Table 1. *Comparison of corrected and accurately known values of the equi-inclination angle*

Substance	Layer	μ_{calc}	s_{calc} (mm)	s_{act} (mm)	δ	μ_{cor}	μ
Struvite	3	22°18′	9·6	9·5	22° 1′	22°10′	22° 8′
	4	30 24	13·8	13·5	29 52	30 8	30 9
	5	39 14	19·2	18·7	38 30	38 52	38 53
Roesslerite	5	19 21	8·3	8·3	19 18	19 19	19 32
	6	23 25	10·3	10·5	23 54	23 40	23 39
	7	27 38	12·3	12·8	28 22	28 0	27 54
	8	32 0	14·8	15·2	32 40	32 20	32 20
	9	36 36	17·6	18·3	37 40	37 8	36 59

angle are still considerably better than the initial ones. Presumably the accuracy could be increased further by irradiating only the tip of the crystal. Hence it would appear that this method is capable of high accuracy and, in addition, has two advantages over Sayre's (1954) method; namely, it is perfectly general and there is no need to take Weissenberg photographs before applying it. However, it has a disadvantage that incorrect settings have, in general, a larger effect on Sayre's parameter, $d\omega$, than the error in the layer line screen movement, leading, in theory, to lower accuracy. To some extent this is counteracted by the fact that the spots on the Weissenberg photograph are likely to be more elongated than those on the oscillation photograph, owing to strain (combined with the velocity factor) and to the Phillips (1954) effect.

In practice, of course, one need not take photographs of each layer line, only of the most accurate.

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The Crystal and Molecular Structure of Neothiobinupharidine Dihydrobromide Tetrahydrate *

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Crystals of neothiobinupharidine dihydrobromide tetrahydrate, $C_{30}H_{42}O_2N_2S.2HBr.4H_2O$, are orthorhombic, space group $P2_12_12_1$, with $a=14·71$, $b=16·07$, $c=14·68$ Å. The structure was solved by the heavy-atom method and the refinement was carried out by the least-squares procedure. The final R value is 0·097. All six-membered rings are in chair conformation and all substituents in equatorial positions. There is some molecular disorder associated with an approximate non-crystallographic twofold axis. The alkaloid molecules are embedded between hydrogen-bonded sheets composed of water molecules and bromide ions.

Introduction

A new class of natural products was discovered by Achmatowicz & Bellen (1962), who isolated four alkaloids containing sulphur from *Nuphar luteum* (L) *Sm.* (yellow water lily). Subsequently, Achmatowicz & Wróbel (1964) isolated another substance from the same source which with empirical formula $C_{30}H_{42}O_2N_2S$ and this was named neothiobinupharidine. The authors investigated this compound using ultraviolet, infrared and nuclear magnetic resonance spectra, but it was mostly on the basis of mass spectra that Achmatowicz, Banaszek, Spiteller & Wróbel (1964) proposed a complete structure (I). At the same time, Prof. Achmatowicz very kindly sent us crystals of neothiobinupharidine dihydrobromide tetrahydrate for X-ray structure analysis. This investigation revealed a somewhat different structure (II) which was first reported in a preliminary communication (Birnbaum, 1965).

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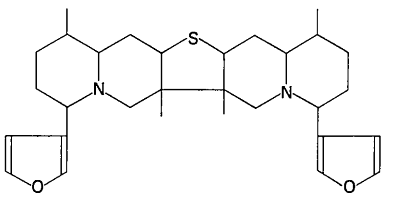
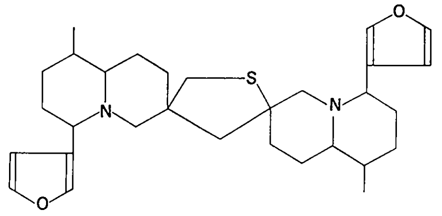


(I)



(II)

Experimental

Crystal data

Empirical formula $C_{30}H_{42}O_2N_2S.2HBr.4H_2O$;
F.W. 728·5
Orthorhombic
$a = 14{\cdot}71 \pm 0{\cdot}02$ Å
$b = 16{\cdot}07 \pm 0{\cdot}02$
$c = 14{\cdot}68 \pm 0{\cdot}02$
$V = 3471{\cdot}4$ Å^3
$D_x = 1{\cdot}39$ g.cm^{-3}
$D_m = 1{\cdot}38$ g.cm^{-3}
$Z = 4$
$\mu = 41{\cdot}2$ cm^{-1} (Cu $K\alpha$), $F(000) = 1520$
Absent reflexions: $h00$ when h is odd, $0k0$ when k is odd, $00l$ when l is odd. Space group: $P2_12_12_1$.

The crystals were recrystallized from aqueous ethanol, giving colorless prisms elongated parallel to [100]. The data were recorded with an equi-inclination Weissenberg camera, using the multiple-film technique and nickel-filtered Cu $K\alpha$ radiation ($\lambda = 1{\cdot}5418$ Å).

The crystals were deteriorating as a result of X-ray exposure and thus it was necessary to use three of them in order to collect the zones ($0kl$) to ($12kl$) and another one to collect ($h0l$) to ($h5l$). All crystals had been cut to an approximately cylindrical shape to give a maximum beam path of 0·25 mm. The intensities were estimated visually; their range was about 1 to 2500. Of the 3967 possible independent reflexions within the Cu sphere only 2022 were observed. No corrections were made for absorption.

Structure determination

After applying Lorentz, polarization and spot shape corrections all the values of F^2 were correlated by using common reflexions from a-axis and b-axis photographs. The layer scale factors were obtained by a simple averaging procedure. All the data were then put

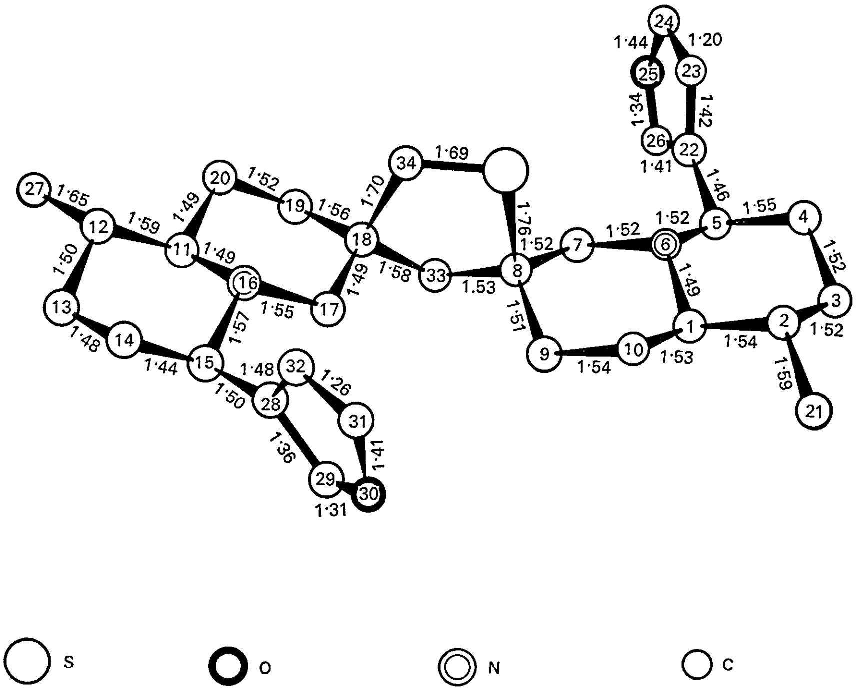


Fig. 1. A perspective view of the molecule showing intramolecular bond distances.

on a single scale. The positions of the bromide ions were determined from a three-dimensional Patterson synthesis sharpened to point atoms at rest. Structure factors were calculated based on the bromide atoms only, giving an $R=43\%$. In this and all future calculations the atomic scattering factors used were those given in *International Tables for X-ray Crystallography* (1962) for carbon, nitrogen, oxygen, sulphur (p. 202) and bromide (p. 206).

A model of the first three-dimensional Fourier synthesis revealed a flat five-membered ring, a six-membered ring in chair conformation, and a sequence of five other atoms with approximately correct bond lengths and angles. All these atoms were assumed to be carbon. There was also a peak, higher than any of those mentioned above, which was assumed to correspond to the sulphur atom. The second Fourier synthesis showed all the atoms of the molecule itself except C(34). To find its position as well as the positions of the four water molecules a difference-Fourier synthesis was calculated. All the missing atoms were now located and it was also possible to identify one of the nitrogen atoms. The position of the other nitrogen atom was assigned from biogenetic considerations. At this stage

Table 1. *The final atomic coordinates and their estimated standard deviations**

	x/a	y/b	z/c	$\sigma(x/a)$	$\sigma(y/b)$	$\sigma(z/c)$
C(1)	0·2726	0·5648	−0·1048	±0·0012	±0·0010	±0·0010
C(2)	0·2082	0·6401	−0·0981	0·0010	0·0009	0·0010
C(3)	0·2590	0·7194	−0·0737	0·0012	0·0011	0·0013
C(4)	0·3125	0·7063	0·0138	0·0012	0·0009	0·0012
C(5)	0·3791	0·6318	0·0048	0·0010	0·0009	0·0011
N(6)	0·3235	0·5549	−0·0175	0·0007	0·0007	0·0007
C(7)	0·3871	0·4803	−0·0221	0·0010	0·0009	0·0012
C(8)	0·3415	0·3989	−0·0475	0·0011	0·0009	0·0010
C(9)	0·2935	0·4132	−0·1368	0·0015	0·0013	0·0014
C(10)	0·2231	0·4838	−0·1279	0·0014	0·0011	0·0015
C(11)	0·4137	0·0688	−0·0349	0·0010	0·0010	0·0010
C(12)	0·3867	−0·0239	−0·0098	0·0014	0·0011	0·0013
C(13)	0·3543	−0·0655	−0·0951	0·0016	0·0012	0·0019
C(14)	0·2699	−0·0229	−0·1235	0·0012	0·0012	0·0017
C(15)	0·2870	0·0624	−0·1490	0·0011	0·0009	0·0011
N(16)	0·3279	0·1099	−0·0651	0·0008	0·0007	0·0008
C(17)	0·3436	0·2019	−0·0919	0·0011	0·0008	0·0009
C(18)	0·3906	0·2492	−0·0187	0·0010	0·0009	0·0010
C(19)	0·4774	0·2021	0·0139	0·0013	0·0010	0·0017
C(20)	0·4515	0·1148	0·0444	0·0014	0·0011	0·0012
C(21)	0·1553	0·6519	−0·1921	0·0017	0·0019	0·0014
C(22)	0·4313	0·6219	0·0887	0·0010	0·0010	0·0012
C(23)	0·5273	0·6207	0·1000	0·0013	0·0017	0·0016
C(24)	0·5483	0·6164	0·1792	0·0015	0·0017	0·0020
O(25)	0·4696	0·6043	0·2363	0·0013	0·0012	0·0011
C(26)	0·3995	0·6079	0·1781	0·0017	0·0018	0·0013
C(27)	0·4809	−0·0690	0·0253	0·0015	0·0013	0·0019
C(28)	0·2019	0·1046	−0·1814	0·0013	0·0011	0·0013
C(29)	0·1171	0·1067	−0·1439	0·0012	0·0015	0·0017
O(30)	0·0583	0·1484	−0·1925	0·0013	0·0014	0·0015
C(31)	0·1064	0·1748	−0·2701	0·0015	0·0017	0·0019
C(32)	0·1900	0·1595	−0·2618	0·0017	0·0031	0·0014
C(33)	0·4206	0·3373	−0·0566	0·0012	0·0008	0·0013
C(34)	0·3142	0·2692	0·0651	0·0016	0·0008	0·0008
S	0·2651	0·3600	0·0347	0·0004	0·0003	0·0005
Br(1)	0·1789	0·0781	0·1058	0·0002	0·0002	0·0002
Br(2)	0·0824	0·7325	0·1519	0·0002	0·0002	0·0002
W(1)	0·0436	−0·0689	0·1725	0·0011	0·0010	0·0012
W(2)	0·1003	0·2506	0·2167	0·0011	0·0010	0·0011
W(3)	0·0852	0·4189	0·1993	0·0009	0·0009	0·0009
W(4)	0·1876	0·5493	0·1253	0·0008	0·0008	0·0008

Mean standard deviations

C	0·0013 (0·019 Å)	0·0012 (0·020 Å)	0·0013 (0·020 Å)
N	0·0008 (0·011 Å)	0·0007 (0·011 Å)	0·0008 (0·011 Å)
O	0·0011 (0·016 Å)	0·0010 (0·016 Å)	0·0011 (0·016 Å)
S	0·0004 (0·006 Å)	0·0003 (0·005 Å)	0·0005 (0·007 Å)
Br	0·0002 (0·003 Å)	0·0002 (0·003 Å)	0·0002 (0·003 Å)

* In order to allow for block-diagonal approximation about 30% should be added to each e.s.d.

the value of R was 25% and the refinement of the structure by least-squares was begun.

