

The Crystal Structure of 2,3,7,8-Tetrachloro-5,10,11,12-tetrathia-1,4,6,9-tetraazatricyclo[5.3.1.1.^{2,6}]dodeca-3,8-diene, (SNCCl)₄

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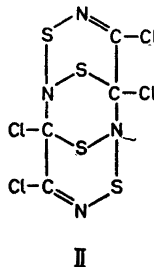
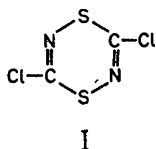
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A three-dimensional crystal structure analysis has shown the compound of empirical formula SNCCl which is obtained from the reaction of ammonia and trichloromethanesulphenyl chloride to be the tricyclic compound (SNCCl)₄.

The crystals are monoclinic with cell dimensions $a = 6.275 \pm 0.005$ Å, $b = 13.205 \pm 0.01$ Å, $c = 7.556 \pm 0.005$ Å, and $\beta = 115.9 \pm 0.3^\circ$, and with space group $P2_1/a$ (C_{2h}^5 , No. 14). There are two tetrameric molecules per cell implying the molecular symmetry $\bar{1}(C_i)$.

The diffuse scattering observed on the photographs of the $h0l$ zone is explained qualitatively in terms of the Difference-Fourier-Transform.

The reaction between trichloromethanesulphenylchloride, Cl₃CSCl, and ammonia in a benzene-water system has been shown by Senning¹ to yield a colourless crystalline compound of empirical formula SNCCl. This compound was believed to be 3,6-dichloro-1,4-dithia-2,5-diazine I, however the chemical reactions were not in agreement with this formula² and so an X-ray investigation of the compound was begun to determine the molecular structure.



Whilst the structure determination was in progress it was discovered³ from the mass spectrum that the molecule was really a tetramer.

The X-ray study shows the compound to be tetrameric and to have the formula II.

EXPERIMENTAL

The compound, which was kindly provided by Senning, crystallised from benzene as colourless needles elongated in the [100] direction.

Intensity data were obtained by a variety of methods; a total of 719 independent non-zero intensities were measured. Layers $0kl$ to $4kl$, and $h0l$, were measured from integrated equi-inclination Weissenberg photographs using the multiple film technique; intensities with optical density $0.1 < D < 1.0$ were measured photometrically. The reflections with $D < 0.1$ were estimated visually by means of a calibration strip. Sufficient data were measured by both methods to enable the two sets of data to be placed on the same scale. Layers $4kl$ to $6kl$ were estimated visually from timed equi-inclination Rinsky-Retigraph photographs. All these photographs were taken with $\text{CuK}\alpha$ radiation and with a crystal which was roughly cylindrical and with a radius such that $\mu r \sim 1$. Timed precession photographs were used to obtain visual intensities for the $hk0$ zone of reflections. In this case $\text{MoK}\alpha$ radiation was used, the crystal was equant with a mean radius such that $\mu r \sim 0.4$. No corrections were made for absorption.

CRYSTAL DATA

$\text{S}_4\text{N}_4\text{C}_4\text{Cl}_4$, $M = 364.2$. m.p. = 201 ± 0.5 .³ Monoclinic, $a = 6.275 \pm 0.005$ Å, $b = 13.205 \pm 0.01$ Å, $c = 7.556 \pm 0.005$ Å, $\beta = 115.9 \pm 0.3^\circ$, $U = 563.2$ Å³, $D_m = 2.19$ (by flotation), $Z = 2$, $D_c = 2.15$, $F(000) = 368$. Space group $P2_1/a$ (C_{2h}^5 , No. 14). $\text{CuK}\alpha$ and $\text{MoK}\alpha$ radiations, single crystal oscillation, Weissenberg, and precession photographs. Linear absorption coefficients $\mu = 180$ cm⁻¹ for $\text{CuK}\alpha$ and $\mu = 17.2$ cm⁻¹ for $\text{MoK}\alpha$. The crystals are often twinned with (100) as the twin plane and [102] as the twin axis.

The coordinates and the thermal parameters, bond lengths and angles, torsion angles, and the observed and calculated structure factors, are listed in Tables 3, 4, 5, and 7.

