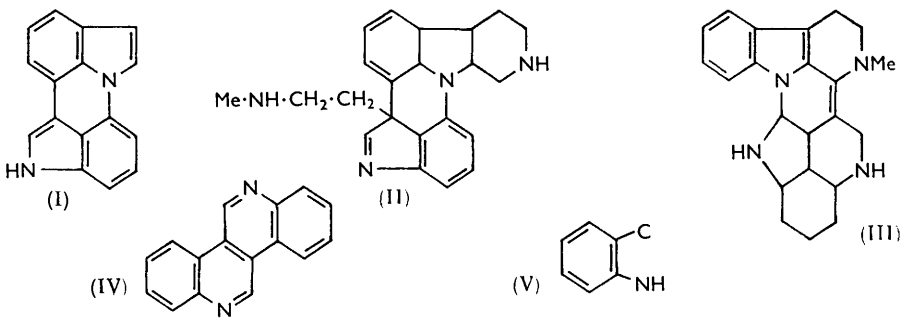
38. *The Structure of Calycanthine. X-Ray Analysis of the Dihydrobromide Dihydrate.*

By T. A. Hamor and J. Monteath Robertson.

The structure and stereochemistry of the alkaloid calycanthine, $C_{22}H_{26}N_4$, has been determined, apart from absolute configuration, by *X*-ray analysis of the dihydrobromide dihydrate, $C_{22}H_{28}N_4Br_2,2H_2O$. The crystals are orthorhombic, space group $P2_12_12_1$ with four molecules per unit cell. The results are summarised in formula (IX) for calycanthine and in Figs. 1—3 which show projections of the molecule. In the final results the discrepancy *R* over the 2116 observed structure factors is 9·6%. The bond lengths and angles (Figs. 4 and 5) have standard deviations of about 0·03 Å and 1·7°.

As long ago as 1888 interest had been aroused in the shrub *Calycanthus glaucus* (Willd.), native to Georgia, North Carolina, and Tennessee, owing to the poisonous nature of its seeds. In that year Eccles [1] isolated the principal poisonous constituent of the seeds which he named calycanthine, and in 1905 Gordin [2] assigned to it the formula $C_{11}H_{14}N_2$ and prepared a number of crystalline salts including the hydrochloride, hydrobromide, and hydriodide. In 1925 Späth and Stroh [3] doubled the molecular formula and this was revised in 1939 by Barger, Madinaveitia, and Streuli [4] to $C_{22}H_{26}N_4$, the formula accepted

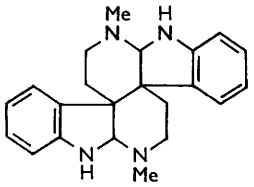


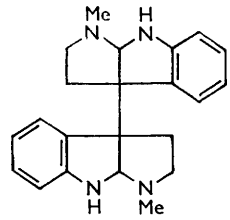
at present and confirmed by the present analysis. Barger *et al.* found that on degradation of calycanthine by a variety of reactions such as heating with zinc dust, copper oxide, selenium, or sulphur, or by treatment with chromic acid, a stable, weakly basic substance, calycanine, $C_{16}H_{10}N_2$, was obtained. Pyrolysis of calycanthine with soda-lime gave a high yield of *N*-methyltryptamine. It was concluded that calycanthine was an indole alkaloid derived biogenetically from *N*-methyltryptamine, and structures (I) and (II) were proposed for calycanine and calycanthine respectively. These formulæ were criticised by Manske and Marion [5] who proposed (III) as a possible structure for calycanthine.

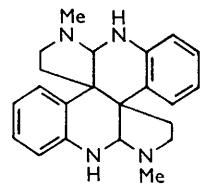
In 1951 Woodward and Clark showed that calycanine has the structure (IV) and confirmed this by synthesis. This result necessitated revision of the previously postulated structures for calycanthine and subsequent spectroscopic and chemical studies indicated that the molecule contains two residues of structure (V) and two *N*-methyl groups.

