,(10)$ Å, $V = 1231.0\,(1)$ Å^3, $Z = 4$, $D_x = 1.308$ g cm^{-3}, $\lambda(\text{Mo } K\alpha) = 0.71069$ Å, $\mu = 0.82$ cm^{-1}, $F(000) = 520$, $T = 293$ K, $R = 0.036$ for 1452 observed reflections. The molecule consists of a five- and a six-membered ring, which are planar [maximum deviations 0.005 (2) and 0.007 (2) Å, respectively], and a seven-membered ring, which adopts a chair-like conformation. The N(3)═C(8) and O═C(7) double bonds are clearly defined by bond lengths [1.296 (3) and 1.222 (3) Å, respectively].

Experimental. The title compound belongs to the class of 4,5-heteroannulated 2,3-dihydro-2-phenyl-4*H*-1,2,4-triazol-3-ones. The compound has been obtained as the sole product by potassium permanganate oxidation of 7-methyl-2-phenyl-1,2,4,7-tetraazaspiro[4.5]decan-3-one (Seil, 1989; Gstach & Seil, 1990*a*). The crystals were grown from propan-2-ol. Pale yellow crystal, 0.2 × 0.3 × 0.5 mm. Cell dimensions from setting angles of 32 reflections having $10 < 2\theta < 30°$. Data collection on a Phillips PW 1100 four-circle diffractometer, Mo $K\alpha$ radiation, graphite monochromator, ω–2θ scans, scan width 1.2°, scan speed 2° min^{-1}, $4 < 2\theta < 48°$, $0 \leq h \leq 9$, $-12 \leq k \leq 12$, $-14 \leq l \leq 14$, 4135 reflections, 1937 independent reflections ($R_{\text{int}} = 0.021$), 1452 with $I > 3\sigma(I)$ observed. The intensities of three standard reflections (112, 222, 213) measured every 2 h were found to be constant within 2%. Data corrected for Lorentz and polarization effects, but not for absorption.

The structure was solved by direct methods and difference Fourier syntheses and refined by least-squares methods, based upon F with weights $w = [\sigma^2(F_o) + 0.00033F_o^2]^{-1}$ (Sheldrick, 1976). H atoms included using a riding model (C—H = 1.08 Å, H—C—H = 109.5°, phenyl H atoms on the external bisector of the appropriate C—C—C angle). C, N and O atoms anisotropic, H atoms isotropic. Atomic scattering factors were taken from *International Tables for X-ray Crystallography* (1974, Vol. IV). Final $R = 0.036$, $wR = 0.035$, maximum and minimum electron density in final difference Fourier map 0.13 and -0.12 e Å^{-3}, respectively. $(\Delta/\sigma)_{\max} = 0.005$.

Final atomic coordinates and equivalent isotropic thermal parameters are given in Table 1.[*] Bond

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[*] Lists of structure factors, anisotropic thermal parameters and positional and isotropic thermal parameters for the H atoms, have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 54484 (13 pp.). Copies may be obtained through The Technical Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

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Table 1. *Atomic coordinates and isotropic thermal parameters for non-*H *atoms with e.s.d.'s in parentheses.*

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

	x	y	z	U_{eq} (Å^2 × 10^3)
O	0.4655 (2)	0.4382 (2)	1.0170 (2)	66 (1)
N(1)	0.3062 (2)	0.4356 (2)	0.8689 (2)	44 (1)
N(2)	0.4477 (3)	0.2783 (2)	0.8995 (2)	47 (1)
N(3)	0.2745 (3)	0.3518 (2)	0.7896 (1)	49 (1)
N(4)	0.4956 (3)	0.0763 (2)	0.9707 (2)	54 (1)
C(1)	0.2327 (4)	0.5499 (2)	0.8667 (2)	45 (1)
C(2)	0.2486 (4)	0.6302 (2)	0.9479 (2)	57 (2)
C(3)	0.1714 (4)	0.7408 (3)	0.9419 (2)	70 (2)
C(4)	0.0815 (4)	0.7698 (3)	0.8575 (3)	71 (2)
C(5)	0.0689 (4)	0.6897 (3)	0.7768 (3)	64 (2)
C(6)	0.1444 (3)	0.5786 (2)	0.7794 (2)	53 (2)
C(7)	0.4123 (3)	0.3912 (2)	0.9387 (2)	46 (2)
C(8)	0.3623 (3)	0.2604 (2)	0.8110 (2)	47 (2)
C(9)	0.5574 (4)	0.1915 (3)	0.9488 (3)	58 (2)
C(10)	0.4695 (4)	−0.0029 (2)	0.8812 (3)	65 (2)
C(11)	0.3402 (4)	0.0322 (2)	0.8070 (3)	64 (2)
C(12)	0.3689 (4)	0.1489 (3)	0.7467 (2)	61 (2)
C(13)	0.3743 (4)	0.0717 (3)	1.0496 (2)	67 (2)