STRUCTURE DETERMINATION AND REFINEMENT

A three-dimensional Patterson function was calculated using unitary structure factors as coefficients; however, it was not possible to interpret this sharpened Patterson function in terms of structure I, nor, when it was known that the molecular weight should have been doubled was it possible to fit a 12-membered ring model. The high concentration of peaks on the section $v = 0$ and on the Harker-section $v = \frac{1}{2}$ made it impossible to locate more than one heavy atom from the Harker peaks. Structure factors calculated for this atom (S(2)) gave an R -factor of 0.61.

A vector-convergence map was obtained with S(2) as a searcher-atom by calculating a Fourier synthesis with $F_{\text{obs}}^2 \times U_{\text{calc}}/f_s$ as coefficients. This procedure may be shown⁴ to be similar to the weighted Fourier synthesis of Woolfson⁵ for cases when only a small amount of the scattering material has been located. Four possible positions for the remaining three heavy atoms were chosen, structure factor calculations were made using S(2) and the 4 possible selections of 3 out of the 4 possible positions (all atoms being regarded as sulphur). The residuals for the possible arrangements are shown in Table 1.

Table 1. Residuals calculated for a sulphur atom S(2) at (0.38, 0.15, 0.16) and at three of the following: A(0.20, 0.65, 0.24), B(0.32, 0.65, 0.46), C(0.24, 0.425, 0.08), D(0.18, 0.425, 0.70).

Combination	$R = \frac{\sum F_o - k F_c }{\sum F_o }$	$r = \frac{\sum (F_o - k F_c)^2}{\sum F_o^2}$
S(2), A, B, C	0.53	0.33
S(2), A, B, D	0.56	0.37
S(2), A, C, D	0.53	0.31
S(2), B, C, D	0.42	0.18

A Fourier synthesis calculated with the signs obtained from combination S2, B, C, D gave the positions of the light atoms. These positions (with the chlorine and sulphur atoms given their appropriate scattering curves and the carbon curve used for the light atoms) gave an *R*-factor of 0.30. A further Fourier synthesis followed by least-squares refinement using the diagonal approximation reduced *R* to 0.15.

The temperature factors for the light atoms, calculated assuming that all the atoms are carbon atoms, are shown in Table 2. In each case the nitrogen atoms have lower temperature factors than the carbon atoms. The difference between the values for C(1) and N(1) is $\sim 2\sigma$, there is a similar difference between the values for C(2) and N(2). The values obtained on refining with the correct scattering curves are also shown in Table 2 as is the number

Table 2. Isotropic temperature factors of the light atoms, B (all C) from the refinement in which all the light atoms were assumed to be carbon atoms, and B (C and N) from the refinement in which the carbon and nitrogen atoms were given their appropriate scattering curves. The estimated standard deviation of these values is $\sim 0.3 \text{ \AA}^2$.

Atom	Coordination No.	B (all C)	B (C and N)
N(1)	3	1.59 \AA^2	2.47 \AA^2
N(2)	2	2.16 \AA^2	3.10 \AA^2
C(1)	4	2.36 \AA^2	2.27 \AA^2
C(2)	3	2.86 \AA^2	2.80 \AA^2

of atoms to which each atom is bound. A difference-map calculated with the structure factors from the refinement in which all the light atoms were assumed to be carbon atoms showed small peaks in the neighbourhood of the nitrogen atoms.

The choice of N(1) and N(2) as nitrogen is in agreement with the stereochemistry and the valencies of carbon and nitrogen.

Least-squares refinement of coordinates and anisotropic thermal parameters reduced *R* to 0.121 for 716 non-zero reflections. Towards the end of the refinement 3 reflections which appeared to be suffering from extinction were removed, these are marked with an E in the final Table 7 of observed and calculated structure factors. If these planes are included the *R*-factor is 0.125. A final

three-dimensional difference Fourier synthesis showed no peaks larger than $1 \text{ e}\text{\AA}^{-3}$.

The final coordinates and thermal parameters together with their standard deviations are listed in Table 3. The standard deviations were estimated by Cruickshank's ⁶ method.

Table 3.

Coordinates (in \AA) and their standard deviations $\times 10^4$.