The first five cycles* were calculated with the block-diagonal approximation and the weighting scheme used was $\sqrt{w}=1/\{1+[(F_o-37{\cdot}5)/60]^2\}^{\frac{1}{2}}$, according to one suggested by Mills & Rollett (1961). Full shifts were applied. Individual isotropic thermal vibrations were assumed for all atoms except the two bromine atoms and the sulphur, which were given anisotropic vibration parameters. After two cycles atom 25, which until this stage was assumed to be a carbon atom, had a significantly lower temperature factor than its neighbours in the five-membered ring and it was thus identified as an oxygen atom. After one more cycle O(30) was identified on the same basis. This completed the determination of the molecular structure. Two more cycles of least-squares reduced the value of R from 14·7% to 11·8%.

Subsequent refinement was carried out with the least-squares program of Smith & Cruickshank (1965), again using the block-diagonal approximation. With this program it was possible to calculate anisotropic thermal parameters for all atoms. The following weighting scheme was used:

$$\sqrt{w}=1/(10{\cdot}0+|F_o|+0{\cdot}0083|F_o|^2+0{\cdot}0001|F_o|^3)^{\frac{1}{2}}$$

(Cruickshank, Pilling, Bujosa, Lovell & Truter, 1961). The function minimized was $\Sigma w(F_o-F_c)^2$. A partial shift factor of 0·8 was applied in each cycle.

After one cycle of fully anisotropic least-squares refinement the value of R dropped to 10·1%. Refinement was continued until, in the last cycle, the average coordinate shift was equal to $0{\cdot}1\sigma$ and the maximum shift equaled $0{\cdot}38\sigma$. The final agreement index calculated for the 2022 observed reflexions is 9·7%. The final atomic coordinates are given in Table 1, and the temperature parameters are listed in Table 2. Observed and calculated structure factors are shown in Table 3.

Table 2. *The final anisotropic temperature parameters and their e.s.d.'s*

The parameters are defined by
$\exp\,[-2\pi^2(U_{11}h^2a^{*2}+U_{22}k^2b^{*2}+U_{33}l^2c^{*2}+2U_{23}klb^*c^*+2U_{31}lhc^*a^*+2U_{12}hka^*b^*)]$.

	U_{11}	U_{22}	U_{33}	$2U_{23}$	$2U_{31}$	$2U_{12}$
C(1)	0·074 (10)	0·046 (8)	0·031 (7)	0·001 (14)	−0·003 (14)	−0·003 (18)
C(2)	0·050 (8)	0·041 (8)	0·038 (7)	0·006 (14)	−0·008 (13)	0·018 (15)
C(3)	0·063 (10)	0·045 (9)	0·070 (11)	0·007 (17)	0·023 (18)	−0·005 (19)
C(4)	0·070 (10)	0·024 (7)	0·077 (11)	0·004 (15)	0·002 (20)	−0·009 (17)
C(5)	0·048 (8)	0·028 (7)	0·061 (9)	−0·025 (14)	−0·005 (15)	−0·004 (15)
N(6)	0·030 (5)	0·031 (6)	0·042 (6)	−0·011 (10)	−0·005 (10)	−0·004 (11)
C(7)	0·040 (8)	0·035 (8)	0·069 (10)	−0·033 (16)	0·011 (15)	−0·009 (14)
C(8)	0·053 (9)	0·034 (7)	0·052 (9)	0·006 (14)	0·000 (14)	−0·017 (15)
C(9)	0·108 (16)	0·071 (12)	0·077 (13)	−0·064 (22)	−0·103 (25)	0·031 (26)
C(10)	0·088 (14)	0·047 (10)	0·101 (15)	−0·044 (21)	−0·110 (26)	0·052 (22)
C(11)	0·048 (8)	0·042 (8)	0·055 (9)	0·011 (15)	−0·009 (14)	0·017 (17)
C(12)	0·083 (12)	0·052 (10)	0·076 (12)	0·021 (20)	−0·028 (22)	0·039 (20)
C(13)	0·087 (13)	0·040 (9)	0·145 (20)	0·002 (24)	0·008 (29)	0·011 (22)
C(14)	0·049 (10)	0·064 (12)	0·124 (18)	−0·001 (25)	−0·036 (23)	−0·020 (20)
C(15)	0·054 (9)	0·036 (8)	0·055 (9)	−0·026 (15)	0·007 (15)	0·019 (16)
N(16)	0·050 (7)	0·029 (5)	0·046 (6)	0·000 (11)	−0·019 (12)	0·014 (12)
C(17)	0·064 (9)	0·020 (6)	0·039 (8)	0·000 (12)	−0·022 (14)	−0·017 (14)
C(18)	0·063 (9)	0·033 (7)	0·039 (8)	−0·031 (13)	−0·004 (14)	−0·008 (15)
C(19)	0·065 (11)	0·038 (9)	0·134 (18)	0·000 (22)	−0·097 (25)	0·028 (19)
C(20)	0·088 (13)	0·050 (9)	0·058 (10)	0·011 (18)	−0·052 (20)	0·015 (20)
C(21)	0·087 (15)	0·160 (25)	0·064 (12)	0·059 (30)	0·001 (24)	0·054 (35)
C(22)	0·036 (8)	0·048 (9)	0·079 (11)	−0·050 (18)	−0·020 (16)	−0·012 (16)
C(23)	0·052 (11)	0·126 (20)	0·096 (15)	−0·065 (31)	−0·010 (22)	−0·020 (27)
C(24)	0·065 (13)	0·120 (20)	0·138 (21)	−0·087 (37)	−0·092 (28)	0·091 (29)
O(25)	0·131 (14)	0·115 (13)	0·083 (10)	−0·003 (20)	−0·065 (20)	−0·026 (25)
C(26)	0·120 (19)	0·141 (22)	0·050 (11)	0·083 (28)	−0·046 (24)	−0·085 (36)
C(27)	0·080 (13)	0·064 (12)	0·142 (21)	0·027 (29)	0·010 (30)	0·058 (25)
C(28)	0·074 (12)	0·056 (10)	0·071 (12)	−0·031 (19)	−0·031 (20)	−0·002 (20)
C(29)	0·042 (9)	0·095 (16)	0·121 (17)	−0·017 (30)	−0·002 (22)	0·045 (21)
O(30)	0·110 (13)	0·143 (16)	0·148 (16)	−0·074 (29)	−0·079 (26)	0·056 (27)
C(31)	0·069 (14)	0·114 (20)	0·127 (19)	0·073 (36)	−0·061 (28)	0·030 (27)
C(32)	0·077 (16)	0·364 (57)	0·054 (12)	−0·006 (48)	−0·021 (24)	0·059 (52)
C(33)	0·055 (9)	0·023 (7)	0·087 (12)	−0·020 (16)	0·030 (18)	0·027 (16)
C(34)	0·166 (18)	0·022 (6)	0·008 (5)	−0·020 (11)	0·062 (18)	0·041 (22)
S	0·078 (3)	0·052 (3)	0·122 (5)	0·022 (6)	0·082 (7)	0·002 (6)
Br(1)	0·108 (2)	0·105 (2)	0·102 (2)	0·059 (3)	0·118 (3)	0·031 (3)
Br(2)	0·100 (2)	0·089 (2)	0·121 (2)	−0·048 (3)	0·053 (3)	0·001 (3)
W(1)	0·092 (10)	0·088 (10)	0·116 (12)	0·046 (20)	0·002 (19)	0·005 (20)
W(2)	0·114 (12)	0·081 (10)	0·100 (10)	−0·024 (18)	0·012 (20)	0·012 (20)
W(3)	0·070 (8)	0·077 (8)	0·074 (8)	−0·016 (15)	−0·034 (14)	0·027 (16)
W(4)	0·073 (7)	0·068 (7)	0·061 (7)	−0·021 (12)	0·036 (13)	0·016 (14)

---

* In these and in all preceding calculations the programs of Ahmed, Gabe, Mair & Pippy (1963) were used.

