chloro and fluoro derivatives. Unfortunately, above 170 K, torsional oscillation averages out the presence of any disorder.

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The Crystal Structures of *s*-Triazolo[4,3-*b*]pyridazine, *s*-Triazolo[1,5-*b*]pyridazine and Tetrazolo[1,5-*b*]pyridazine

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The crystal structures of three azolopyridazines have been determined with three-dimensional Mo $K\alpha$ X-ray diffraction data. The crystal data at 20 (1)°C are as follows: *s*-triazolo[4,3-*b*]pyridazine, $C_5H_4N_4$, $M_r = 120{\cdot}1$, monoclinic, $P2_1/c$, $a = 7{\cdot}378$ (2), $b = 10{\cdot}845$ (2), $c = 7{\cdot}533$ (2) Å, $\beta = 118{\cdot}78$ (2)°, $V = 528{\cdot}3$ Å^3, $D_m = 1{\cdot}52$ (5), $D_x = 1{\cdot}510$ g cm^{-3}, $Z = 4$; *s*-triazolo[1,5-*b*]pyridazine, $C_5H_4N_4$, $M_r = 120{\cdot}1$, monoclinic, $P2_1$, $a = 3{\cdot}737$ (2), $b = 6{\cdot}647$ (2), $c = 10{\cdot}756$ (2) Å, $\beta = 93{\cdot}30$ (2)°, $V = 266{\cdot}7$ Å^3, $D_m = 1{\cdot}52$ (5), $D_x = 1{\cdot}496$ g cm^{-3}, $Z = 2$; tetrazolo[1,5-*b*]pyridazine, $C_4H_3N_5$, $M_r = 121{\cdot}1$, monoclinic, $P2_1/n$, $a = 6{\cdot}148$ (2), $b = 8{\cdot}026$ (2), $c = 10{\cdot}509$ (2) Å, $\beta = 94{\cdot}84$ (2)°, $V = 516{\cdot}7$ Å^3, $D_m = 1{\cdot}57$ (5), $D_x = 1{\cdot}557$ g cm^{-3}, $Z = 4$. The structures were solved with direct methods and refined with full-matrix least-squares techniques to R values of 0·055, 0·085, 0·054 and R_w values of 0·053, 0·106 and 0·040 respectively. The molecules are essentially planar. Most of the bond lengths and angles are within normal ranges for aromatic heterocyclic systems, apart from the N(5)–C(6) and C(7)–C(8) bonds in *s*-triazolo[4,3-*b*]pyridazine [1·296 (3) and 1·354 (3) Å] and tetrazolo[1,5-*b*]pyridazine [1·298 (3) and 1·353 (3) Å] which demonstrate a considerable double-bond character.

Introduction

The crystal structures of *s*-triazolo[4,3-*b*]pyridazine (hereinafter STPI), *s*-triazolo[1,5-*b*]pyridazine (STPII) and tetrazolo[1,5-*b*]pyridazine (TP) were determined as part of a continuing study of the parent bicyclic heterocyclic systems. The present X-ray investigation was motivated by the desire to obtain accurate geometries (bond lengths and angles) of these azolopyridazines with a bridgehead N atom in order to find a possible correlation between the structure and the chemical reactivity.

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Experimental

Crystals suitable for X-ray analysis were obtained by cooling the corresponding warm solution to room temperature. The choice of a space group was made from an inspection of the systematic absences on Weissenberg and/or precession photographs and from the distribution of $|E|$ values. Unit-cell dimensions were obtained from a least-squares fit of the 2θ values of 20–25 high-order reflections measured on a CAD-4 diffractometer with Mo $K\alpha_1$ radiation [$\lambda = 0{\cdot}70926$ Å, $t = 20\ (1)$°C]. The diffraction data were collected on an automatic computer-controlled Enraf–Nonius CAD-4 four-circle diffractometer with Mo $K\alpha$ radiation and a graphite monochromator by an ω–2θ scan technique (moving crystal–moving counter) with a variable scan rate. Three reference reflections were monitored at intervals of 100 reflections to correct for intensity fluctuations and crystal orientation. The crystal and intensity-measurement data are summarized in Table 1. The data were corrected for variations in reference reflections and Lorentz–polarization effects, but not for absorption. The three structures were solved in a routine manner with *MULTAN* 74 (Main, Woolfson, Lessinger, Germain & Declercq, 1974). *E* maps with the highest combined figure of merit (CFOM) obtained with the unit-weighting of ABSFOM, ψ_0 and RESID revealed the locations of all the C and N atoms. The structure refinements were done by full-matrix least squares, minimizing the function $\Sigma w(|F_o| - k|F_c|)^2$, where the weighting function was determined empirically:

$$w = w_F w_S$$

where

$$w_F(|F_o| < A) = (|F_o|/A)^C,$$
$$w_F(|F_o| > B) = (B/|F_o|)^P,$$
$$w_F(A < |F_o| < B) = 1{\cdot}0$$

Table 1. *Crystal and intensity-measurement data summary for STPI, STPII and TP at* 20 (1) °C

	STPI	STPII	TP
Molecular formula	$C_5H_4N_4$	$C_5H_4N_4$	$C_4H_3N_5$
Molecular weight	120·1	120·1	121·1
Crystallization solvent	Ethanol	Chloroform/*n*-hexane	Ethanol
Crystal system	Monoclinic	Monoclinic	Monoclinic
Space group	$P2_1/c$ (No. 14)	$P2_1$ (No. 4)	$P2_1/n$ (No. 14)
Unit-cell parameters			
a (Å)	7·378 (2)	3·737 (2)	6·148 (2)
b (Å)	10·845 (2)	6·647 (2)	8·026 (2)
c (Å)	7·533 (2)	10·756 (2)	10·509 (2)
β (°)	118·78 (2)	93·30 (2)	94·84 (2)
Volume (Å^3)	528·3	266·7	516·7
Z	4	2	4
D_x (g cm^{-3})	1·510	1·496	1·557
D_m (g cm^{-3}) (flotation)	1·52 (5)	1·52 (5)	1·57 (5)
Shape of crystal	Prismatic	Prismatic	Prismatic
Size of crystal (mm)	0·62 × 0·54 × 0·13	0·15 × 0·20 × 0·70	0·20 × 0·25 × 0·50
Prominent axis	(001)	(100)	(100)
2θ scan width (°)	0·6 + 0·2 tan θ	0·8 + 0·2 tan θ	0·6 + 0·2 tan θ
Scan rate (° min^{-1}): minimum	0·91	2·01	0·91
maximum	20·11	20·11	20·11
Background	$\frac{1}{4}$ of the scan time at each of the scan limits		
$2\theta_{\max}$ (°)	60	60	60
Maximum scan time (s)	60	40	60
Aperture (mm)	2·5 + 0·9 tan θ	2·8 + 0·9 tan θ	2·5 + 0·9 tan θ
Reference reflections	$10\bar{2}$, $1\bar{2}1$, $2\bar{1}2$	$\bar{1}11$, $\bar{1}\bar{1}1$, 111	$23\bar{3}$, 230, 221
Intensity decrease	–	22·7	–
Measured reflections	6380 ($\pm h$, $\pm k$, $\pm l$)	2380 ($\pm h$,$\pm k$,$\pm l$)	6063 ($\pm h$,$\pm k$,$\pm l$)
Averaged reflections	1552	837	1510
Mean discrepancy on *I*(%)	3·2 (for 5705 reflections)	4·0 (for 2206 reflections)	2·7 (for 5421 reflections)
Observed reflections	820	431	917
Unobserved reflections	732[$I < 3\sigma(I)$]	406[$I < 1{\cdot}5\sigma(I)$]	593[$I < 3\sigma(I)$]
$\sigma(I)$ based on	counting statistics		
Linear absorption coefficient (cm^{-1})	1·136	1·125	1·225
Distribution of $|E|$ values			
$\langle E^2 \rangle$	1·000	1·000	1·000
$\langle |E^2 - 1| \rangle$	1·012	0·783	0·994
$\langle |E| \rangle$	0·788	0·859	0·791
$|E| > 1$ (%)	28·9	35·2	30·0
$|E| > 2$ (%)	5·3	2·0	5·1
$|E| > 3$ (%)	0·5	0·0	0·3

and

$$w_S(\sin\theta < G) = (\sin\theta/G)^J,$$
$$w_S(\sin\theta > H) = (H/\sin\theta)^K,$$
$$w_S(G < \sin\theta < H) = 1{\cdot}0.$$