Cl(1)	-1.1732	(50)	1.0418	(44)	2.2869	(48)
Cl(2)	2.0923	(51)	2.1326	(47)	3.4423	(45)
S(1)	-1.5654	(42)	0.9747	(40)	-0.6797	(45)
S(2)	0.8698	(50)	1.9093	(41)	-1.1018	(46)
N(1)	-0.4445	(147)	0.6415	(124)	-1.5240	(141)
N(2)	1.4897	(174)	2.2624	(126)	0.6741	(163)
C(1)	-0.1746	(180)	0.7408	(148)	1.2098	(176)
C(2)	1.0418	(183)	1.7191	(145)	1.5901	(184)

Thermal parameters together with their standard deviations (in $\text{\AA}^2, \times 10^4$).

	u_{11}	$\sigma(u_{11})$	u_{22}	$\sigma(u_{22})$	u_{33}	$\sigma(u_{33})$	u_{12}	$\sigma(u_{12})$	u_{13}	$\sigma(u_{13})$	u_{23}	$\sigma(u_{23})$
Cl(1)	525	(25)	456	(23)	355	(19)	33	(19)	267	(19)	-20	(18)
Cl(2)	510	(25)	502	(24)	269	(18)	-100	(20)	79	(18)	-23	(18)
S(1)	333	(19)	318	(19)	304	(18)	70	(16)	113	(16)	27	(15)
S(2)	554	(26)	305	(19)	327	(19)	-52	(18)	229	(19)	30	(16)
N(1)	419	(76)	296	(66)	255	(60)	-4	(57)	104	(60)	8	(53)
N(2)	730	(104)	193	(59)	413	(75)	-82	(66)	281	(77)	-40	(58)
C(1)	477	(93)	292	(76)	361	(82)	-57	(68)	300	(78)	-33	(66)
C(2)	424	(88)	235	(70)	391	(82)	-38	(65)	155	(75)	68	(66)

Computational details

The weighting scheme used for the refinement using Danielsen's diagonal least-squares program was $w = 1/(10 + |F_{\text{obs}}|)$, and for Grønbaek's block-diagonal program $w = 1/(a + |F_{\text{obs}}| + c|F_{\text{obs}}|^2)$ where a and c were selected so that the average value of $w(F_{\text{obs}} - F_{\text{calc}})^2$ was independent of the magnitude of F_{obs} . The scattering curves were those of *International Tables* ⁷ approximated by Bassi's ⁸ polynomials calculated from the values of the scattering factors at $\sin \theta/\lambda = 0.1, 0.2, 0.3, 0.5,$ and 0.7 , (the carbon curve used was that of Freeman ⁹). Fourier syntheses were calculated using a machine-order program written by S. Lauesen. All the calculations were carried out on the Aarhus University GIER computer.

DIFFUSE SCATTERING

Weissenberg photographs taken about the [010] axis with $\text{CuK}\alpha$ radiation revealed considerable diffuse scattering.

The thermal diffuse scattering of X-rays by molecular crystals has been discussed by Hoppe ¹⁰ and by Amorós, Canut and Bujosa. ¹¹ The positions of the diffuse peaks are given approximately by the square of the Fourier-Transform of one molecule, or better, by the Difference-Fourier-Transform (the D.F.T.) which is the square of the Fourier-Transform of one molecule at 0°K minus

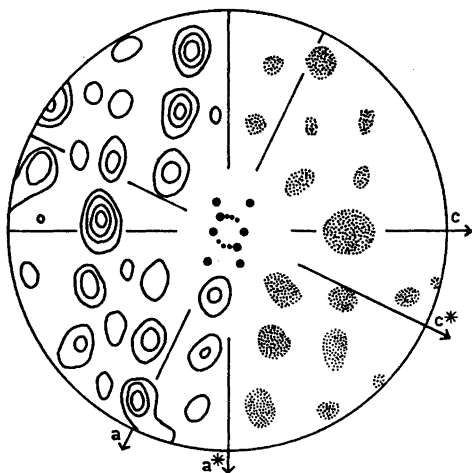


Fig. 1. Diffuse scattering in the $h0l$ projection. The observed scattering is represented by groups of dots, the D.F.T. is shown by contours at equal arbitrary intervals. The $h0l$ projection of one molecule is shown superimposed at the origin, the large circles representing the heavy atoms.

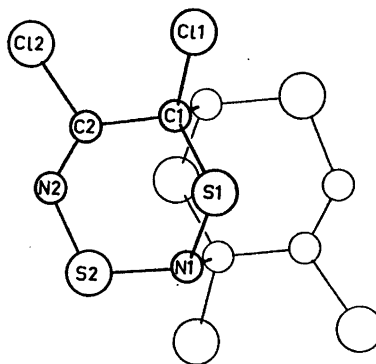


Fig. 2. The molecule viewed perpendicular to the plane through C1-C2-N2-S2-N1.