In 1954 Robinson and Teuber [6] proposed (VI) and (VII) as possible structures for calycanthine and suggested a biogenesis involving ββ′-oxidative coupling of two molecules

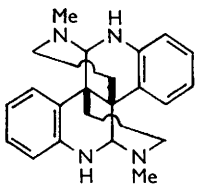
[1] Eccles, *Proc. Amer. Pharm. Assoc.*, 1888, 84, 382.
[2] Gordin, *J. Amer. Chem. Soc.*, 1905, **27**, 144, 1418.
[3] Späth and Stroh, *Ber.*, 1925, **58**, 2131.
[4] Barger, Madinaveitia, and Streuli, *J.*, 1939, 510.
[5] Manske and Marion, *Canad. J. Res.*, 1939, B**17**, 293.
[6] Robinson and Teuber, *Chem. and Ind.*, 1954, 783; Robinson, " The Structural Relations of Natural Products," Clarendon Press, 1955, p. 105.


(VI)


(VII)


(VIII)

of *N*-methyltryptamine. Degradation to calycanine would require a rearrangement and Robinson and Teuber give a mechanism for this.


(IX)

Our *X*-ray work began in 1957 when Mr. Harley-Mason kindly supplied us with a quantity of the pure material and several crystalline derivatives. At that time the structures regarded as most probable for calycanthine were those represented by (VI) and (VIII).[7] Our results are summarised in formula (IX) for calycanthine and in Figs. 2 and 3 which are projection drawings of the molecule as it appears in the crystal. It was only after the completion of the *X*-ray analysis that we learned that this structure had also been considered.[8]

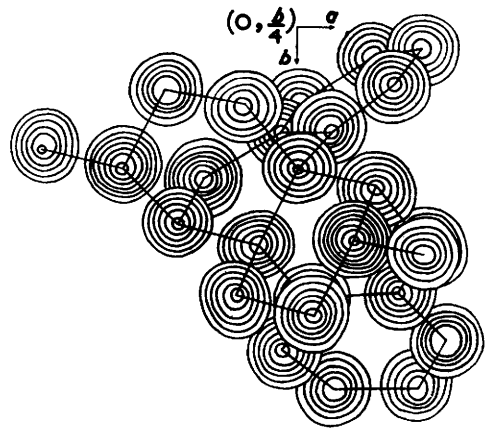


FIG. 1. Final electron-density distribution as superimposed contour sections parallel to (001). Contours are at unit interval but those below 2 eÅ^{-3} are omitted. Atoms are drawn on the nearest section level.

A possible biogenetic route to the alkaloid in accordance with the views of Robinson [6] and Woodward *et al.*[8] is shown in Scheme 1. It may be noted that the calycanine skeleton is preformed in the calycanthine structure, but that it does not contain an indole system.

It had originally been hoped that a study of the isomorphous calcycanthine dihydrochloride dihydrate and dihydrobromide dihydrate by two-dimensional methods would lead to the elucidation of the structure. However, although reliable electron-density projections were obtained, extensive overlap prevented their full interpretation. Certain other derivatives were also examined,[9] but the dihydrobromide was finally selected for three-dimensional study by the heavy-atom phase-determining method.[10]

After location of the bromide ions from Patterson maps the analysis proceeded by means of triple Fourier syntheses, based initially on the bromine phases alone, and then on an increasing number of atoms as these became clearly visible in successive calculations. This was followed by three cycles of least-squares refinement, giving a final discrepancy

[7] Harley-Mason, personal communication.
[8] Ingleby, Ph.D. Dissertation, Cambridge, 1956; Woodward, Yang, Katz, Clark, Harley-Mason, Ingleby, and Sheppard, *Proc. Chem. Soc.*, 1960, 76.
[9] Hamor, Robertson, Shrivastava, and Silverton, *Proc. Chem. Soc.*, 1960, 78.
[10] Robertson and Woodward, *J.*, 1937, 219; 1940, 36.

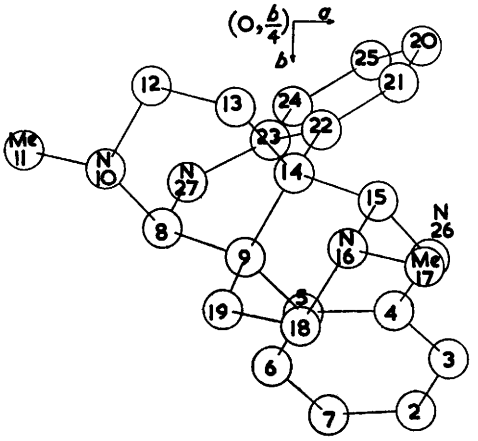


FIG. 2. Drawing of the molecule in projection on (001).