Difference Fourier syntheses revealed all the H atoms at the expected positions. However, the positions of the H atoms were recalculated with a C–H distance of 1·08 Å. An isotropic temperature factor B of 3·5 Å^2 was assumed for all the H atoms and was kept fixed throughout the refinement. The final difference electron-density maps were featureless. The details of the structure determination and the final refinement parameters together with the weighting-function parameters are given in Table 2. Atomic scattering factors for H were those of Stewart, Davidson & Simpson (1965) and for other atoms those of Cromer & Mann (1968).

All calculations were carried out on the CDC Cyber 72 computer at RRC Ljubljana. The XRAY 72 system of crystallographic programs (Stewart, Kruger, Ammon, Dickinson & Hall, 1972) was used.

Table 3 lists the final atomic coordinates of STPI, STPII and TP.*

Table 2. *Structure determination and refinement summary*

	STPI	STPII	TP
Structure determination			
Number of $\lvert E\rvert$ values	250	218	250
$\lvert E\rvert$ threshold	1·36	1·02	1·36
CFOM	2·000	2·840	2·000
ABSFOM	1·087	1·046	1·145
ψ_0	3075	1614	2744
RESID	31·79	33·13	27·79
Final refinement cycle			
Scale factor (k)	0·972	0·741	0·884
$R = \Sigma\lvert\Delta F\rvert/\Sigma\lvert F_o\rvert$	0·055	0·085	0·054
$R_w = [\Sigma\, w(\Delta F)^2/\Sigma\, wF_o{}^2]^{1/2}$	0·053	0·106	0·040
Average shift/error	0·023	0·009	0·225
Maximum shift/error	0·125	0·109	0·829
Data(m)-to-variable(n) ratio	10·0	5·3	11·2
$[\Sigma\, w(\Delta F)^2/(m-n)]^{1/2}$	0·258	0·468	0·227
Final difference map			
Maximum $\Delta\rho$(e Å^{-3})	0·15	0·24	0·13
Weighting-function parameters			
A	4·0	1·8	5·0
B	12·0	5·0	13·0
C	1·5	3·0	1·5
D	4·0	2·0	4·0
G	0·33	0·33	0·30
H	0·44	0·44	0·45
J	3·5	3·0	3·5
K	1·0	1·0	1·0

Table 3. *Fractional coordinates* (×10^4, *for* H ×10^3) *with e.s.d.'s in parentheses*

	x	y	z
STPI			
N(1)	4893 (3)	2516 (2)	7648 (3)
N(2)	5356 (3)	1287 (2)	8054 (3)
C(3)	3840 (4)	632 (2)	6681 (4)
N(4)	2365 (3)	1402 (2)	5350 (2)
N(5)	532 (3)	1081 (2)	3712 (3)
C(6)	−581 (4)	2022 (2)	2723 (3)
C(7)	−14 (4)	3271 (2)	3225 (3)
C(8)	1818 (4)	3565 (2)	4848 (3)
C(8a)	3072 (3)	2568 (2)	5993 (3)
H(3)	377	−36	662
H(6)	−204	185	142
H(7)	−105	398	229
H(8)	229	451	525
STPII			
N(1)	2027 (19)	4846 (16)	4154 (6)
C(2)	496 (25)	3213 (18)	3593 (8)
N(3)	141 (18)	3219 (16)	2367 (7)
N(4)	1629 (17)	5025	2105 (6)
N(5)	1859 (18)	5678 (14)	953 (6)
C(6)	3420 (26)	7640 (17)	860 (8)
C(7)	4571 (28)	8617 (15)	1883 (9)
C(8)	4333 (24)	7918 (17)	3078 (9)
C(8a)	2766 (17)	6012 (16)	3189 (7)
H(2)	−97	193	405
H(6)	311	781	4
H(7)	589	981	196
H(8)	492	869	393
TP			
N(1)	3262 (2)	−16 (2)	3347 (2)
N(2)	2232 (3)	−279 (2)	2182 (2)
N(3)	371 (3)	524 (2)	2019 (2)
N(4)	200 (2)	1331 (2)	3120 (1)
N(5)	−1532 (2)	2306 (2)	3325 (1)
C(6)	−1387 (3)	2953 (2)	4461 (2)
C(7)	378 (3)	2702 (2)	5412 (2)
C(8)	2083 (3)	1723 (2)	5160 (2)
C(8a)	1983 (2)	1002 (2)	3936 (2)
H(6)	−272	376	470
H(7)	37	330	634
H(8)	346	149	586