Table 3. *Observed and calculated structure factors*

H	K	L	F OBS	F CALC
0	2	0	39.3	20.4
0	4	0	106.1	111.9
0	6	0	192.4	254.4
0	8	0	29.7	29.6
0	10	0	34.7	32.2
0	12	0	15.6	13.6
0	14	0	37.6	33.4
0	16	0	8.3	10.5
2	0	0	108.1	101.9
2	2	0	108.8	123.1
2	4	0	14.5	27.3
2	6	0	14.5	12.7
2	8	0	37.9	42.4
2	10	0	8.5	7.2
2	14	0	25.1	28.3
2	18	0	8.5	5.6
4	0	0	89.7	80.4
4	2	0	55.3	58.4
4	4	0	63.1	67.7
4	6	0	51.8	53.7
4	8	0	13.1	10.6
4	12	0	24.3	21.1
4	14	0	23.3	23.2
6	0	0	60.9	56.7
6	2	0	106.3	115.4
6	4	0	68.2	75.0
6	6	0	29.2	25.5
6	8	0	80.3	91.2
6	10	0	40.3	43.7
8	0	0	110.7	99.9
8	2	0	10.6	6.6
8	4	0	34.2	27.5
8	6	0	70.6	73.2
8	8	0	26.7	28.3
8	12	0	12.2	11.8
8	18	0	7.2	10.1
10	0	0	39.7	36.6
10	2	0	5.8	7.1
10	4	0	31.1	32.9
10	6	0	30.1	26.7
10	12	0	16.1	14.9
12	0	0	35.1	33.7
12	2	0	19.0	20.6
12	4	0	15.7	18.5
12	6	0	18.7	21.8
12	8	0	10.4	9.0
14	0	0	18.6	19.7
14	2	0	14.2	12.8
1	3	1	47.5	49.1
1	5	1	71.8	75.4
1	7	1	57.4	47.2
1	9	1	20.2	20.8
1	11	1	31.0	29.8
1	13	1	9.2	8.2
1	15	1	13.5	12.4
1	17	1	12.4	10.8
3	1	1	145.0	157.7
3	3	1	53.2	48.6
3	5	1	49.3	47.4
3	7	1	92.7	77.9
3	9	1	26.8	23.9
3	11	1	13.1	12.6
3	13	1	23.3	21.4
5	1	1	57.1	58.1
5	3	1	5.0	7.7
5	5	1	44.5	47.8
5	7	1	76.4	81.0
5	9	1	11.1	9.8
5	11	1	29.5	31.0
5	13	1	33.2	30.7
7	1	1	25.5	21.6
7	3	1	35.0	33.5
7	7	1	25.4	18.6
7	9	1	17.0	16.1
9	1	1	40.0	45.3
9	3	1	41.8	47.2
9	5	1	10.3	11.4
9	7	1	25.3	28.8
9	9	1	25.7	28.9
11	1	1	12.4	8.5
11	3	1	31.8	30.0
11	7	1	15.2	14.5
11	9	1	13.7	15.2
11	11	1	17.1	15.4
13	3	1	14.2	15.5
15	1	1	12.1	8.6
17	1	1	12.9	12.7
0	0	2	155.0	145.1
0	2	2	55.1	64.3
0	4	2	27.4	7.6
0	10	2	13.9	13.0
0	12	2	29.5	30.0
0	14	2	22.2	22.4
0	16	2	8.2	8.1
2	0	2	134.9	124.2
2	2	2	36.4	33.4
2	4	2	20.0	29.6
2	6	2	108.9	103.0
2	8	2	58.3	61.5
2	10	2	17.1	19.3
2	12	2	40.8	38.2
2	14	2	20.5	18.9
2	18	2	8.5	7.4
4	0	2	21.3	13.7
4	2	2	152.4	146.3
4	4	2	124.0	121.4
4	6	2	16.0	13.2
4	10	2	54.4	56.3
4	14	2	23.3	20.3
4	16	2	14.0	11.4
6	0	2	72.2	61.0
6	2	2	56.0	44.8
6	4	2	61.8	59.9
6	6	2	62.8	64.1
6	8	2	36.8	31.3
6	10	2	16.7	17.6
6	12	2	14.9	11.3
8	0	2	31.1	29.6
8	2	2	27.1	16.0
8	4	2	25.4	21.9
8	6	2	28.1	29.0
8	8	2	23.1	21.1
8	10	2	20.0	17.4
8	12	2	24.3	23.5
10	0	2	20.3	20.5
10	2	2	28.5	26.2
10	4	2	28.6	24.7
10	6	2	27.1	28.9
10	8	2	29.7	27.7
12	0	2	20.9	15.3
12	2	2	20.2	20.4
12	4	2	25.3	26.3
12	6	2	9.6	7.9
12	8	2	10.6	12.8
14	0	2	17.5	14.8
14	2	2	14.2	11.2
1	1	3	83.9	85.4
1	3	3	69.7	87.4
1	5	3	92.9	98.8
1	7	3	14.4	15.8
1	9	3	23.6	25.5
1	11	3	47.4	52.5
1	13	3	31.0	33.0
1	15	3	25.0	23.1
1	17	3	12.1	10.6
3	1	3	141.5	131.7
3	3	3	27.5	29.3
3	5	3	66.0	51.9
3	7	3	48.6	38.7
3	9	3	32.5	31.2
3	11	3	40.7	39.3
3	15	3	22.1	17.3
5	1	3	30.4	24.7
5	3	3	49.6	37.7
5	5	3	66.7	61.1
5	7	3	46.7	46.5
5	9	3	43.7	43.2
5	11	3	34.0	41.1
5	13	3	25.5	21.8
5	15	3	16.3	13.8
7	1	3	39.3	30.6
7	3	3	37.6	40.5
7	5	3	54.9	60.3
7	7	3	34.5	30.6
7	9	3	17.6	21.0
7	11	3	30.4	37.9
9	1	3	25.5	26.0
9	3	3	12.1	10.1
9	5	3	14.7	13.2
9	7	3	20.5	21.5
9	9	3	13.2	13.9
11	1	3	17.1	18.3
11	3	3	12.2	7.0
11	5	3	24.0	24.9
11	7	3	22.4	20.8
11	11	3	13.9	12.4
11	15	3	7.2	7.3
13	1	3	19.0	19.2
13	3	3	12.8	13.5
0	0	4	122.2	115.5
0	2	4	64.9	56.7
0	4	4	24.9	30.4
0	6	4	101.4	123.6
0	8	4	25.5	22.6
0	12	4	30.3	27.4
0	14	4	8.5	7.6
2	0	4	130.7	107.5
2	2	4	140.0	141.3
2	4	4	83.1	80.2
2	6	4	51.0	44.0
2	8	4	52.8	56.3
2	10	4	41.0	45.0
2	12	4	19.9	18.2
2	14	4	14.5	12.3
4	0	4	26.6	19.9
4	2	4	76.7	76.6
4	4	4	52.5	40.1
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4	8	4	33.1	28.4
4	10	4	38.5	33.2
4	14	4	10.4	10.9
4	16	4	9.6	12.5
6	0	4	8.2	8.3
6	2	4	61.3	58.0
6	4	4	53.9	60.2
6	8	4	23.8	25.6
6	12	4	15.0	15.0
8	0	4	77.8	85.2
8	2	4	17.4	16.5
8	4	4	10.3	8.2
8	6	4	44.3	46.5
8	8	4	18.9	12.8
8	10	4	16.7	14.5
8	12	4	12.2	14.0
10	0	4	14.7	8.8
10	2	4	13.3	9.0
10	4	4	38.4	39.4
10	6	4	20.0	18.4
10	10	4	18.6	19.4
10	12	4	9.1	8.2
12	0	4	13.2	8.3
12	2	4	16.5	18.2
12	4	4	24.9	27.1
12	6	4	10.2	9.1
12	8	4	15.2	13.4
14	0	4	22.8	18.4
14	2	4	13.9	17.5
14	4	4	9.9	11.0
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1	3	5	29.7	27.7
1	5	5	59.6	56.1
1	7	5	39.3	39.2
1	9	5	37.6	38.3
1	11	5	21.8	23.0
1	13	5	26.7	28.4
3	1	5	38.8	41.2
3	3	5	70.8	75.6
3	5	5	77.0	81.4
3	7	5	54.6	59.1
3	9	5	10.3	14.4
3	11	5	27.8	22.2
3	13	5	33.8	34.4
3	15	5	16.3	12.7
5	1	5	70.0	62.9
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5	5	5	30.1	30.8
5	7	5	34.5	33.5
5	9	5	17.2	15.8
5	11	5	23.1	18.5
7	1	5	39.3	38.9
7	5	5	18.6	16.7
7	7	5	39.3	45.3
9	1	5	20.3	19.0
9	3	5	34.9	35.5
9	5	5	38.5	42.7
9	7	5	22.0	22.1
9	9	5	13.6	12.4
11	1	5	30.6	26.0
11	3	5	11.4	15.2
11	5	5	16.7	18.9
11	13	5	8.5	12.8
13	3	5	11.0	12.1
15	1	5	9.9	6.6
15	3	5	9.7	8.5
0	0	6	37.8	39.9
0	2	6	23.8	32.2
0	4	6	61.1	66.0
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0	8	6	33.5	30.0
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2	4	6	44.6	40.6
2	6	6	43.1	45.7
2	8	6	22.8	22.6
2	14	6	20.3	17.6
4	0	6	33.8	24.0
4	2	6	32.4	26.6
4	4	6	75.2	69.6
4	6	6	16.3	17.9
4	8	6	43.3	42.1
4	10	6	44.2	38.2
4	14	6	10.2	9.1
4	16	6	9.0	10.3
6	0	6	95.1	94.4
6	2	6	31.5	28.2
6	4	6	28.9	30.3
6	6	6	52.9	54.0
6	8	6	25.7	22.7
6	10	6	18.1	17.3
8	0	6	26.0	20.8
8	2	6	27.2	24.4
8	4	6	29.5	29.5
8	6	6	21.4	21.8
8	8	6	16.4	16.2
10	0	6	32.4	31.8
10	2	6	26.0	27.1
10	4	6	20.6	18.0
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10	10	6	13.2	11.0
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12	0	6	13.2	13.0
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12	4	6	17.8	18.7
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1	9	7	39.7	39.9
1	11	7	26.3	27.1
1	13	7	19.0	20.0
1	15	7	12.5	13.4
1	17	7	10.3	8.5
3	1	7	37.2	33.9
3	3	7	55.2	52.5
3	5	7	18.1	12.0
3	9	7	29.3	29.8
3	11	7	11.8	13.3
3	13	7	11.9	12.0
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5	3	7	38.8	39.9
5	5	7	15.3	13.5
5	7	7	41.9	38.9
5	9	7	13.1	14.0
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7	5	7	32.5	32.2
7	7	7	18.3	13.3
9	1	7	28.5	27.5
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11	1	7	12.8	14.8
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11	9	7	13.7	11.7
13	1	7	16.3	17.8
13	5	7	14.4	10.8
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2	2	8	61.1	61.8
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2	10	8	22.5	22.2
2	12	8	29.0	27.4
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4	0	8	20.9	19.8
4	2	8	42.2	39.1
4	4	8	24.4	19.7
4	6	8	29.0	30.0
4	8	8	48.2	49.8
4	10	8	10.3	10.0
4	12	8	10.4	10.4
4	14	8	9.6	11.7
6	0	8	10.7	13.6
6	2	8	15.7	15.8
6	4	8	19.0	18.1
6	8	8	25.5	27.4
8	0	8	11.9	9.9
8	2	8	42.1	40.9
8	4	8	12.4	10.2
8	6	8	23.5	23.7
8	8	8	32.1	28.2
8	12	8	16.0	14.9
10	0	8	31.0	30.7
10	2	8	14.5	14.3
10	4	8	17.2	15.5
10	6	8	9.2	12.0
10	8	8	9.3	6.8
10	10	8	17.8	15.4
12	0	8	12.4	11.2
12	2	8	18.3	16.5
14	0	8	7.8	9.3
16	0	8	12.1	11.6
1	1	9	37.6	35.5
1	5	9	51.3	51.1
1	7	9	21.5	17.9
1	9	9	13.2	15.8
1	11	9	21.3	21.5
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3	3	9	12.1	10.2
3	5	9	29.3	32.6
3	7	9	40.4	40.3
3	9	9	16.7	16.6
3	11	9	18.9	18.9
3	13	9	13.9	13.2
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5	3	9	33.2	32.7
5	5	9	40.3	39.0
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5	9	9	16.7	18.6
5	11	9	28.8	27.4
7	3	9	16.1	18.1
7	5	9	18.6	16.8
7	7	9	23.9	30.4
9	1	9	26.6	21.9
9	3	9	21.3	18.0
9	5	9	10.8	9.8
11	1	9	17.9	18.5
11	5	9	13.5	14.4
11	7	9	13.7	12.0
11	9	9	9.1	11.4
13	1	9	8.0	8.1
0	0	10	18.2	16.8
0	2	10	40.8	43.4
0	4	10	26.7	25.2
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0	14	10	12.6	11.6
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2	2	10	12.6	14.3
2	4	10	22.8	22.0
2	6	10	19.6	19.7
2	8	10	17.6	16.3
2	10	10	20.6	21.8
4	0	10	17.2	15.2
4	2	10	22.2	20.9
4	4	10	16.7	17.4
6	0	10	44.5	45.9
6	2	10	24.2	23.0
6	4	10	24.9	26.7
6	6	10	14.9	20.2
8	2	10	27.5	26.5
8	4	10	10.2	11.4
12	2	10	14.5	15.3
12	4	10	11.7	15.1
12	8	10	12.9	13.0
12	10	10	7.6	11.5
14	0	10	14.7	12.5
1	1	11	31.4	31.8
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1	5	11	15.3	15.5
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1	9	11	13.5	12.8
1	11	11	18.3	15.2
3	1	11	18.9	17.6
3	3	11	37.5	35.2
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3	9	11	11.9	14.9
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5	9	11	13.5	13.3
7	3	11	28.2	30.7
9	1	11	19.9	20.8
9	5	11	17.2	19.1
11	9	11	7.6	10.3
13	1	11	11.9	9.8
13	3	11	8.3	11.7
0	2	12	20.7	19.4
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0	12	12	10.3	11.3
2	0	12	17.6	12.7
2	4	12	23.6	24.0
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4	6	12	20.9	18.1