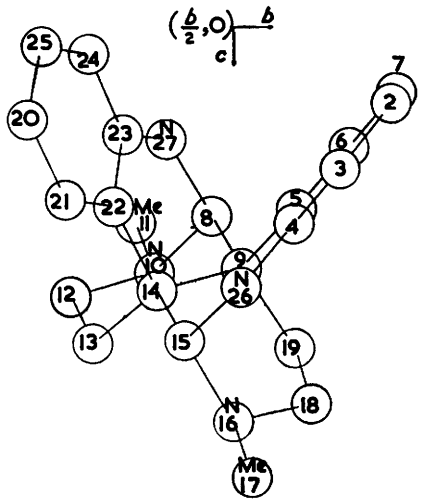
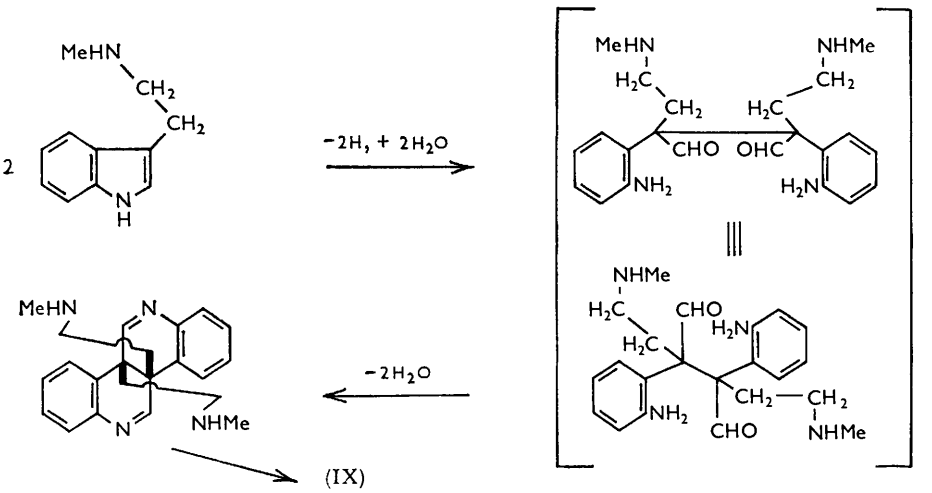


FIG. 3. Drawing of the molecule in projection on (100).



Scheme 1.

over the 2116 observed reflections of 9·6% (Table 4). The course of the analysis is summarised in Table 1.

TABLE 1.

Course of analysis.

Operation	Data used	Atoms included	R (%)	$\Sigma w\Delta^2$
3D Patterson function	2111 F_o	—	—	—
1st 3D F_o synthesis	1858 F_o	2Br	33·8	—
2nd 3D F_o synthesis	1908 F_o	2Br + 15C	28·0	—
3rd 3D F_o synthesis	1987 F_o	2Br + 24C	22·0	—
4th 3D F_o synthesis	2059 F_o	2Br + 26C + 2O	16·0	—
Co-ordinates adjusted	—	2Br + 26C + 2O	15·0	—
Co-ordinates adjusted	—	2Br + 22C + 4N + 2O	14·6	—
5th 3D F_o synthesis	2116 F_o	2Br + 22C + 4N + 2O	14·6	—
1st least-squares cycle	2116 F_o	2Br + 22C + 4N + 2O + 18H	14·6	5100
2nd least-squares cycle	2116 F_o	2Br + 22C + 4N + 2O + 18H	11·1	2700
3rd least-squares cycle	2116 F_o	2Br + 22C + 4N + 2O + 18H	10·9	2515
Co-ordinates adjusted	—	2Br + 22C + 4N + 2O + 18H	9·6	—

The final three-dimensional Fourier synthesis is shown in Fig. 1. The bromide ions and water molecules which lie at some distance from the molecule are not included. The stereochemistry is further indicated by Figs. 2 and 3, which also show the numbering system used in the later Tables. The molecular dimensions are summarised in Figs. 4 and 5. The co-ordinate standard deviations calculated from the least-squares totals