Discussion

Although azolopyridazines with a bridgehead N atom are generally considered to be fully aromatic 10π-electron systems, some reactions [catalytic hydrogenation in which 7,8-dihydro and 5,6,7,8-tetrahydro derivatives were isolated (Kač, Kovač, Stanovnik & Tišler, 1975; Kadaba, Stanovnik & Tišler, 1976; Polanc, Verček, Šek, Stanovnik & Tišler, 1974), opening of the six-membered pyridazine ring either by cleavage of the N(4)–N(5) or N(5)–C(6) double bond with nucleophiles (Pollak, Polanc, Stanovnik & Tišler, 1972; Bezeg, Stanovnik, Šket & Tišler, 1972; Polanc *et al.*, 1974) and photochemical addition and cycloaddition reactions (Bradshaw, Stanovnik & Tišler, 1973*a*, *b*; Bradshaw, Tišler & Stanovnik, 1974; Maas &

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* Lists of structure factors and anisotropic thermal parameters have been deposited with the British Library Lending Division as Supplementary Publication No. SUP 33077 (16 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 13 White Friars, Chester CH1 1NZ, England.

Table 4. *Bond distances* (Å) *and bond angles* (°) *for s-triazolo*[4,3-*b*]*pyridazine*

N(1)–N(2)	1·373 (3)	N(2)–N(1)–C(8a)	106·4 (2)
N(1)–C(8a)	1·322 (2)	N(1)–N(2)–C(3)	108·9 (2)
N(2)–C(3)	1·308 (3)	N(5)–N(4)–C(3)	127·2 (2)
C(3)–N(4)	1·355 (3)	N(5)–N(4)–C(8a)	126·8 (2)
N(4)–N(5)	1·366 (2)	C(3)–N(4)–C(8a)	106·0 (2)
N(4)–C(8a)	1·365 (3)	N(4)–N(5)–C(6)	113·4 (2)
N(5)–C(6)	1·296 (3)	N(2)–C(3)–N(4)	109·1 (2)
C(6)–C(7)	1·415 (4)	N(5)–C(6)–C(7)	125·1 (2)
C(7)–C(8)	1·354 (3)	C(6)–C(7)–C(8)	120·4 (2)
C(8)–C(8a)	1·414 (3)	C(7)–C(8)–C(8a)	116·5 (2)
		N(1) C(8a)–N(4)	109·6 (2)
		N(1)–C(8a)–C(8)	132·6 (2)
		N(4)–C(8a)–C(8)	117·8 (2)

Table 5. *Bond distances* (Å) *and bond angles* (°) *for s-triazolo*[1,5-*b*]*pyridazine*