4	10	12	9.6	12.6
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8	4	12	12.1	8.2
10	2	12	12.8	13.7
10	4	12	11.7	14.4
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10	10	12	8.6	8.6
12	0	12	10.8	7.8
1	3	13	27.5	31.6
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1	11	13	13.7	13.0
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3	3	13	11.5	14.5
3	5	13	14.0	17.3
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5	3	13	24.3	24.3
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7	1	13	10.6	11.1
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11	1	13	7.6	7.8
11	5	13	7.5	7.4
13	3	13	7.8	11.3
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0	4	14	24.7	26.0
0	8	14	15.2	13.2
2	10	14	8.2	8.1
2	12	14	6.7	6.2
4	4	14	11.1	8.7
10	2	14	7.5	6.2
1	1	15	8.9	9.7
1	5	15	8.6	9.6
3	1	15	10.7	9.7
3	3	15	12.4	14.0
3	5	15	10.6	9.6
5	1	15	9.9	6.7
9	1	15	7.8	9.7
0	0	16	7.5	2.2
1	3	17	7.1	10.1
1	3	0	66.2	80.3
1	5	0	97.3	96.7
1	7	0	52.4	57.4
1	9	0	41.7	37.3
1	11	0	78.6	84.0
1	13	0	20.5	18.3
3	1	0	57.1	47.8
3	3	0	18.1	17.7
3	5	0	17.9	16.0
3	7	0	44.5	48.7
3	9	0	56.1	53.2
3	13	0	18.5	14.2
3	15	0	43.7	35.2
3	17	0	15.2	12.1
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5	7	0	32.9	35.9
5	9	0	24.6	20.3
5	13	0	25.4	28.0
7	1	0	85.7	93.4
7	3	0	71.0	57.6
7	5	0	52.1	55.7
7	9	0	23.9	25.7
7	13	0	19.9	23.3
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9	7	0	27.4	30.5
9	11	0	19.4	21.1
11	1	0	11.4	10.9
11	3	0	24.6	22.0
11	9	0	13.7	8.9
11	15	0	7.6	8.7
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0	4	1	67.4	69.6
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0	10	1	33.5	35.3
0	14	1	25.3	27.8
0	16	1	8.3	8.5
0	18	1	7.1	6.4
2	0	1	119.8	84.3
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2	4	1	108.0	90.9
2	6	1	62.4	68.8
2	8	1	27.9	27.3
2	10	1	42.2	52.7
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2	16	1	20.0	15.6
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4	6	1	44.9	32.9
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8	6	1	24.4	21.3
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13	5	2	10.8	15.2
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0	10	3	43.7	40.3
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2	12	3	23.9	25.9
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6	4	3	82.9	74.7
6	6	3	12.9	15.0
6	8	3	19.4	21.5
6	10	3	33.9	31.4
6	12	3	18.3	20.4
8	0	3	37.9	34.9
8	2	3	71.9	78.6
8	4	3	45.7	51.5
8	6	3	18.5	14.7
8	8	3	35.0	38.1
8	10	3	16.5	18.0
8	12	3	17.2	15.1
10	0	3	33.6	35.6
10	2	3	14.4	11.6
10	4	3	26.0	24.9
10	6	3	35.7	33.9
10	8	3	17.5	18.3
12	0	3	24.4	26.7
12	2	3	19.3	19.6
12	4	3	17.5	18.8
12	6	3	19.9	21.0
12	8	3	15.0	14.4
12	10	3	10.7	9.6
14	0	3	10.7	5.5
14	2	3	14.0	14.6
1	1	4	84.6	104.9
1	3	4	13.6	11.4
1	5	4	71.5	76.8
1	7	4	27.7	25.3
1	9	4	13.3	13.2
1	11	4	17.4	18.9
1	15	4	21.0	19.7
1	17	4	11.8	11.7
3	1	4	8.0	14.3
3	3	4	46.7	46.2
3	5	4	46.0	34.9
3	7	4	50.7	47.2
3	9	4	32.8	32.5
3	11	4	38.4	36.1
3	13	4	23.8	23.2
3	15	4	11.7	8.1
5	1	4	52.6	55.9
5	3	4	69.3	64.8
5	5	4	16.8	13.7
5	7	4	60.1	60.0
5	9	4	33.4	31.5
5	11	4	13.1	11.1
5	13	4	16.8	12.0
7	1	4	41.8	41.0
7	3	4	43.8	39.8
7	5	4	24.2	22.2
7	7	4	49.6	60.2
9	1	4	36.4	28.7
9	3	4	46.1	41.0
9	5	4	11.9	10.9
9	7	4	24.4	24.3
9	9	4	18.9	17.6
9	11	4	19.7	24.6
11	1	4	29.9	29.9
11	7	4	9.3	9.8
11	9	4	14.0	15.5
13	1	4	12.8	14.0
13	3	4	9.1	7.1
13	5	4	10.7	11.9
15	1	4	15.4	14.6
0	2	5	47.4	47.1
0	4	5	27.8	32.3
0	6	5	48.5	51.2
0	8	5	31.4	33.2
0	10	5	39.7	40.0
0	12	5	42.6	40.9
0	14	5	20.6	19.5
0	16	5	7.9	3.1
0	18	5	9.0	8.3
2	0	5	57.4	62.9
2	2	5	120.4	110.0
2	4	5	18.9	16.4
2	6	5	39.7	40.3
2	8	5	21.7	24.1
2	10	5	16.0	14.1
2	12	5	14.2	12.1
2	16	5	9.5	9.8
4	0	5	101.7	96.6
4	2	5	67.8	52.1
4	4	5	39.7	32.8
4	6	5	45.8	43.9
4	8	5	51.7	46.1
4	10	5	13.6	11.6
4	14	5	23.2	19.3
4	16	5	9.3	11.3
6	0	5	15.6	15.8
6	2	5	20.5	20.2
6	4	5	28.2	28.9
6	6	5	20.3	21.7
6	8	5	29.5	31.9
6	10	5	22.8	24.4
6	12	5	21.4	19.3
8	0	5	23.8	23.1
8	2	5	21.4	19.7
8	4	5	18.5	19.7
8	6	5	38.1	38.5
8	8	5	27.5	28.3
10	0	5	9.9	6.7
10	2	5	33.6	35.2
10	4	5	19.4	18.3
10	6	5	37.9	32.8
10	8	5	35.3	34.1
10	12	5	12.6	10.6
12	0	5	39.2	45.3
12	2	5	20.1	21.5
12	4	5	9.9	10.2
12	6	5	10.4	12.1
12	8	5	10.8	14.9
12	10	5	14.9	13.3
18	0	5	7.8	7.9
1	1	6	83.2	78.2
1	3	6	22.9	31.2
1	5	6	46.8	44.8
1	7	6	42.4	43.7
1	9	6	23.6	20.8
1	11	6	15.7	18.4
1	13	6	19.0	17.0
1	15	6	9.1	10.7
3	1	6	11.8	6.3
3	3	6	27.9	31.7
3	5	6	32.4	36.8
3	7	6	44.5	43.6
3	9	6	38.4	37.8
3	11	6	24.6	32.5
3	13	6	17.0	15.7
3	15	6	17.9	14.3
5	1	6	119.9	113.4
5	3	6	28.2	24.1
5	5	6	55.9	53.0
5	7	6	56.8	55.6
5	9	6	15.4	17.9
5	11	6	34.5	33.3
5	15	6	17.6	18.9
7	1	6	17.5	14.9
7	3	6	52.0	49.0
7	5	6	37.9	37.9
7	7	6	24.9	25.9
7	9	6	19.0	19.5
9	1	6	10.0	6.9
9	3	6	11.7	9.6
9	5	6	22.1	25.7
9	7	6	39.3	39.3
9	9	6	13.9	13.9
11	1	6	27.5	26.2
11	3	6	13.5	13.9
11	5	6	9.3	10.6
11	7	6	17.0	16.2
11	9	6	9.9	10.9
13	1	6	26.6	27.1
13	3	6	8.9	10.0
13	5	6	16.5	20.6
15	1	6	9.5	9.3
0	2	7	81.8	90.5
0	4	7	111.1	121.9
0	6	7	11.5	14.2
0	10	7	37.6	43.1
0	12	7	25.3	21.7
0	16	7	7.4	7.9
2	0	7	25.0	25.4
2	2	7	65.1	67.2
2	4	7	19.7	16.4
2	6	7	20.2	16.7
2	8	7	28.6	26.9
2	10	7	9.7	11.1
2	14	7	10.0	13.0
2	18	7	6.4	11.3
4	0	7	19.7	20.3
4	2	7	33.5	35.2
4	4	7	40.3	30.0
4	6	7	14.9	14.3
4	10	7	39.2	35.1
4	14	7	9.9	10.5
6	0	7	25.6	32.6
6	2	7	32.5	31.4
6	4	7	38.6	36.2
6	6	7	27.9	30.9
6	8	7	30.3	28.7
8	0	7	25.3	21.9
8	2	7	34.0	29.6
8	4	7	45.7	48.1
8	6	7	19.6	18.0
8	8	7	16.8	17.6
8	10	7	17.2	19.5
10	0	7	23.9	23.9
10	2	7	22.8	20.8
10	4	7	22.8	21.9
10	8	7	22.8	22.2
10	10	7	9.2	11.1
10	12	7	8.5	6.9
12	0	7	13.1	8.9
12	2	7	15.6	16.0
12	4	7	18.7	15.8
14	2	7	14.9	14.4
14	4	7	9.1	10.9
16	0	7	11.7	5.4
1	1	8	68.1	74.3
1	3	8	21.0	20.0
1	5	8	25.8	24.5
1	7	8	34.5	35.7
1	13	8	16.3	18.6
1	15	8	8.5	8.3
3	1	8	15.3	17.1
3	3	8	23.2	23.5
3	5	8	13.7	13.0
3	7	8	10.7	11.0
3	9	8	11.5	10.9
3	11	8	16.8	18.6
5	1	8	20.0	16.4
5	3	8	56.8	56.3
5	5	8	34.0	27.3
7	1	8	31.2	24.7
7	3	8	26.6	26.4
7	7	8	19.0	18.2
7	9	8	19.7	26.0
9	1	8	26.8	17.8
9	3	8	34.0	28.1
9	5	8	33.5	34.6
9	9	8	19.6	19.7
11	1	8	13.2	12.8
11	5	8	23.5	25.2
11	7	8	9.9	10.1
11	11	8	8.5	8.7
13	1	8	8.5	8.4
13	3	8	10.3	12.1
15	1	8	6.9	7.4
0	2	9	13.5	11.9
0	4	9	30.1	24.7
0	6	9	21.4	19.3
0	8	9	29.0	27.4
0	10	9	31.5	31.2
0	12	9	16.7	17.3
0	14	9	18.9	21.1
2	0	9	14.4	15.3
2	2	9	34.6	31.8
2	4	9	37.5	36.5
2	6	9	26.3	24.7
2	8	9	14.0	11.7
2	12	9	25.0	21.8
4	0	9	67.5	65.7
4	2	9	12.2	10.7
4	4	9	26.6	22.4
4	6	9	41.0	39.1
4	8	9	10.2	12.7
4	10	9	10.4	14.3
4	14	9	9.1	8.2
6	0	9	34.9	39.0
6	4	9	25.7	24.0
6	6	9	27.1	28.1
8	0	9	10.4	10.0
8	2	9	16.0	15.5
8	4	9	17.6	11.4
8	6	9	20.9	17.8
8	8	9	12.2	11.5
10	0	9	21.5	23.7
10	2	9	21.3	22.7
10	4	9	21.1	20.0
10	6	9	13.2	16.6
10	8	9	9.1	11.2
10	12	9	10.3	10.0
12	2	9	19.3	19.2
12	4	9	15.2	15.0
14	4	9	7.9	9.1
1	1	10	17.8	17.6
1	5	10	14.9	13.9
1	7	10	26.0	25.5
1	9	10	9.5	13.1
1	13	10	15.2	14.6
1	15	10	7.4	9.5
3	1	10	13.9	12.9
3	3	10	23.9	24.0
3	5	10	20.3	24.0
3	7	10	24.7	26.1
5	1	10	16.0	17.8
5	3	10	24.4	24.6
5	5	10	21.3	22.6
5	7	10	31.7	29.9
5	9	10	19.4	19.4
5	11	10	18.6	15.0
7	1	10	26.3	25.5
7	3	10	10.8	10.8
7	5	10	25.3	25.8
9	1	10	12.9	16.2
9	5	10	13.6	13.3
11	1	10	11.1	13.0
13	1	10	15.0	14.5
15	3	10	9.1	12.5
0	2	11	11.0	8.3
0	4	11	19.3	20.4
0	8	11	14.6	13.5
0	12	11	19.2	19.9
2	0	11	10.0	9.3
2	2	11	28.3	28.8
2	4	11	25.0	29.6
2	6	11	14.4	14.7
2	8	11	29.0	29.4
2	14	11	11.3	12.7
4	0	11	24.3	22.9
4	6	11	25.4	20.0
4	8	11	20.9	17.8
6	0	11	20.2	21.7
6	2	11	34.5	33.8
6	4	11	15.6	19.0
6	8	11	18.3	24.2
8	0	11	17.4	16.1
8	2	11	17.8	17.8
8	4	11	12.5	12.1
8	6	11	17.1	18.2
10	2	11	16.8	17.0
10	4	11	9.3	8.7
10	8	11	11.7	11.3
14	0	11	9.1	7.4
1	1	12	24.7	24.2
1	5	12	15.7	16.5
1	7	12	19.0	17.5
1	9	12	9.3	7.5
3	1	12	18.1	16.1
3	3	12	12.9	9.8
3	11	12	10.7	9.8
5	1	12	15.4	14.4
5	5	12	13.7	9.5
5	7	12	16.5	15.1
7	3	12	11.0	11.8
9	9	12	11.1	13.3
11	1	12	13.2	11.4
11	3	12	9.0	5.8
11	5	12	13.2	16.5
0	8	13	16.3	13.4
0	10	13	10.7	8.5
2	0	13	12.5	12.5
2	2	13	14.4	14.5
2	4	13	14.5	13.0
4	0	13	31.2	33.7
4	2	13	18.7	16.4
4	4	13	16.3	14.8
4	6	13	17.8	19.0
6	0	13	16.3	15.7
6	2	13	11.4	9.8
8	0	13	8.3	7.9
8	2	13	11.3	10.0
10	2	13	9.6	6.3
10	4	13	10.7	10.8
12	0	13	16.3	15.7
12	4	13	8.3	9.6
1	7	14	17.6	13.9
3	3	14	14.4	10.3
5	1	14	8.6	7.6
7	1	14	12.9	14.0
9	1	14	7.4	8.2
0	4	15	11.1	9.8
0	10	15	5.8	7.1
4	0	15	20.5	15.3
4	4	15	9.3	4.2
4	6	15	15.2	14.8
8	0	15	9.6	7.2
8	4	15	10.4	12.0
10	0	15	11.5	7.7
1	7	16	7.1	9.1
6	4	17	9.1	10.5
1	2	0	22.5	9.2
1	4	0	52.5	50.4
1	6	0	87.5	82.9
1	8	0	20.7	13.0
1	10	0	10.8	9.8