Table 4.

Bond lengths and their standard deviations (in Å).

S(1) — C(1)	1.806	0.014
Cl(1) — C(1)	1.785	0.022
Cl(2) — C(2)	1.734	0.016
S(1) — N(1)	1.705	0.018
S(2) — N(1)	1.740	0.014
S(2) — N(2)	1.644	0.016
C(1) — N(1)	1.491	0.019
C(2) — N(2)	1.301	0.028
C(1) — C(2)	1.476	0.023

Angles and their standard deviations (in degrees).

Cl(1) — C(1) — S(1)	103.4	0.85
Cl(1) — C(1) — N(1)	108.6	1.23
Cl(1) — C(1) — C(2)	110.4	1.23
S(1) — C(1) — N(1)	113.55	1.01
S(1) — C(1) — C(2)	110.8	1.19
N(1) — C(1) — C(2)	109.9	1.31
C(1) — C(2) — N(2)	126.4	1.46
C(1) — C(2) — Cl(2)	118.7	1.43
N(2) — C(2) — Cl(2)	115.7	1.43
S(1) — N(1) — C(1)	112.3	1.21
S(2) — N(1) — C(1)	114.1	1.02
S(1) — N(1) — S(2)	109.1	0.75
S(2) — N(2) — C(2)	130.0	1.45
N(1) — S(1) — C(1)	96.9	0.77
N(1) — S(2) — N(2)	107.9	0.80

the square of the Fourier-Transform at the temperature at which the measurements were made.

The positions of the maxima together with the calculated D.F.T. are shown in Fig. 1.

Table 5. Torsion angles φ (in degrees) with their standard deviations.

	φ	$\sigma(\varphi)$
S(2) — N(1) — C(1) — Cl(1)	60.1	0.8
S(2) — N(1) — C(1) — C(2)	— 179.1	1.1
S(2) — N(1) — C(1) — S(1)	— 54.3	0.9
S(1) — N(1) — C(1) — S(1)	71.3	0.9
S(1) — N(1) — C(1) — C(2)	— 53.5	1.1
S(1) — N(1) — C(1) — Cl(1)	— 174.3	0.8
Cl(1) — C(1) — S(1) — N(1)	— 179.5	0.7
C(2) — C(1) — S(1) — N(1)	62.3	1.1
N(1) — C(1) — S(1) — N(1)	— 62.0	1.0
C(1) — S(1) — N(1) — S(2)	— 67.9	0.7
C(1) — S(1) — N(1) — C(1)	61.0	1.0
Cl(1) — C(1) — C(2) — Cl(2)	41.7	0.9
S(1) — C(1) — C(2) — Cl(2)	155.5	0.9
N(1) — C(1) — C(2) — Cl(2)	— 78.1	1.1
S(1) — C(1) — C(2) — N(2)	— 29.2	1.4
Cl(1) — C(1) — C(2) — N(2)	— 143.1	1.4
N(1) — C(1) — C(2) — N(2)	97.1	1.5
C(1) — C(2) — N(2) — S(2)	— 4.1	1.1
Cl(2) — C(2) — N(2) — S(2)	171.3	0.9
C(2) — N(2) — S(2) — N(1)	— 2.07	1.4
N(2) — S(2) — N(1) — S(1)	43.0	0.9
N(2) — S(2) — N(1) — C(1)	— 84.3	1.1

Table 6. Distances, Δ , from the least-squares best plane through C(1)—C(2)—N(2)—S(2)—N(1). The standard deviations, σ , are the r.m.s. values.

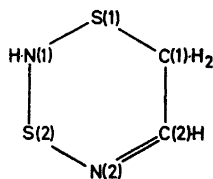
	Δ (in Å)	σ (in Å)	Δ/σ
C(1)	+0.0481	0.0163	3.0
C(2)	—0.0389	0.0164	—2.4
N(2)	—0.0153	0.0156	—1.0
S(2)	+0.0210	0.0046	4.6
N(1)	—0.0407	0.0138	—3.0
Cl(1)	+0.9985	0.0047	
Cl(2)	—0.3125	0.0048	
S(1)	+1.0030	0.0042	

DISCUSSION

The molecule, which is tricyclic, is best described pictorially (Fig. 2). The bond lengths and angles together with their standard deviations are listed in Table 4, and the torsion angles in Table 5.