N(1)–C(2)	1·352 (15)	C(2)–N(1)–C(8a)	102·7 (7)
N(1)–C(8a)	1·337 (11)	N(1)–C(2)–N(3)	117·3 (9)
C(2)–N(3)	1·318 (11)	C(2)–N(3)–N(4)	101·0 (8)
N(3)–N(4)	1·359 (11)	N(3)–N(4)–N(5)	122·3 (6)
N(4)–N(5)	1·320 (10)	N(3)–N(4)–C(8a)	110·7 (6)
N(4)–C(8a)	1·383 (10)	N(5)–N(4)–C(8a)	127·0 (6)
N(5)–C(6)	1·326 (14)	N(4)–N(5)–C(6)	114·6 (7)
C(6)–C(7)	1·390 (14)	N(5)–C(6)–C(7)	123·5 (8)
C(7)–C(8)	1·373 (14)	C(6)–C(7)–C(8)	121·3 (9)
C(8)–C(8a)	1·404 (14)	C(7)–C(8)–C(8a)	115·7 (9)
		N(1)–C(8a)–N(4)	108·2 (8)
		N(1)–C(8a)–C(8)	134·1 (8)
		N(4)–C(8a)–C(8)	117·7 (7)

Table 6. *Bond distances* (Å) *and bond angles* (°) *for tetrazolo*[1,5-*b*]*pyridazine*

N(1)–N(2)	1·346 (2)	N(2)–N(1)–C(8a)	105·7 (1)
N(1)–C(8a)	1·324 (2)	N(1)–N(2)–N(3)	112·3 (2)
N(2)–N(3)	1·312 (2)	N(2)–N(3)–N(4)	104·9 (1)
N(3)–N(4)	1·338 (2)	N(3)–N(4)–N(5)	122·5 (1)
N(4)–N(5)	1·353 (2)	N(3)–N(4)–C(8a)	109·5 (1)
C(4)–C(8a)	1·359 (2)	N(5)–N(4)–C(8a)	128·0 (1)
N(5)–C(6)	1·298 (2)	N(4)–N(5)–C(6)	112·5 (1)
C(6)–C(7)	1·426 (3)	N(5)–C(6)–C(7)	125·3 (2)
C(7)–C(8)	1·353 (3)	C(6)–C(7)–C(8)	120·0 (2)
C(8)–C(8a)	1·406 (2)	C(7)–C(8)–C(8a)	116·3 (1)
		N(1)–C(8a)–N(4)	107·7 (1)
		N(1)–C(8a)–C(8)	134·4 (1)
		N(4)–C(8a)–C(8)	117·9 (1)

Bradshaw, 1977; Bradshaw, Tueller, Baxter, Maas & Carlock, 1977)] suggested that these compounds had localized N(5)–C(6) and C(7)–C(8) double bonds. The C(7)–C(8) double-bond character was also indicated by the magnitude of the coupling constant $J_{7,8}$ = 8·9–9·5 Hz (Japelj, Stanovnik & Tišler, 1969; Pirc, Stanovnik & Tišler, 1973; Polanc, Verček, Stanovnik & Tišler, 1973).