Table 3 (*cont.*)

H	K	L	F OBS	F CALC
1	12	0	36.1	38.0
1	14	0	46.3	52.1
1	16	0	13.2	12.0
1	18	0	8.0	6.1
3	2	0	237.8	210.4
3	4	0	101.1	113.2
3	6	0	43.8	27.5
3	8	0	44.3	40.0
3	10	0	24.3	21.9
3	14	0	29.2	25.2
5	2	0	81.7	70.3
5	4	0	114.2	108.9
5	6	0	95.9	77.9
5	10	0	49.3	48.8
5	16	0	18.2	15.9
7	2	0	11.9	16.4
7	4	0	92.2	88.4
7	6	0	9.3	4.0
7	10	0	28.5	37.0
9	4	0	22.8	19.0
9	6	0	22.9	20.3
9	8	0	17.0	16.8
9	10	0	29.9	29.3
9	12	0	19.7	19.8
11	2	0	17.6	17.3
11	4	0	9.5	7.6
11	6	0	25.8	20.9
11	8	0	18.6	22.2
11	14	0	8.5	10.4
13	4	0	14.5	19.4
0	1	1	57.4	48.0
0	3	1	72.1	116.4
0	5	1	43.3	54.8
0	7	1	49.2	39.9
0	9	1	54.9	50.3
0	13	1	30.6	29.8
0	15	1	14.6	14.8
0	17	1	7.8	5.3
0	19	1	6.1	7.8
2	1	1	67.4	77.0
2	3	1	101.4	102.7
2	5	1	50.9	58.7
2	7	1	77.4	76.7
2	9	1	28.2	31.0
2	11	1	27.1	28.1
2	13	1	10.0	11.6
2	15	1	25.1	24.6
2	17	1	13.3	10.8
4	1	1	76.8	70.4
4	3	1	77.8	63.3
4	5	1	64.6	57.3
4	7	1	36.9	36.5
4	9	1	43.6	45.8
4	11	1	16.3	15.0
4	13	1	41.1	38.3
4	19	1	6.8	8.6
6	1	1	55.7	55.1
6	3	1	39.3	33.9
6	5	1	46.7	52.6
6	7	1	62.4	56.7
6	9	1	31.0	28.1
6	11	1	24.7	20.9
6	13	1	18.6	15.6
8	1	1	26.5	24.5
8	3	1	56.8	58.3
8	7	1	46.1	48.1
8	9	1	30.7	33.8
8	13	1	17.2	18.6
10	1	1	9.7	9.1
10	3	1	8.2	7.1
10	5	1	32.3	33.4
10	9	1	12.6	11.5
10	11	1	18.7	17.9
10	13	1	12.8	13.4
12	1	1	6.9	6.5
12	3	1	32.4	29.7
12	5	1	16.4	20.5
12	7	1	10.0	7.9
12	9	1	10.7	8.5
14	1	1	11.0	10.5
16	1	1	7.9	4.8
1	0	2	10.7	19.1
1	2	2	57.8	57.5
1	4	2	127.2	115.4
1	6	2	88.9	90.3
1	8	2	12.6	10.6
1	10	2	33.2	37.6
1	12	2	23.5	22.4
1	14	2	19.0	20.0
3	0	2	163.2	147.9
3	2	2	74.5	73.3
3	4	2	105.0	110.9
3	6	2	86.7	79.8
3	8	2	48.3	35.4
3	10	2	17.4	7.9
3	14	2	11.9	13.4
5	0	2	10.7	8.6
5	2	2	78.1	73.3
5	4	2	9.6	10.0
5	6	2	31.7	25.5
5	8	2	32.2	34.1
5	12	2	31.1	34.4
7	0	2	41.4	36.3
7	2	2	76.4	76.4
7	4	2	33.2	22.1
7	6	2	30.8	36.3
7	8	2	49.7	53.6
9	0	2	77.4	85.0
9	2	2	24.7	25.4
9	4	2	28.5	31.2
9	6	2	46.4	49.5
9	8	2	27.4	25.0
9	10	2	13.5	14.2
11	0	2	22.9	20.5
11	2	2	16.1	16.0
11	6	2	16.8	17.2
11	8	2	13.3	12.4
11	12	2	9.6	8.5
13	0	2	26.3	23.6
13	2	2	14.2	15.7
15	2	2	15.4	15.6
15	4	2	9.5	9.1
17	0	2	10.6	10.9
0	1	3	126.0	143.0
0	3	3	104.7	119.9
0	5	3	10.8	9.7
0	7	3	35.0	37.6
0	9	3	61.3	73.7
2	1	3	134.9	126.2
2	3	3	33.4	24.5
2	5	3	84.9	96.5
2	7	3	57.0	60.7
2	9	3	43.8	44.4
2	11	3	13.2	12.1
2	13	3	10.2	12.5
2	15	3	20.5	20.2
2	19	3	6.9	8.4
4	1	3	98.4	95.5
4	3	3	71.4	68.9
4	5	3	57.4	61.3
4	7	3	36.3	34.5
4	9	3	25.7	22.1
4	11	3	13.7	13.4
4	13	3	29.3	27.9
4	19	3	9.1	8.7
6	1	3	82.6	85.2
6	3	3	25.8	28.5
6	5	3	51.7	51.2
6	7	3	44.1	36.9
6	9	3	27.7	21.2
6	11	3	22.9	22.5
8	1	3	12.5	13.0
8	3	3	35.6	35.8
8	5	3	8.6	10.8
8	7	3	39.6	38.5
8	9	3	15.7	16.7
10	1	3	34.9	32.3
10	3	3	42.2	49.7
10	5	3	34.5	35.7
10	7	3	27.5	31.1
10	9	3	15.7	16.4
10	11	3	16.1	14.1
12	1	3	27.7	26.7
12	7	3	20.6	21.9
14	1	3	8.9	7.0
16	1	3	9.3	9.2
1	0	4	30.6	31.4
1	2	4	67.2	69.6
1	4	4	50.6	64.5
1	6	4	32.4	33.3
1	8	4	59.0	61.4
1	10	4	14.2	13.0
1	12	4	28.8	28.0
1	14	4	13.5	14.5
1	18	4	7.5	7.4
3	0	4	135.0	132.1
3	2	4	89.9	88.8
3	4	4	119.6	112.7
3	6	4	51.1	52.5
3	8	4	26.2	25.0
3	10	4	42.1	40.6
3	12	4	28.3	33.2
3	14	4	11.9	7.8
5	0	4	14.7	8.9
5	2	4	52.8	46.1
5	4	4	76.8	69.5
5	6	4	41.8	41.5
5	8	4	35.1	35.1
5	10	4	26.5	26.5
5	12	4	39.3	31.4
5	16	4	15.0	13.6
7	0	4	79.6	73.9
7	2	4	26.3	25.4
7	4	4	32.7	33.6
7	6	4	39.9	42.1
7	8	4	24.2	20.0
9	0	4	37.2	34.7
9	2	4	31.0	26.7
9	4	4	32.3	30.6
9	6	4	32.7	30.6
9	10	4	13.7	15.1
11	2	4	25.8	29.9
11	4	4	22.7	26.4
11	6	4	27.0	25.0
11	8	4	16.8	17.6
11	10	4	14.0	13.9
11	12	4	16.3	14.4
13	0	4	17.6	19.0
13	2	4	21.7	21.6
15	0	4	15.2	10.7
0	1	5	20.7	13.9
0	3	5	61.8	67.8
0	5	5	22.5	25.3
0	7	5	14.0	9.5
0	9	5	57.1	61.5
0	15	5	11.7	10.8
2	1	5	45.8	37.1
2	3	5	16.7	20.7
2	5	5	28.9	27.4
2	7	5	39.3	39.8
2	9	5	45.3	47.9
2	11	5	43.3	41.2
2	13	5	17.8	16.1
2	17	5	8.6	8.0
2	19	5	6.0	6.7
4	1	5	60.1	54.6
4	3	5	26.6	22.7
4	5	5	48.3	40.9
4	7	5	33.5	33.3
4	9	5	20.3	22.3
4	11	5	20.1	17.7
4	13	5	31.4	27.6
4	15	5	9.9	10.0
6	1	5	85.0	77.0
6	3	5	65.6	63.1
6	5	5	42.9	40.1
6	7	5	30.6	29.1
6	9	5	19.6	20.2
6	11	5	21.0	24.4
8	1	5	25.3	20.3
8	3	5	26.0	25.7
8	5	5	33.8	32.3
8	7	5	10.7	12.2
8	9	5	23.3	23.2
8	11	5	24.3	21.0
8	15	5	10.3	9.1
10	1	5	8.5	7.4
10	3	5	9.1	11.1
10	5	5	23.3	26.0
10	7	5	26.5	23.7
10	9	5	16.0	16.3
10	11	5	9.2	8.7
12	3	5	13.2	12.6
12	5	5	10.2	8.6
14	1	5	10.6	10.5
14	3	5	18.3	18.2
1	0	6	19.0	19.7
1	2	6	32.3	25.6
1	4	6	39.0	38.6
1	6	6	53.5	55.6
1	8	6	27.4	26.2
1	10	6	43.7	47.0
1	12	6	18.7	21.3
1	14	6	18.9	20.9
1	16	6	8.5	9.0
1	18	6	6.7	7.2
3	0	6	61.6	53.7
3	2	6	65.1	63.0
3	4	6	57.0	54.8
3	6	6	28.1	28.8
3	8	6	38.5	34.5
3	10	6	37.8	38.3
3	12	6	26.6	31.5
5	0	6	68.4	60.8
5	2	6	64.2	63.9
5	4	6	46.7	44.1
5	6	6	46.4	46.6
5	8	6	12.1	14.3
5	10	6	16.1	19.0
5	12	6	21.7	20.7
7	0	6	60.1	57.3
7	2	6	39.3	35.9
7	4	6	46.3	38.3
7	6	6	20.6	26.3
7	8	6	22.5	25.9
9	0	6	63.1	60.4
9	2	6	13.7	12.9
9	4	6	37.9	38.0
9	6	6	36.0	32.3
9	10	6	13.9	15.0
11	0	6	21.5	22.0
11	2	6	22.9	19.7
11	4	6	12.4	10.5
11	6	6	9.6	8.0
11	10	6	13.7	13.9
13	0	6	13.7	14.0
13	4	6	12.2	13.8
15	4	6	8.5	9.5
0	1	7	72.6	75.4
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0	9	7	23.1	22.8
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0	17	7	6.5	6.1
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2	3	7	13.1	15.5
2	5	7	26.8	26.0
2	7	7	43.2	42.6
2	9	7	18.9	18.3
2	11	7	20.2	22.7
2	13	7	14.5	14.2
2	15	7	13.5	12.3
4	1	7	69.9	68.2
4	3	7	47.8	41.9
4	5	7	49.6	49.5
4	7	7	38.2	36.3
4	9	7	22.0	20.6
4	11	7	23.2	23.7
4	15	7	9.3	8.8
6	1	7	28.5	30.8
6	3	7	43.9	42.6
6	5	7	27.4	25.2
6	7	7	21.7	22.5
6	9	7	14.6	14.4
8	1	7	49.6	45.6
8	3	7	15.0	17.9
8	5	7	32.8	34.0
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8	9	7	12.1	12.4
10	1	7	27.5	26.5
10	3	7	23.9	26.0
10	5	7	21.7	23.6
10	7	7	18.5	15.5
10	9	7	16.1	16.7
10	13	7	7.8	8.1
12	1	7	25.0	24.9
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12	7	7	10.7	13.2
12	13	7	7.1	11.5
14	1	7	10.0	8.8
14	3	7	11.5	10.0
1	0	8	17.6	15.7
1	2	8	61.2	66.0
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1	6	8	37.7	36.3
1	8	8	32.5	32.4
1	12	8	23.3	27.0
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3	2	8	31.8	32.7
3	4	8	49.7	45.3
3	6	8	31.0	27.2
3	8	8	11.1	12.1
3	10	8	16.7	15.3
3	12	8	18.9	16.4
5	0	8	74.2	75.1
5	2	8	16.4	13.5
5	4	8	16.3	13.5
5	6	8	30.1	33.1
5	10	8	21.5	19.9
7	0	8	30.8	32.3
7	2	8	21.5	21.6
7	4	8	22.4	22.1
7	6	8	22.8	28.8
9	0	8	22.2	18.6
9	2	8	26.6	26.5
9	4	8	28.9	27.6
9	8	8	19.7	17.9
11	2	8	23.3	23.3
11	4	8	19.3	22.8
11	6	8	9.9	9.8
13	0	8	8.3	8.7
13	2	8	18.9	19.8
13	4	8	11.5	11.2
15	0	8	15.3	13.3
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0	5	9	45.6	47.1
0	7	9	11.0	14.2
0	9	9	23.3	24.0
0	11	9	14.6	12.6
0	15	9	14.2	12.5
2	1	9	40.6	37.4
2	3	9	19.9	20.8
2	5	9	10.3	12.1
2	7	9	19.2	17.7
2	9	9	14.2	15.4
2	13	9	14.0	12.7
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4	5	9	26.6	26.2
4	7	9	17.2	18.4
4	9	9	27.4	22.7
6	1	9	27.7	25.5
6	3	9	31.0	30.0
6	5	9	10.3	14.3
6	7	9	14.7	16.8
6	9	9	15.2	15.9
8	1	9	41.5	35.5
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8	5	9	16.7	14.4
8	7	9	21.0	20.7
8	9	9	17.1	14.5
10	1	9	23.5	21.0
10	5	9	14.7	14.6
10	7	9	9.2	12.1
10	11	9	8.0	11.0
12	1	9	11.5	14.2
16	3	9	6.4	9.0
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1	2	10	14.4	13.6
1	4	10	42.6	44.6
1	8	10	16.1	13.6
1	10	10	21.3	20.8
1	14	10	8.2	4.7
1	16	10	6.4	7.6
3	0	10	32.2	33.0
3	2	10	14.0	15.1
3	4	10	29.9	29.8
3	6	10	14.0	11.6
3	10	10	14.6	16.0
5	0	10	14.7	13.1
5	2	10	24.2	23.9
5	4	10	12.5	9.2
5	8	10	19.4	17.4
5	10	10	16.5	13.0
7	0	10	24.9	31.6
7	2	10	11.4	11.7
9	2	10	27.4	25.1
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9	6	10	13.9	12.4
11	2	10	17.6	11.8
11	4	10	16.1	16.1
11	6	10	9.5	9.4
11	8	10	12.6	13.0
15	0	10	7.8	8.0
0	1	11	10.6	10.8
0	3	11	25.7	27.9
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0	11	11	11.5	10.6
2	1	11	12.8	12.5
2	3	11	16.4	17.0
2	7	11	10.2	9.4
2	13	11	8.9	6.9
4	1	11	17.8	17.3
4	5	11	20.2	23.6
4	11	11	26.0	22.6
6	3	11	24.2	24.3
8	1	11	15.9	9.2
10	1	11	9.9	9.9
10	7	11	14.7	12.0
10	13	11	4.2	7.1
12	1	11	10.8	11.6
12	7	11	8.9	9.2
1	2	12	17.4	22.7
1	4	12	23.2	24.7
1	6	12	13.5	13.3
1	8	12	16.4	12.8
1	10	12	17.8	15.1
3	0	12	23.1	21.6
3	2	12	17.9	17.8
3	4	12	23.5	22.0
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3	8	12	14.5	10.6
3	10	12	11.1	6.7
5	0	12	16.0	15.0
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5	4	12	14.5	15.0
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5	8	12	16.3	13.1
7	0	12	12.5	13.6
7	2	12	11.5	12.2
7	4	12	18.9	14.0
9	2	12	13.6	15.0
9	4	12	9.5	11.2
9	8	12	11.8	13.4
11	2	12	9.9	5.4
11	8	12	7.2	8.7
13	0	12	13.3	11.1
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2	1	13	16.7	15.4
2	5	13	14.7	13.7
4	5	13	13.9	9.7
6	1	13	17.0	17.0
6	3	13	13.1	11.4
8	1	13	11.0	10.3
10	1	13	8.0	11.8
10	5	13	7.8	7.4
10	7	13	6.9	5.7
3	0	14	13.1	9.2
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3	4	14	11.9	10.3