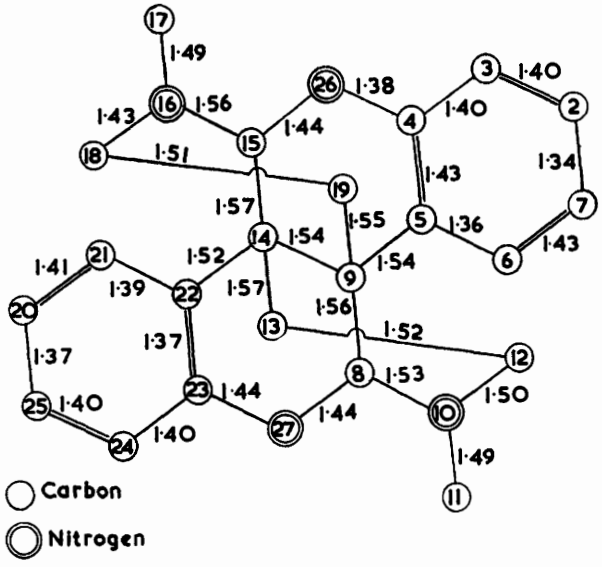


FIG. 4. Bond lengths (Å).

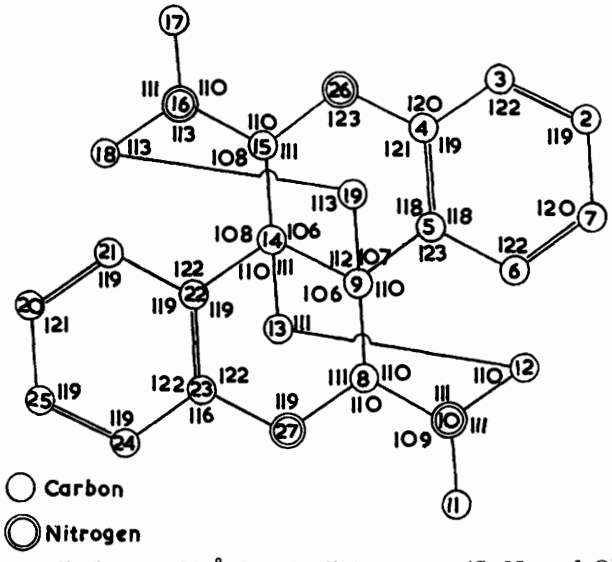


FIG. 5. Bond angles.

Angles: 5—9—14, 109°.
8—9—19, 113°.
9—14—22, 108°.
13—14—15, 113°.

are all about 0·02 Å for the light atoms (C, N, and O) and 0·002 Å for the bromide ions. The standard deviation of a bond length is therefore about 0·03 Å for bonds involving only light atoms. Distances involving halogen have standard deviation 0·02 Å. The corresponding values for angles are 1·7° and 1·3°.

The molecule is a very compact three-dimensional structure consisting of six six-membered rings fused together. The two benzene rings are inclined at 61° to each other, and the other four rings are all of " chair " form, with the *N*-methyl groups, 11 and 17, equatorial. It is of interest that the calycanthine molecule as a whole shows almost exact 2-fold symmetry. This is, of course, permissible for an optically active molecule, as

there is no reflection symmetry present. The benzene rings are planar to within 0·025 Å, and the four adjacent atoms, N(26), C(9), and N(27), C(14) lie near the mean planes of the corresponding aromatic rings. The maximum displacement is 0·13 Å for N(27), which may be compared with a displacement of 0·16 Å found for a similar grouping in the structure of (—)-aspidospermine *N*(b)-methiodide.[11]