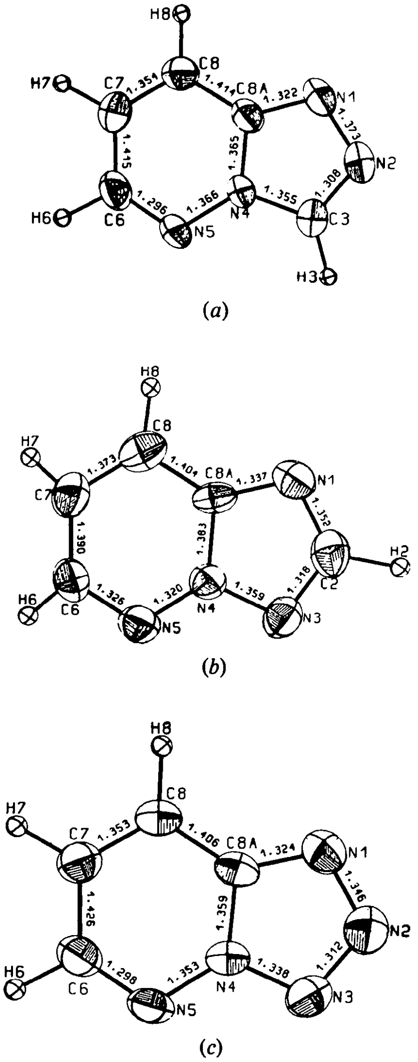
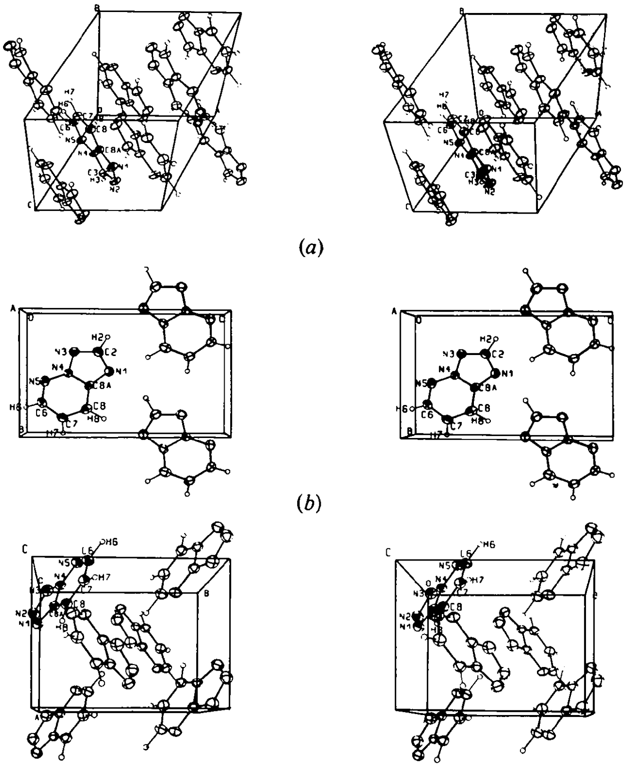


Fig. 1. The molecules of (*a*) STPI, (*b*) STPII and (*c*) TP viewed along the normal to the mean plane. The atoms are represented by thermal ellipsoids drawn at the 50% probability level (Johnson, 1965).

Fig. 1 shows molecules of STPI, STPII, and TP, together with values of the interatomic bond distances. Interatomic distances and angles with estimated standard deviations are given in Tables 4, 5 and 6. From an inspection of Tables 4, 5 and 6 it can be seen that the N(5)–C(6) and C(7)–C(8) bond lengths exhibit double-bond character, especially in STPI [1·296 (3) and 1·354 (3) Å] and TP [1·298 (3) and 1·353 (3) Å], whereas in STPII the values [1·326 (14) and 1·373 (14) Å] suggest an aromatic character, which is generally in accordance with the chemical reactivity of these compounds. However, it must be pointed out that the distances in STPII are less accurate owing to the moderate accuracy of this structure determination. The other bond lengths and angles are within the normal ranges of similar heterocyclic systems.



(a)

(b)

(c)

Fig. 2. Stereoscopic views of the unit cells as seen (*a*) along [010] for STPI, (*b*) along [100] for STPII and (*c*) along [001] for TP.

The molecules are planar, described by the equations: STPI: $-0{\cdot}8218X + 0{\cdot}0006Y + 0{\cdot}5698Z = 2{\cdot}1815$ Å; STPII: $0{\cdot}8990X - 0{\cdot}4300Y - 0{\cdot}0828Z = -1{\cdot}0746$ Å; and TP: $0{\cdot}5019X + 0{\cdot}7916Y - 0{\cdot}3485Z = -0{\cdot}3747$ Å, where $X = ax \sin\beta$, $Y = by$ and $Z = ax\cos\beta + cz$. The largest deviations from the least-squares planes are: STPI: 0·011, 0·008 and −0·012 Å for N(1), C(6) and C(8); STPII: 0·009, −0·019 and 0·015 Å for N(3), C(6) and C(7); and TP: −0·007, −0·008 and 0·012 Å for N(2), C(7) and C(8a) respectively.

The angles between the pyridazine and azole parts of the molecules (0·7° in STPI, 0·4° in STPII and 0·8° in TP) are not significantly different from zero. These small values are probably due to molecular packing.

Stereoscopic views of the molecular packing showing the crystal chemical units are in Fig. 2. The packing arrangements are quite different and there are no abnormally short contacts between the molecules.

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