3	6	14	13.7	10.3
5	2	14	14.9	16.5
9	0	14	7.2	8.3
9	2	14	9.3	10.5
9	4	14	11.3	12.8
11	0	14	12.1	12.3
11	2	14	7.1	7.8
11	4	14	7.6	9.2
0	7	15	7.2	8.1
1	0	16	9.7	8.6
7	4	16	10.6	13.8
0	7	17	7.1	7.8
2	1	0	63.8	56.8
2	3	0	191.3	200.1
2	5	0	17.1	19.6
2	7	0	32.8	28.4
2	9	0	52.8	62.1
2	11	0	12.8	8.4
2	13	0	10.0	8.5
2	17	0	16.3	12.5
4	1	0	74.7	89.7
4	3	0	15.3	18.0
4	5	0	121.3	138.6
4	9	0	33.8	31.8
4	11	0	35.1	30.1
4	15	0	10.3	9.1
4	19	0	9.7	8.1
6	1	0	31.8	29.1
6	3	0	16.4	16.3
6	5	0	31.7	28.1
6	9	0	26.1	25.7
6	13	0	21.4	22.8
8	1	0	25.8	23.4
8	3	0	20.1	22.1
8	5	0	43.2	39.8
8	9	0	18.7	20.2
8	11	0	47.5	51.2
8	13	0	12.2	14.7
10	1	0	12.9	10.4
10	3	0	47.8	53.2
10	5	0	13.9	16.5
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10	9	0	19.9	18.8
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10	13	0	12.8	12.7
10	17	0	9.2	9.4
12	1	0	23.3	21.0
12	3	0	14.4	19.4
12	5	0	10.7	11.0
12	7	0	9.9	8.8
12	9	0	10.7	9.7
14	1	0	27.5	24.7
14	5	0	13.1	14.7
16	1	0	7.9	7.9
16	5	0	11.0	10.6
1	6	1	118.3	128.9
1	8	1	84.6	88.8
1	10	1	42.4	40.9
1	12	1	38.2	34.2
1	14	1	26.7	27.0
1	20	1	7.5	8.4
3	0	1	117.4	116.9
3	2	1	84.2	82.6
3	4	1	68.2	67.5
3	6	1	108.6	94.8
3	8	1	34.0	35.4
3	10	1	57.1	52.9
3	12	1	23.8	20.5
5	0	1	43.6	34.9
5	2	1	131.0	136.6
5	4	1	77.2	80.5
5	6	1	28.5	27.7
5	8	1	72.1	67.2
5	10	1	34.6	26.8
5	16	1	18.1	16.8
7	0	1	58.5	55.1
7	2	1	79.2	71.1
7	4	1	59.4	59.8
7	6	1	54.3	55.6
7	8	1	19.4	20.1
9	0	1	20.9	17.1
9	2	1	53.9	55.6
9	4	1	35.1	41.1
9	6	1	10.4	11.5
9	8	1	17.1	18.5
9	12	1	13.9	17.2
11	0	1	42.1	46.8
11	2	1	12.2	11.0
11	4	1	12.8	11.5
11	6	1	34.9	33.4
11	8	1	16.1	13.9
13	0	1	19.7	19.9
13	2	1	14.2	15.5
13	4	1	12.5	12.9
15	2	1	11.0	10.5
0	1	2	7.4	2.2
0	3	2	71.9	79.7
0	5	2	50.2	57.0
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0	11	2	68.8	72.6
0	13	2	16.4	16.5
0	15	2	18.9	14.4
0	17	2	20.3	17.4
2	1	2	125.5	102.5
2	3	2	55.7	42.5
2	5	2	59.3	56.0
2	7	2	49.2	56.5
2	9	2	28.6	33.7
2	11	2	9.1	7.7
2	13	2	41.4	43.2
4	1	2	47.6	31.0
4	3	2	43.9	42.3
4	5	2	43.8	32.1
4	7	2	63.8	61.8
4	9	2	40.0	37.0
4	11	2	26.8	23.9
6	1	2	71.4	71.1
6	3	2	29.0	27.2
6	5	2	68.8	68.2
6	7	2	44.1	48.2
6	9	2	27.1	23.6
6	11	2	14.4	15.4
6	13	2	26.2	29.1
8	1	2	33.5	32.3
8	3	2	38.2	41.1
8	5	2	36.5	44.0
8	9	2	26.8	23.0
8	11	2	35.8	39.3
8	17	2	8.9	12.2
10	1	2	20.0	17.6
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10	7	2	19.9	18.9
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1	12	3	29.7	31.2
1	14	3	21.1	19.2
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3	4	3	104.3	87.3
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3	8	3	61.2	69.5
3	10	3	57.1	54.0
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3	14	3	36.9	33.9
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5	12	3	34.0	37.0
7	0	3	17.0	12.7
7	2	3	23.2	21.3
7	4	3	40.3	40.3
7	6	3	14.2	14.6
7	10	3	26.2	31.2
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9	10	3	13.6	12.9
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11	2	3	8.0	8.7
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11	12	3	13.5	9.6
13	4	3	10.2	10.7
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0	11	4	7.8	4.4
0	13	4	22.1	22.1
0	15	4	16.7	18.8
0	17	4	7.5	5.3
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2	3	4	127.7	129.8
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2	7	4	33.6	32.9
2	9	4	54.6	53.8
2	11	4	18.9	15.5
2	15	4	10.2	11.0
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4	3	4	13.5	11.0
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4	7	4	63.6	61.2
4	9	4	15.3	11.0
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4	13	4	25.5	28.7
4	15	4	14.4	16.8
6	1	4	17.4	13.7
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6	7	4	16.8	18.3
6	9	4	30.0	33.2
6	11	4	14.6	14.4
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8	5	4	16.4	15.9
8	7	4	20.6	17.6
8	9	4	11.4	11.2
10	1	4	22.4	22.0
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10	5	4	16.5	18.7
10	9	4	24.3	23.3
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12	1	4	9.6	4.1
12	3	4	7.8	5.7
12	7	4	25.8	24.0
12	9	4	10.8	13.8
14	3	4	6.3	8.4
16	1	4	12.9	8.3
1	0	5	88.9	78.1
1	2	5	95.3	102.7
1	4	5	78.2	77.6
1	6	5	52.6	48.2
1	8	5	25.0	21.1
1	10	5	32.8	31.1
1	12	5	16.0	13.0
1	16	5	12.5	14.2
3	0	5	11.8	25.1
3	2	5	41.5	42.6
3	4	5	42.9	33.9
3	6	5	25.7	25.6
3	8	5	13.7	10.1
3	10	5	15.3	16.3
3	12	5	31.1	33.9
3	14	5	14.5	14.8
5	0	5	60.7	53.0
5	2	5	61.6	67.0
5	4	5	49.8	51.6
5	6	5	16.3	16.5
5	8	5	28.3	28.6
5	10	5	15.7	12.3
5	12	5	28.9	24.3
7	0	5	94.9	94.4
7	2	5	35.3	34.4
7	4	5	46.4	46.4
7	6	5	42.1	40.4
7	8	5	35.4	32.5
7	10	5	19.2	22.1
9	0	5	20.3	15.2
9	2	5	19.9	20.8
9	4	5	32.8	34.7
9	6	5	17.1	13.4
11	2	5	26.6	27.3
11	4	5	24.6	25.4
11	6	5	13.2	13.2
11	8	5	17.0	17.2
11	10	5	9.7	8.1
13	2	5	16.3	15.6
13	4	5	17.5	18.1
0	1	6	46.3	49.1
0	3	6	17.9	27.1
0	5	6	80.6	80.6
0	11	6	21.4	19.3
0	13	6	14.6	17.4
0	15	6	16.3	16.1
0	17	6	15.3	15.1
2	1	6	66.6	72.5
2	3	6	48.7	46.3
2	5	6	73.6	81.9
2	7	6	16.3	17.4
2	9	6	27.2	23.1
2	11	6	37.1	37.7
2	13	6	20.6	17.5
2	19	6	5.3	7.7
4	1	6	77.2	65.7
4	3	6	79.9	76.0
4	5	6	12.9	13.1
4	7	6	45.3	41.0
4	9	6	28.3	26.6
4	11	6	36.8	27.7
4	15	6	9.7	8.8
6	1	6	26.2	15.5
6	3	6	35.0	31.7
6	5	6	12.5	8.0
6	7	6	22.7	18.4
6	9	6	26.7	27.1
8	1	6	27.7	30.3
8	3	6	56.0	54.9
8	5	6	25.7	28.8
8	7	6	15.7	18.4
8	9	6	16.8	17.1
8	11	6	24.3	22.7
10	1	6	23.2	25.0
10	5	6	15.2	14.1
10	7	6	22.2	23.8
10	11	6	9.1	8.9
12	3	6	15.0	17.6
14	1	6	13.5	11.4
1	0	7	13.6	12.8
1	2	7	22.2	16.5
1	4	7	26.5	27.4
1	6	7	18.5	19.1
1	8	7	24.9	21.5
1	12	7	21.1	20.7
1	16	7	11.5	13.1
3	0	7	66.2	64.1
3	2	7	34.5	33.3
3	4	7	44.6	44.5
3	6	7	46.5	43.6
3	8	7	35.3	39.8
3	10	7	22.9	18.9
3	12	7	23.9	18.0
3	14	7	23.1	23.2
5	0	7	89.6	93.1
5	2	7	11.8	8.3
5	4	7	15.7	13.8
5	6	7	44.2	45.5
5	8	7	26.7	24.9
7	2	7	16.5	15.1
7	4	7	11.1	12.3
7	6	7	21.7	21.1
9	0	7	29.0	24.7
9	2	7	21.5	23.6
9	4	7	12.4	15.6
9	6	7	26.5	26.3
9	8	7	13.9	15.3
9	12	7	13.1	16.2
11	0	7	15.4	13.0
11	2	7	31.2	29.7
11	6	7	13.9	13.1
11	10	7	9.5	8.4
17	0	7	10.7	9.7
0	1	8	34.2	36.0
0	3	8	20.3	25.6
0	7	8	31.5	30.3
0	11	8	31.5	30.5
0	13	8	32.3	31.9
2	1	8	73.5	69.6
2	3	8	40.3	42.5
2	5	8	43.6	44.5
2	9	8	19.6	19.3
2	15	8	18.2	16.3
4	1	8	53.1	46.2
4	5	8	25.0	20.3
4	7	8	37.9	32.6
4	9	8	10.2	10.2
4	11	8	10.4	9.5
6	1	8	12.8	10.5
6	3	8	38.2	34.9
6	5	8	26.2	26.6
6	11	8	21.3	27.4
8	1	8	18.1	13.2
8	3	8	19.6	15.1
8	7	8	31.7	28.6
8	13	8	10.6	10.0
10	1	8	9.9	10.6
10	3	8	10.0	8.6
10	9	8	15.9	18.0
12	1	8	14.5	12.5
12	7	8	15.0	12.9
12	13	8	5.8	8.5
1	0	9	27.1	23.5
1	2	9	42.6	45.4
1	4	9	38.2	40.3
1	8	9	31.4	32.1
1	10	9	28.3	28.0
1	12	9	18.9	18.4
1	14	9	8.6	10.0
3	0	9	8.0	4.9
3	2	9	27.4	25.7
3	4	9	8.9	8.2
3	6	9	18.9	17.3
3	8	9	18.2	18.7
3	10	9	11.9	11.6
3	12	9	16.5	14.7
5	0	9	13.2	14.5
5	2	9	31.0	30.6
5	4	9	29.0	28.0
5	8	9	26.8	25.2
7	0	9	40.6	43.9
7	2	9	30.7	30.3
7	4	9	20.3	25.0
7	6	9	38.5	43.2
7	8	9	19.7	19.2
9	0	9	17.1	10.4
9	2	9	19.4	10.0
9	4	9	14.5	14.3
9	6	9	13.9	10.8
9	8	9	13.9	10.7
11	0	9	10.7	11.1
13	4	9	8.9	12.2
15	0	9	18.2	15.8
0	1	10	21.3	19.9
0	3	10	17.8	11.1
0	5	10	11.5	13.3
0	7	10	16.3	14.9
0	9	10	25.3	25.4
0	11	10	14.5	12.4
2	1	10	54.0	54.1
2	3	10	28.5	31.8
2	5	10	49.0	46.6
2	7	10	28.2	29.5
2	11	10	27.8	22.7
4	1	10	9.3	10.8
4	3	10	57.4	53.7
4	5	10	23.3	21.2
4	9	10	31.4	26.7
4	11	10	14.4	10.6
6	1	10	14.7	15.8
6	3	10	13.5	11.8
6	5	10	15.0	17.2
8	1	10	27.9	26.5
8	3	10	25.5	20.8
8	5	10	20.0	19.2
8	9	10	11.8	13.0
10	1	10	28.1	28.1
10	3	10	9.3	10.4
10	5	10	18.5	18.2
10	7	10	15.6	15.6
10	9	10	8.5	10.3
12	1	10	10.0	8.6
12	5	10	9.3	8.4
1	2	11	41.8	43.7
1	12	11	8.9	8.9
1	16	11	7.5	8.9
3	0	11	34.9	33.3
3	2	11	19.3	16.8
3	6	11	23.6	24.6
3	8	11	17.0	14.6
3	10	11	11.7	7.9
3	12	11	10.8	10.8
5	0	11	15.9	17.8
5	2	11	17.1	21.5
5	4	11	13.9	11.0
5	6	11	21.5	19.3
5	8	11	19.3	18.7
7	2	11	14.2	17.0
7	4	11	17.8	17.4
11	0	11	14.5	15.7
11	2	11	17.5	16.8
11	4	11	13.9	14.9
11	6	11	9.0	8.6
11	8	11	6.2	9.8
11	10	11	6.8	6.7
13	0	11	16.7	14.0
0	1	12	23.9	23.9
0	3	12	14.7	12.4
0	7	12	30.6	27.1
0	13	12	14.7	14.0
2	1	12	21.5	19.6
2	3	12	29.2	26.3
2	5	12	17.4	17.8
2	9	12	10.0	12.0
4	5	12	10.8	7.1
4	7	12	10.4	9.2
4	9	12	10.0	8.3
6	1	12	25.8	26.5
6	3	12	13.1	14.5
6	5	12	17.8	18.5
8	1	12	16.3	16.5
8	5	12	12.6	12.5
8	7	12	11.4	11.6
12	1	12	6.9	7.8
14	3	12	6.1	9.6
1	2	13	11.5	7.9
1	4	13	16.3	17.1
1	8	13	9.1	10.2
1	10	13	11.9	12.9
3	0	13	13.7	16.4
3	4	13	15.0	13.1
3	6	13	11.8	11.5
3	8	13	17.9	15.6
7	0	13	17.1	19.2
7	2	13	11.3	12.1
9	0	13	9.7	4.7
9	2	13	15.7	15.2
9	4	13	11.7	13.5
9	8	13	10.6	13.7
0	3	14	11.4	12.5
0	9	14	14.5	13.8
2	1	14	10.6	10.3
2	5	14	10.3	11.6
2	7	14	13.5	10.7
2	11	14	10.6	11.7
4	1	14	8.9	9.6
4	3	14	20.7	22.2
4	5	14	14.0	14.6
4	9	14	11.9	11.7
6	1	14	11.9	11.5
10	1	14	13.3	12.8
10	7	14	11.1	11.3
12	3	14	6.1	9.7
1	0	15	12.4	11.7
1	2	15	9.1	8.2
1	4	15	12.5	11.7
1	6	15	11.9	11.3
1	8	15	7.8	10.0
5	0	15	11.3	10.5
5	4	15	10.8	5.3
0	1	16	7.5	8.7
0	5	16	6.9	5.1
4	5	16	11.3	10.3
8	1	16	6.9	8.5