The average length of the C–C aromatic bond is 1·39 ± 0·03 Å (maximum deviation 0·05 Å), and of the C–C single bond 1·54 ± 0·02 Å with a maximum deviation of 0·03 Å. The C–N bond distances show a greater spread, varying from 1·38 to 1·56 Å. However, these are not all of the same type. The atoms N(10) and N(16) carry a positive charge since the protons are associated with them, while N(26) and N(27) are directly linked to aromatic nuclei, but do not carry a formal charge. Three different kinds of C–N bond have therefore to be considered, *viz.*, C–N^+, C(sp^2 hybridised)–N, and C(sp^3 hybridised)–N. The C(sp^2)–N bond length has been measured [12] in acetanilide as 1·43 Å and in 2-chloro-4-nitroaniline [13] as 1·37 Å, and for bonds involving a positively charged nitrogen atom, Hahn [14] has listed the results of seven accurate investigations involving six different amino-acids. The C–N^+ lengths of the " zwitterions " vary from 1·474 to 1·528 Å, the mean value being 1·503 Å. If we then assume C–N^+ = 1·50 Å, C(sp^2)–N = 1·40 Å, and for the C(sp^3)–N single bond distance take the generally accepted value of 1·47 Å, all the bond lengths in calycanthine appear to be reasonable and within the limits of experimental error. Even the very long bond C(15)–N^+(16) = 1·56 Å does not deviate significantly from 1·50 Å, nor does the bond C(18)–N^+(16) = 1·43 Å. However, this is actually shorter than the bond C(23)–N(27) which involves an aromatic carbon atom. Owing to the limited accuracy of our results it would not be profitable to discuss these bond lengths in detail, but it is of interest that long C–N^+ bonds ranging from 1·49 to 1·57 Å (mean 1·53 Å) have been reported in the structures of ibogaine hydrobromide,[15] (±)-α-prodine hydrochloride [16] and (+)-demethanolaconinone hydriodide trihydrate.[17] Similarly in echitamine bromide [18] we have a quaternary nitrogen atom linked to carbon atoms at distances varying from 1·54 to 1·56 Å. This structure and that of ibogaine hydrobromide also contain short C (aromatic)–N bonds of length 1·38 and 1·40 Å, but in these analyses the standard deviations are quite large (0·03—0·04 Å for bond lengths).

The bond angles fall within the range of values expected for trigonal and tetrahedral atoms, apart from the angles at N(26) (123°) and N(27) (119°), which appear to be significantly larger than tetrahedral. If these two angles are excluded, the mean angle at sp^3-hybridised atoms is 110° ± 2° (maximum deviation 4°) and at sp^2-hybridised atoms 120° ± 2° (maximum deviation 4°). The whole structure seems to be quite free from strain.

In the crystal the molecules are arranged in layers parallel to (010). Within each layer the molecules are held together by hydrogen bonds involving the bromide ions, the water molecule, $H_2O(28)$, and the nitrogen atoms. The closest approach between atoms of different layers is the $H_2O(28) \cdots H_2O(29)$ distance of 3·15 Å which can represent only a very weak hydrogen bond. This is illustrated in Fig. 6 which shows the structure projected on (100), with the possible hydrogen bond contacts as broken lines. The extra-molecular contacts of less than 4 Å and some of the angles are listed in Table 3. Each of the crystallographically independent bromide ions has three close contacts, Br(0) to nitrogen atoms of two different molecules at 3·22 and 3·35 Å and to a water molecule at 3·36 Å, and Br(1) to a nitrogen atom at 3·42 Å and to water molecules at 3·35 and 3·47 Å. These distances are similar to the $Br^- \cdots N$ and $Br^- \cdots O$ contacts found in the crystal

[11] Mills and Nyburg, *J.*, 1960, 1458.
[12] Brown and Corbridge, *Acta Cryst.*, 1954, **7**, 711.
[13] McPhail and Sim, unpublished results.
[14] Hahn, *Z. Krist.*, 1957, **109**, 438.
[15] Arai, Coppola, and Jeffrey, *Acta Cryst.*, 1960, **13**, 553.
[16] Kartha, Ahmed, and Barnes, *Acta Cryst.*, 1960, **13**, 525.
[17] Przybylska, *Acta Cryst.*, 1961, **14**, 429.
[18] Hamilton, Hamor, Robertson, and Sim, *Proc. Chem. Soc.*, 1961, 63, and unpublished results.

structures of D(−)-isoleucine hydrobromide monohydrate,[19] 11-aminoundecanoic acid hydrobromide hemihydrate,[20] L-cystine dihydrobromide,[21] and ibogaine hydrobromide.[15] The fourth nitrogen atom is hydrogen bonded to a water molecule $N(10)\cdots H_2O(28) = 2{\cdot}89$ Å and the closest bromide ion is Br(1) at a distance of 3·62 Å. The protons are