The centre (SNC)₂ ring is in the chair form and has dimensions similar to those in 1,4-dithiane,¹² which suggests that little strain has been introduced by the addition of the side rings. Space group considerations require that the molecule has a centre symmetry.

With the exception of S(1) the outer (SNC)₂ rings are essentially, although not accurately, planar (Table 6).



III

The possible conformations for the "parent" ring III are, by analogy with those of cyclohexene,¹³ the half chair and the half-boat forms. The atoms C(1), C(2), N(2), and S(2) must lie in a plane as a result of the carbon-nitrogen double bond. The geometry of the ring is such that there are two possible half-chair forms having five atoms almost in a plane and one (S(1) or N(1)) out of the plane and a boat form with S(1) and N(1) on the same side of the C(1)—C(2)—N(2)—S(2) plane. The conformation observed in this case is that with one sulphur atom out of the plane. (In the case where there are 5 atoms in a plane the "half-chair" could equally well be described as a "half-boat").

The atom Cl(2) would be expected to lie in the plane, but is 0.31 Å out of it. The atom is on the opposite side of the plane from Cl(1) and the twisting of the C(2)—Cl(2) bond out of the plane (by some 10°) helps to reduce the strain arising from the close contact between Cl(1) and Cl(2) (3.145 Å, s.d. 0.007 Å compared with the sum of the van der Waals radii¹⁴ 3.6 Å). The torsion angle Cl(1)—C(1)—C(2)—Cl(2) is increased from 30° to 42°. This close contact also causes a distortion of the angles about C(1), the angle S(1)—C(1)—Cl(1) being only 103.4°.

The carbon-sulphur bond length (1.806 Å) agrees well with the value (1.817 Å) usually found¹⁵ for single S—C bonds in saturated heterocyclic compounds. The two carbon-chlorine distances (1.734 Å, 1.785 Å) are in agreement with the average values,¹⁵ 1.719 Å for a single *sp*² carbon-chlorine bond, and 1.767 Å for a single *sp*³ carbon-chlorine bond.

The carbon-nitrogen bond length in the centre ring (1.491 Å) is typical for a single bond whereas that of the side ring is 1.301 Å. Whilst there is a wealth of information on carbon-nitrogen single¹⁵ (1.479 Å), conjugated¹⁵

Table 7. Observed and calculated structure factors.