Discussion of the structure

The molecule consists of two substituted quinolizidine systems joined together through a *spiro*-thiophane ring. Each quinolizidine moiety is built of two *trans*-fused chairs. The methyl and furan substituents are in the more stable equatorial orientations. The thiophane ring is puckered with C(18) lying outside the plane formed by the other four atoms. C(33) is equatorial with respect to both quinolizidine systems while C(34) and the sulphur are axial. Some calculated mean planes and their relationships to one another are given in Table 4.

The intramolecular bond lengths and their e.s.d.'s are listed in Table 5. All coordinate e.s.d.'s had been increased by 30% to allow for the block-diagonal approximation. The most unusual bond lengths are the three in the thiophane ring involving C(18), C(34), S and C(8). The bond length of 1·70 Å between C(18) and C(34) is significantly longer than the accepted value for a C–C single bond. The difference between this bond length and the normally accepted one amounts to 5σ, which puts it in the 'highly significant' range as defined by Cruickshank & Robertson (1953). It is postulated that this is due to a partial disorder of the molecule. The molecule very nearly possesses a noncrystallographic twofold axis going through C(33) and bisecting the thiophane ring. It is therefore capable of occupying two distinct positions. This is supported by the bond lengths S–C(8) and S–C(34) which are shorter than the accepted value of 1·82 Å (Abrahams, 1956), the latter bond being significantly shorter. A final difference-Fourier map showed a peak whose height was 1·1 e.Å^{-3} and whose distance from C(18) was 1·79 Å. The position of this peak with respect to C(34) and its distance from that atom (0·49 Å) indicates that the peak cannot correspond to a hydrogen atom. Therefore it must be due to the sulphur atom which is placed in that position by a rotation of the molecule. A disorder of about 10% would be consistent with these results. Finally, the thermal vibration parameters of atoms C(34) and S, shown in Table 2, exhibit a much higher anisotropy than the other three atoms of this ring and their direction cosines agree with what one would expect to obtain in view of the postulated disorder.

Table 5. *Intramolecular bond lengths and angles*

Standard deviations* in parentheses are for the last place listed.

Distances		Angles	
C(1)—C(2)	1·54 (3) Å	C(2)—C(1)—N(6)	110 (1)°
C(1)—N(6)	1·49 (2)	C(2)—C(1)—C(10)	113 (2)
C(1)—C(10)	1·53 (3)	N(6)—C(1)—C(10)	110 (2)
C(2)—C(3)	1·52 (3)	C(1)—C(2)—C(3)	112 (2)
C(2)—C(21)	1·59 (3)	C(1)—C(2)—C(21)	110 (2)
C(3)—C(4)	1·52 (3)	C(3)—C(2)—C(21)	110 (2)
C(4)—C(5)	1·55 (3)	C(2)—C(3)—C(4)	110 (2)
C(5)—N(6)	1·52 (2)	C(3)—C(4)—C(5)	111 (2)
C(5)—C(22)	1·46 (3)	C(4)—C(5)—C(6)	108 (2)
N(6)—C(7)	1·52 (2)	C(4)—C(5)—C(22)	110 (2)
C(7)—C(8)	1·52 (3)	N(6)—C(5)—C(22)	112 (2)
C(8)—C(9)	1·51 (3)	C(1)—N(6)—C(5)	112 (1)
C(8)—C(33)	1·53 (3)	C(1)—N(6)—C(7)	111 (1)
C(8)—S	1·76 (2)	C(5)—N(6)—C(7)	109 (1)
C(9)—C(10)	1·54 (4)	N(6)—C(7)—C(8)	115 (2)
C(11)–C(12)	1·59 (3)	C(7)—C(8)—C(9)	107 (2)
C(11)–N(16)	1·49 (3)	C(7)—C(8)—C(33)	104 (2)
C(11)–C(20)	1·49 (3)	C(7)—C(8)—S	115 (1)
C(12)–C(13)	1·50 (4)	C(9)—C(8)—C(33)	112 (2)
C(12)–C(27)	1·65 (4)	C(9)—C(8)—S	111 (2)
C(13)–C(14)	1·48 (4)	C(33)–C(8)—S	108 (1)
C(14)–C(15)	1·44 (3)	C(8)—C(9)—C(10)	111 (2)
C(15)–N(16)	1·57 (3)	C(1)—C(10)–C(9)	109 (2)
C(15)–C(28)	1·50 (3)	C(12)–C(11)–N(16)	106 (2)
N(16)–C(17)	1·55 (2)	C(12)–C(11)–C(20)	112 (2)
C(17)–C(18)	1·49 (3)	N(16)–C(11)–C(20)	109 (2)
C(18)–C(19)	1·56 (3)	C(11)–C(12)–C(13)	108 (2)
C(18)–C(33)	1·58 (3)	C(11)–C(12)–C(27)	106 (2)
C(18)–C(34)	1·70 (3)	C(13)–C(12)–C(27)	109 (2)
C(19)–C(20)	1·52 (3)	C(12)–C(13)–C(14)	107 (2)
C(22)–C(23)	1·42 (3)	C(13)–C(14)–C(15)	112 (2)
C(22)–C(26)	1·41 (4)	C(14)–C(15)–N(16)	109 (2)
C(23)–C(24)	1·20 (5)	C(14)–C(15)–C(28)	112 (2)
C(24)–O(25)	1·44 (4)	N(16)–C(15)–C(28)	110 (2)
O(25)–C(26)	1·34 (4)	C(11)–N(16)–C(15)	110 (1)
C(28)–C(29)	1·36 (3)	C(11)–N(16)–C(17)	112 (1)
C(28)–C(32)	1·48 (5)	C(15)–N(16)–C(17)	109 (1)
C(29)–O(30)	1·31 (4)	N(16)–C(17)–C(18)	112 (1)
O(30)–C(31)	1·41 (4)	C(17)–C(18)–C(19)	111 (2)
C(31)–C(32)	1·26 (5)	C(17)–C(18)–C(33)	109 (2)
C(34)–S	1·69 (2)	C(17)–C(18)–C(34)	108 (2)

Table 4. *Mean planes*

Plane *A*		Plane *B*		Plane *C*		Plane *D*		Plane *E*	
	Δ		*Δ*		*Δ*		*Δ*		*Δ*
C(1)	−0·036 Å	C(11)	−0·057 Å	C(22)	−0·026 Å	C(28)	0·038 Å	C(8)	0·008 Å
C(3)	−0·003	C(13)	0·043	C(23)	0·036	C(29)	−0·010	C(33)	−0·005
C(4)	0·032	C(14)	0·020	C(24)	−0·029	O(30)	−0·025	C(34)	0·005
N(6)	−0·023	N(16)	−0·068	O(25)	0·008	C(31)	0·055	S	−0·007
C(8)	−0·011	C(18)	0·046	C(26)*	0·012	C(32)	−0·058	C(18)*	0·487
C(9)	0·040	C(19)	0·016	C(5)*	−0·064	C(15)*	0·102		

* Not included in the calculation of the plane.

Plane *A* $0{\cdot}8641X+0{\cdot}1697Y-0{\cdot}4739Z-5{\cdot}7699=0$
Plane *B* $0{\cdot}4070X+0{\cdot}1572Y-0{\cdot}8998Z-3{\cdot}1688=0$
Plane *C* $0{\cdot}0446X-0{\cdot}9910Y-0{\cdot}1258Z+9{\cdot}7594=0$
Plane *D* $0{\cdot}2087X+0{\cdot}8343Y+0{\cdot}5103Z-0{\cdot}7021=0$
Plane *E* $0{\cdot}4995X+0{\cdot}4761Y+0{\cdot}7237Z-5{\cdot}0649=0$

∠*AB*	∠*AC*	∠*AD*	∠*AE*	∠*BC*	∠*BD*	∠*BE*	∠*CD*	∠*CE*	∠*DE*
36·4°	86·0°	85·4°	80·2°	88·6°	75·9°	68·1°	28·1°	57·3°	29·5°

Table 5 (*cont.*)

Angles	
C(19)–C(18)–C(33)	108 (2)
C(19)–C(18)–C(34)	114 (2)
C(33)–C(18)–C(34)	106 (1)
C(18)–C(19)–C(20)	109 (2)
C(11)–C(20)–C(19)	109 (2)
C(5)—C(22)–C(23)	128 (2)
C(5)—C(22)–C(26)	129 (2)
C(23)–C(22)–C(26)	103 (2)
C(22)–C(23)–C(24)	112 (3)
C(23)–C(24)–O(25)	111 (3)
C(24)–O(25)–C(26)	104 (2)
C(22)–C(26)–O(25)	110 (3)
C(15)–C(28)–C(29)	130 (2)
C(15)–C(28)–C(32)	128 (2)
C(29)–C(28)–C(32)	101 (2)
C(28)–C(29)–O(30)	114 (3)
C(29)–O(30)–C(31)	105 (2)
O(30)–C(31)–C(32)	111 (3)
C(28)–C(32)–C(31)	108 (3)
C(8)—C(33)–C(18)	110 (2)
C(18)–C(34)–S	105 (1)
C(8)—S——C(34)	102 (1)

* All standard deviations were increased by 30% in order to allow for block diagonal approximation.