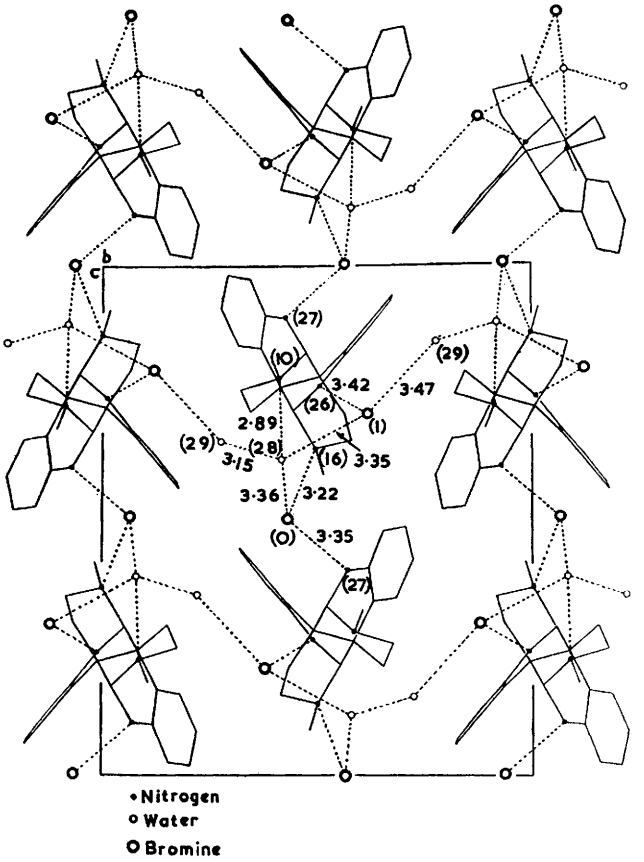


FIG. 6. Projection of the structure on (100).

obviously associated with the tertiary nitrogen atoms N(10) and N(16), and all the available hydrogen atoms appear to take part in hydrogen bonding. The $C{-}N\cdots Br^-$ and $C{-}N\cdots O$ angles deviate considerably from the expected tetrahedral value, but this certainly does not exclude the possibility of hydrogen-bond formation, although the bonds would be comparatively weak. The shortest distance between a carbon atom and a bromide ion is $Br(0)\cdots C(8f) = 3{\cdot}60$ Å. Between the water molecule $H_2O(28)$ and carbon atom C(12) there is a close contact of 3·18 Å but this distance appears to be normal for $C\cdots O$ contacts, values of about 3·2 Å occurring in the crystal structures of other alkaloids (see, *e.g.*, refs. 11 and 17). The closest approach between two calycanthine molecules is 3·49 Å and occurs between C(7) of the reference molecule and C(13) of the molecule related to it by the two-fold screw axis parallel to *b*. The bonding perpendicular to the plane of the projection of Fig. 6 is by means of the following system of hydrogen bonds:

$$N(26)\cdots 3{\cdot}42\text{ Å}\cdots Br(1a)\cdots 3{\cdot}35\text{ Å}\cdots H_2O(28a)\cdots 2{\cdot}89\text{ Å}\cdots N(10a)$$

where the letter *a* refers to the position $x + 1, y, z$.

[19] Trommel and Bijvoet, *Acta Cryst.*, 1954, **7**, 703.
[20] Sim, *Acta Cryst.*, 1955, **8**, 833.
[21] Peterson, Steinrauf, and Jensen, *Acta Cryst.*, 1960, **13**, 104.

Consideration of the distances between the calycanthine molecule and the bromide ions and water molecules leads directly to the location of the four nitrogen atoms, since the $Br^- \cdots$ [calycanthine] contacts of 3·22, 3·35, and 3·42 Å and the $H_2O \cdots$ [calycanthine] contact of 2·89 Å are too short to involve carbon atoms. These results are confirmed by the peak heights in the various Fourier syntheses, by the final temperature factors [22] (Table 2), and by the chemical evidence.