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

O—C(7)	1.222 (3)	N(4)—C(13)	1.459 (4)
N(1)—N(3)	1.404 (3)	C(1)—C(2)	1.377 (4)
N(1)—C(1)	1.419 (3)	C(1)—C(6)	1.393 (4)
N(1)—C(7)	1.373 (3)	C(2)—C(3)	1.398 (4)
N(2)—C(7)	1.383 (3)	C(3)—C(4)	1.371 (5)
N(2)—C(8)	1.370 (3)	C(4)—C(5)	1.369 (5)
N(2)—C(9)	1.492 (4)	C(5)—C(6)	1.396 (4)
N(3)—C(8)	1.296 (3)	C(8)—C(12)	1.487 (4)
N(4)—C(9)	1.414 (3)	C(10)—C(11)	1.520 (5)
N(4)—C(10)	1.463 (4)	C(11)—C(12)	1.528 (4)

C(1)—N(1)—N(3)	119.3 (2)	C(2)—C(3)—C(4)	121.4 (3)
C(1)—N(1)—C(7)	129.3 (2)	C(3)—C(4)—C(5)	119.4 (3)
N(3)—N(1)—C(7)	111.4 (2)	C(4)—C(5)—C(6)	121.2 (3)
C(7)—N(2)—C(8)	108.2 (2)	C(5)—C(6)—C(1)	118.6 (3)
C(7)—N(2)—C(9)	124.8 (2)	O—C(7)—N(1)	129.4 (2)
C(8)—N(2)—C(9)	126.9 (2)	O—C(7)—N(2)	126.9 (2)
N(1)—N(3)—C(8)	104.5 (2)	N(1)—C(7)—N(2)	103.7 (2)
C(9)—N(4)—C(10)	116.4 (2)	N(2)—C(8)—N(3)	112.2 (2)
C(9)—N(4)—C(13)	116.2 (2)	N(2)—C(8)—C(12)	124.0 (2)
C(10)—N(4)—C(13)	114.4 (2)	N(3)—C(8)—C(12)	123.7 (2)
N(1)—C(1)—C(2)	121.2 (2)	N(2)—C(9)—N(4)	115.2 (2)
N(1)—C(1)—C(6)	117.8 (2)	N(4)—C(10)—C(11)	116.8 (2)
C(2)—C(1)—C(6)	121.0 (2)	C(10)—C(11)—C(12)	114.4 (3)
C(1)—C(2)—C(3)	118.5 (3)	C(8)—C(12)—C(11)	114.7 (2)

distances and bond angles are summarized in Table 2. Fig. 1 shows the atom-numbering scheme of the molecule and Fig. 2 shows the molecular packing.

Related literature. The known reaction behaviour of 5-heterospiro-substituted 2-phenyl-1,2,4-triazolidin-3-ones upon action of a variety of oxidizing agents is oxidative ring opening to 1-isocyanato-1-(phenylazo)-heterocycloalkanes, which rearrange to 1,5-heteroannulated 1,2-dihydro-2-phenyl-3*H*-1,2,4-triazol-3-ones on treatment with acid (Gstach, Seil, Schantl, Gieren, Hübner & Wu, 1986; Seil, 1989; Gstach & Seil, 1990*b*, *c*). The observed unusual 4,5-annulation of the azoheterocycle in the title compound is a consequence of both the reaction path of the oxidation and the availibility of the free electron pair of the tertiary amine function of the starting material.

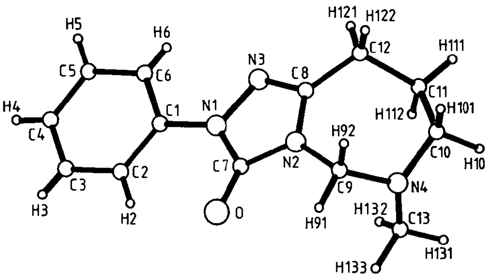

Fig. 1. Perspective view of the molecule showing the atom-numbering scheme. Radii of the atoms are arbitrary.

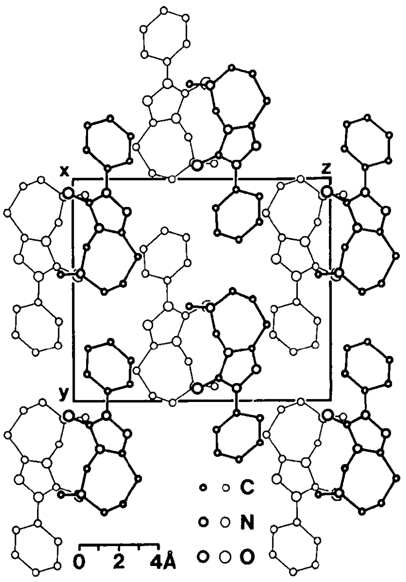

Fig. 2. Packing of the molecules in the unit cell.

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