h	k	l	Obs	Calc	h	k	l	Obs	Calc	h	k	l	Obs	Calc	h	k	l	Obs	Calc
1	0	0	292	-222	1	0	0	292	-222										
0	2	0	80	56	0	2	0	80	56										
0	0	2	81	56	0	0	2	81	56										
0	5	0	49	-25	0	5	0	49	-25										
0	6	0	288	257	0	6	0	288	257										
0	7	0	117	174	0	7	0	117	174										
0	8	0	85	-76	0	8	0	85	-76										
1	1	1	121	117	1	1	1	121	117										
1	2	1	140	-103	1	2	1	140	-103										
1	4	1	52	-80	1	4	1	52	-80										
1	5	1	70	87	1	5	1	70	87										
1	7	1	153	179	1	7	1	153	179										
1	8	1	77	-95	1	8	1	77	-95										
2	1	2	128	-102	2	1	2	128	-102										
2	2	2	109	-102	2	2	2	109	-102										
2	3	2	95	96	2	3	2	95	96										
2	4	2	75	-73	2	4	2	75	-73										
2	5	2	203	194	2	5	2	203	194										
2	6	2	77	71	2	6	2	77	71										
2	8	2	141	125	2	8	2	141	125										
2	9	2	102	-141	2	9	2	102	-141										
2	10	2	84	-76	2	10	2	84	-76										
2	11	2	65	65	2	11	2	65	65										
2	12	2	125	145	2	12	2	125	145										
2	13	2	126	-135	2	13	2	126	-135										
2	14	2	231	236	2	14	2	231	236										
2	15	2	112	-139	2	15	2	112	-139										
2	16	2	66	-82	2	16	2	66	-82										
2	17	2	137	139	2	17	2	137	139										
2	18	2	181	174	2	18	2	181	174										
2	19	2	148	-153	2	19	2	148	-153										
2	20	2	175	-99	2	20	2	175	-99										
2	21	2	170	-228	2	21	2	170	-228										
2	22	2	87	-118	2	22	2	87	-118										
2	23	2	209	199	2	23	2	209	199										
2	24	2	96	-75	2	24	2	96	-75										
2	25	2	69	-20	2	25	2	69	-20										
2	26	2	124	-178	2	26	2	124	-178										
2	27	2	171	-168	2	27	2	171	-168										
2	28	2	127	-285	2	28	2	127	-285										
2	29	2	97	102	2	29	2	97	102										
2	30	2	226	-178	2	30	2	226	-178										
2	31	2	208	-25	2	31	2	208	-25										
2	32	2	375	-307	2	32	2	375	-307										
2	33	2	161	-183	2	33	2	161	-183										
2	34	2	72	144	2	34	2	72	144										
2	35	2	74	87	2	35	2	74	87										
2	36	2	102	86	2	36	2	102	86										
2	37	2	128	-128	2	37	2	128	-128										
2	38	2	126	128	2	38	2	126	128										
2	39	2	76	93	2	39	2	76	93										
2	40	2	78	95	2	40	2	78	95										
2	41	2	252	-226	2	41	2	252	-226										
2	42	2	160	-172	2	42	2	160	-172										
2	43	2	309	-399	2	43	2	309	-399										
2	44	2	86	85	2	44	2	86	85										
2	45	2	89	89	2	45	2	89	89										
2	46	2	67	96	2	46	2	67	96										
2	47	2	191	15	2	47	2	191	15										
2	48	2	272	901	2	48	2	272	901										
2	49	2	157	169	2	49	2	157	169										
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2	57	2	101	90	2	57	2	101	90										
2	58	2	72	101	2	58	2	72	101										
2	59	2	84	-99	2	59	2	84	-99										
2	60	2	117	-186	2	60	2	117	-186										
2	61	2	94	-96	2	61	2	94	-96										
2	62	2	56	-27	2	62	2	56	-27										
2	63	2	98	-86	2	63	2	98	-86										
2	64	2	241	282	2	64	2	241	282										
2	65	2	47	42	2	65	2	47	42										
2	66	2	238	235	2	66	2	238	235										
2	67	2	94	36	2	67	2	94	36										
2	68	2	258	-394	2	68	2	258	-394										
2	69	2	71	61	2	69	2	71	61										
2	70	2	139	-131	2	70	2	139	-131										
2	71	2	99	-126	2	71	2	99	-126										
2	72	2	129	86	2	72	2	129	86										
2	73	2	471	-941	2	73	2	471	-941										
2	74	2	256	-307	2	74	2	256	-307										
2	75	2	116	110	2	75	2	116	110										
2	76	2	112	-112	2	76	2	112	-112										
2	77	2	40	94	2	77	2	40	94										
2	78	2	272	-289	2	78	2	272	-289										
2	79	2	94	-74	2	79	2	94	-74										
2	80	2	66	63	2	80	2	66	63										
2	81	2	349	346	2	81	2	349	346										
2	82	2	72	69	2	82	2	72	69										
2	83	2	48	-54	2	83	2	48	-54										

(1.33 Å), and triple¹⁵ (1.16 Å), bonds, there is little to be found on double bonds.

The π -bond order of C(2)—N(2) as calculated from the bond order-bond length curve of Goodwin and Porte¹⁶ is ~ 0.8 . Hall and Llewellyn,¹⁷ however, deduce a π -bond order of only 0.6 for a C—N bond of length 1.301 Å in formamidoxime.

The two sulphur-nitrogen bonds S(1)—N(1) (1.705, s.d. 0.018 Å) and S(1)—N(2) (1.740, s.d. 0.014 Å) do not differ significantly in length. The weighted mean value, 1.725, s.d. 0.011 Å, is typical for a single bond.¹⁵ The third bond S(2)—N(2) (1.644, s.d. 0.016) differs significantly from 1.725 Å. The shortening corresponds to a π -bond order of ~ 0.5 (*cf.* Chapman and Waddington¹⁸).

The double bond is thus shared between the neighbouring S(2)—N(2) and N(2)—C(2) bonds. This delocalisation might explain the increase of the S(2)—N(2)—C(2) angle to 130°.

The carbon-carbon-distance (1.476 Å) agrees with the value expected (1.510 Å)¹⁵ for a single bond adjacent to a double bond.

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