The determination of the bond lengths in the two furan rings was adversely affected by the relatively high motion of these rings. This is probably a consequence of the molecular disorder as well as some rotation around the single bonds by which these rings are attached. Nevertheless, the values obtained do not deviate significantly from previously reported ones.

The C–O bond was found to be 1·362 Å in furan (Bak, Christensen, Dixon, Hansen-Nygaard, Andersen & Schottländer, 1962) and 1·361 Å in furan-3,4-dicarboxylic acid (Williams & Rundle, 1964). In the same investigations the C=C bonds were determined to be 1·361 and 1·351 Å respectively. Our results agree with these values and even the very short bond length between C(23) and C(24) is not significantly different. The lengths of the two C(sp^2)–C(sp^2) single bonds are in good agreement with those found by Bak *et al.* (1·431 Å) and by Williams & Rundle (1·462 Å). The two C(sp^2)–C(sp^3) single bonds by which the furan rings are attached to the quinolizidines agree well in their lengths with the accepted value for such bonds of 1·501 Å (Lide, 1962).

There are six C(sp^3)–N$^+$ bonds in the molecule, each protonated nitrogen atom being attached to three C(sp^3) atoms, and the weighted mean of their lengths is 1·515 (σ=0·009) Å. This is in good agreement with the weighted mean of 1·499 (σ=0·002) Å calculated from 30 such bonds which have been found in the following structures: ibogaine HBr (Arai, Coppola & Jeffrey, 1960), alphaprodine HCl (Kartha, Ahmed & Barnes, 1960), methadone HBr (Hanson & Ahmed, 1958), des(oxymethylene)lycoctonine (Przybylska, 1961*a*), demethanol-aconinone (Przybylska, 1961*b*), betaprodine HBr (Ahmed, Barnes & Masironi, 1963), betaprodine HCl (Ahmed & Barnes, 1963), hetisine HBr (Pzrybylska, 1963), heteratisine HBr (Przybylska, 1965) and mitragynine HI (Zacharias, Rosenstein & Jeffrey, 1965). There appears to be sufficient evidence now for accepting 1·500 Å as the usual length for such a bond.

The average distance of the 20 C(sp^3)–C(sp^3) single bonds is 1·53 Å, which is in good agreement with the accepted value. Apart from the discrepancy in the thiophane ring, which is due to the molecular disorder (*vide supra*), none of the bonds differs significantly from this mean value.

The valence angles and their e.s.d.'s, the latter increased by 30% as explained above, are also shown in Table 5. The average of 48 angles which should be tetrahedral equals 109·8°. Those angles whose values deviate significantly from the theoretical one are in the vicinity of the thiophane ring and the deviations can be ascribed to the disorder. Thus, for example, the angle at C(18)–C(34)–S is 105°, 4·3σ less than a tetrahedral angle, and this is a consequence of the increased bond length C(18)–C(34) caused by the disorder. The C–S–C angle of 102 ± 1° has the expected value for sulphur linked to two atoms (Abrahams, 1956). All angles in the furan rings agree with those found previously in similar systems (Bak *et al.*, 1962; Williams & Rundle, 1964).

An interesting feature of this crystal structure is the extensive network of hydrogen bonds composed of four molecules of water, two bromide ions and two quaternary nitrogen atoms. The hydrogen positions indicated in Fig. 2 are given only to show the proton donor–proton acceptor relationship of the various atoms participating in hydrogen bonding. The assignment is unambiguous when one considers that HBr, being a strong acid, must be completely dissociated and that the amine nitrogen atoms must be protonated in preference to water. Although the positions of hydrogen atoms were not determined, the presence of hydrogen bonds can be inferred from the distances and angles shown in

Table 6. *Distances and angles involving hydrogen bonds*

′ $-x,\ \frac{1}{2}+y,\ \frac{1}{2}-z$
″ $x,\ 1+y,\ z$
‴ $-x,\ -\frac{1}{2}+y,\ \frac{1}{2}-z$
iv $x,\ -1+y,\ z$

Distances		Angles	
Br(1)–W(1)	3·24 Å	W(1) —Br(1)–N(16)	137°
Br(1)–W(2)	3·42	W(1) —Br(1)–W(2)	104
Br(1)–N(16)	3·37	N(16)—Br(1)–W(2)	117
Br(2)–$W(1'')$	3·26	$W(1'')$ –Br(2)–$W(2')$	74
Br(2)–$W(2')$	3·32	$W(1'')$ –Br(2)–W(4)	162
Br(2)–W(4)	3·35	$W(2')$—Br(2)–W(4)	121
W(1)–$W(3''')$	2·68	$W(3''')$–W(1)–Br(1)	134
W(2)–W(3)	2·73	$W(3''')$–W(1)–Br(2^{iv})	97
W(3)–W(4)	2·80	Br(1)—W(1)–Br(2^{iv})	125
W(4)–N(6)	2·90	W(3)—W(2)–Br($2'''$)	94
		W(3)—W(2)–Br(1)	142
Angles		Br(1)—W(2)–Br($2'''$)	119
C(1)—N(6)–W(4)	106°	$W(1')$—W(3)–W(2)	94
C(5)—N(6)–W(4)	104	$W(1')$—W(3)–W(4)	127
C(7)—N(6)–W(4)	116	W(2)—W(3)–W(4)	137
C(11)–N(16)–Br(1)	105	W(3)—W(4)–N(6)	132
C(15)–N(16)–Br(1)	105	W(3)—W(4)–Br(2)	111
C(17)–N(16)–Br(1)	116	N(6)—W(4)–Br(2)	112

Table 6. The e.s.d.'s of the distances range from 0·02 to 0·03 Å, and those of the angles from 0·5 to 1·0°. The distances between water oxygen atoms do not deviate significantly from the corresponding value, 2·76 Å, found in ice. All distances between water oxygen atoms and bromide ions are close to the weighted average of 3·34 Å obtained by Clark (1963) in a review of hydrated compounds. Similarly, the distance between the quaternary nitrogen and the water oxygen agrees with the average of 2·94 Å found by Clark. The $N^+–H\cdots Br^-$ distance is, as it should be, somewhat shorter than the value of 3·40 Å found in heteratisine $HBr.H_2O$ (Przybylska, 1965) for a bifurcated hydrogen bond and somewhat longer than the average $H–O–H\cdots Br^-$ distance.

The environment of all water molecules is planar and approximately trigonal, *i.e.* each oxygen atom is surrounded by three heavy-atom (non-hydrogen) neighbours. In each case the sum of the three angles approaches 360°. The environment of the bromide ions is also planar. Such planar arrangements of hydrogen bonds, although less common than tetrahedral, have been reported in the past (Clark, 1963). The angles $X\cdots H–O–H\cdots X$ (where X can be Br^- or another water oxygen atom) vary from 93·7 to 125·4°, very reasonable values compared with the range of angles reported by Clark. Each bromide ion is surrounded by three protons donated by its three neighbours.

The crystal structure can be thus described as consisting of sheets of water molecules and bromide ions to which the alkaloid molecules are attached *via* their quaternary nitrogen atoms. The sheets are formed by two chains running parallel to the y axis and the chains which are related by the screw axes are cross-linked in three positions within each unit cell. The sheets are approximately parallel to (304) and their thickness, as determined by a mean-plane calculation, is 1 Å.

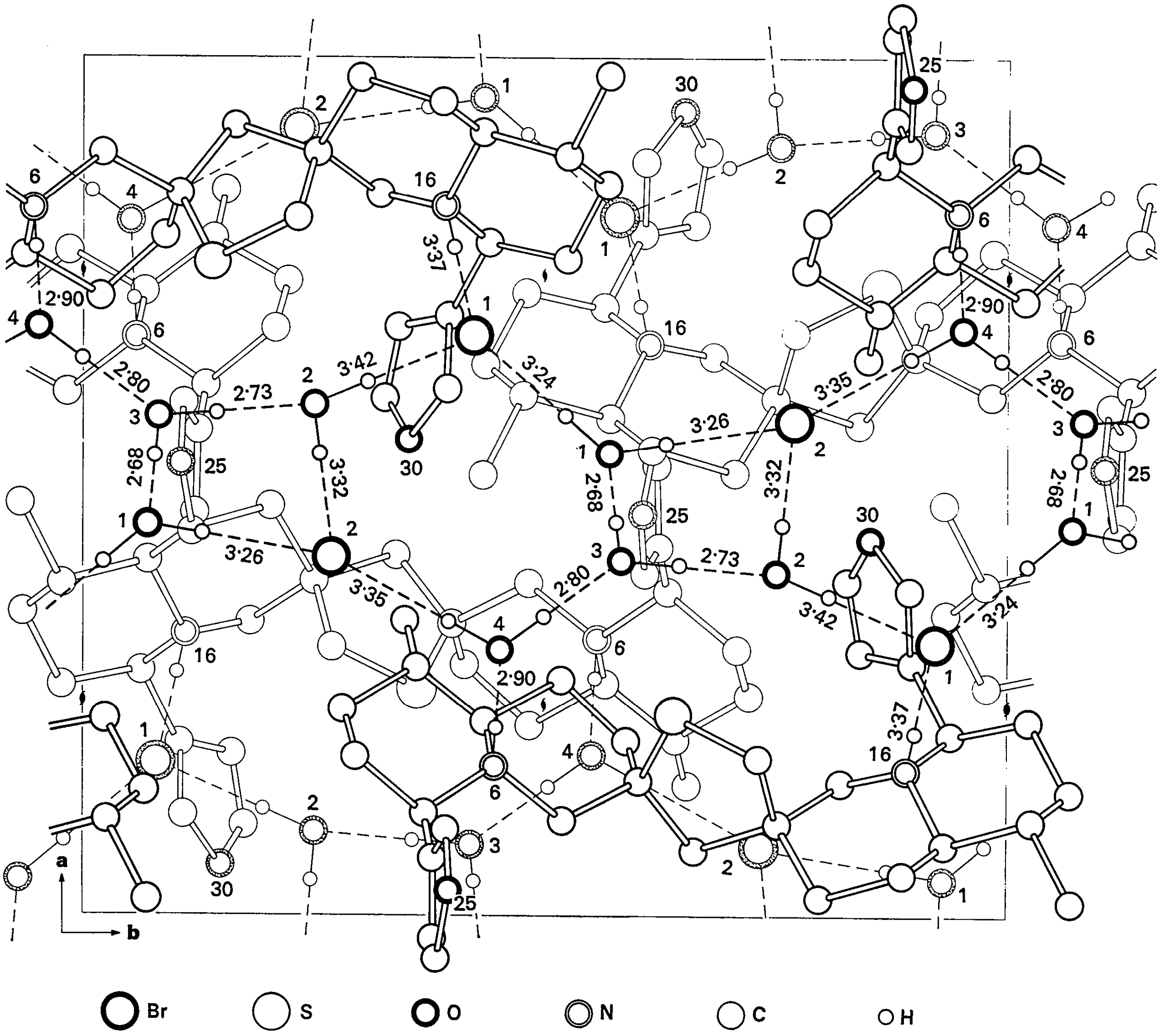

Fig. 2. Projection of the structure along the c axis. Hydrogen atoms are in assigned positions; hydrogen bonds are shown by broken lines. The distances are between non-hydrogen atoms.

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Instrumental Corrections for X-ray Reflexions from Thin Monocrystalline Films

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Correction curves for instrumental broadening are given for the cases that the physical broadening profile is $(\sin^2 Kx)/(Kx)^2$ and the instrumental profile is either $(1+\kappa^2x^2)^{-1}$ or $(1+\kappa^2x^2)^{-2}$ or $\exp\{-\kappa^2x^2\}$.

The interpretation of recent diffractometer measurements of thin monocrystals requires the observed reflexions to be corrected for instrumental influences. Only then is it possible to compare the thickness calculated from the breadth of the reflexions with that determined from fringe spacings, electron microscopy, gravimetry, optical interferrometry *etc.* It is customary and convenient to obtain the instrumental broadening from a standard specimen which is known to be free from physical broadening. For thin films there is apparently the complication that the standard specimen has to be thicker in order to be free from size broadening. The penetration of the X-ray beam into the specimen then causes an increase of the instrumental broadening for the thicker film. However, the total width of the aberration profile due to penetration is in most cases so small that it and its changes with specimen thickness can be neglected. An estimate can be obtained by assuming that the crystal reflects as a delta function. Calculating the penetration into the specimen