Experimental

Crystal Data.—Calycanthine dihydrobromide dihydrate, $C_{22}H_{26}N_4,2HBr,2H_2O$; M, 544·3; m. p. 213—214°; [2] d calc. 1·568, d found 1·568 (flotation in CCl_4–C_6H_6); orthorhombic, a = 9·61 ± 0·01, b = 14·13 ± 0·01, c = 16·97 ± 0·02 Å. Absent spectra, (h00) when h is odd, (0k0) when k is odd, (00l) when l is odd. Space group $P2_12_12_1$ (D_2^4). Four molecules per unit

Table 2.

Atomic co-ordinates and temperature factors.

Atom	x/a	y/b	z/c	$10^4\beta_{11}$	$10^4\beta_{22}$	$10^4\beta_{33}$	$10^4\beta_{23}$	$10^4\beta_{31}$	$10^4\beta_{12}$	B (Å^2)
Br(0)	−0·1147	0·4338	0·5024	158	88	45	12	33	−11	4·2
Br(1)	−0·4787	0·6204	0·2936	127	89	52	11	−12	11	4·2
C(2)	0·2011	0·6732	0·0704	228	92	34	−4	−8	−93	4·5
C(3)	0·2545	0·6158	0·1305	126	92	48	−2	44	−26	4·0
C(4)	0·1672	0·5634	0·1800	146	64	40	8	−12	−55	3·5
C(5)	0·0205	0·5683	0·1680	100	69	37	−28	5	1	3·1
C(6)	−0·0289	0·6265	0·1105	178	70	52	−29	8	11	4·2
C(7)	0·0627	0·6780	0·0598	197	80	37	6	−21	−65	4·1
C(8)	−0·2055	0·4756	0·1728	104	74	44	12	−34	−35	3·4
C(9)	−0·0737	0·5068	0·2202	73	57	38	−9	16	3	2·7
N(10)	−0·2982	0·4120	0·2235	108	72	47	11	−5	−31	3·5
C(11)	−0·4286	0·3918	0·1792	127	92	65	−14	−38	−8	4·5
C(12)	−0·2241	0·3220	0·2454	180	59	48	−23	−30	2	3·9
C(13)	−0·0898	0·3449	0·2893	179	55	34	8	1	5	3·5
C(14)	0·0047	0·4146	0·2406	83	71	40	27	−34	22	3·1
C(15)	0·1394	0·4452	0·2862	158	57	35	−1	5	4	3·3
N(16)	0·0923	0·4972	0·3625	113	83	41	−17	0	9	3·6
C(17)	0·2151	0·5158	0·4141	157	136	29	−11	−8	−28	4·6
C(18)	0·0172	0·5823	0·3462	179	100	37	−18	2	9	4·4
C(19)	−0·1082	0·5655	0·2949	121	70	49	−32	−16	−20	3·6
C(20)	0·2062	0·2758	0·0840	142	92	59	−15	66	36	4·5
C(21)	0·1703	0·3155	0·1572	154	61	43	0	28	−21	3·6
C(22)	0·0488	0·3683	0·1635	122	74	37	9	−20	−17	3·4
C(23)	−0·0334	0·3796	0·0982	138	81	36	−13	−6	54	3·6
C(24)	0·0027	0·3428	0·0245	115	81	48	−10	13	−27	3·7
C(25)	0·1271	0·2920	0·0179	196	69	48	−32	58	−37	4·2
N(26)	0·2223	0·5075	0·2387	111	76	37	4	−27	−43	3·3
N(27)	−0·1667	0·4263	0·1022	94	79	41	4	3	5	3·3
H_2O(28)	−0·3944	0·4182	0·3848	118	105	37	3	5	−2	3·9
H_2O(29)	−0·6405	0·2772	0·3483	187	146	52	−5	−7	51	5·7
H(2)	0·270	0·719	0·040	—	—	—	—	—	—	4·0
H(3)	0·358	0·614	0·130	—	—	—	—	—	—	4·0
H(6)	−0·133	0·623	0·111	—	—	—	—	—	—	4·0
H(7)	0·017	0·719	0·017	—	—	—	—	—	—	4·0
H(8)	−0·241	0·541	0·170	—	—	—	—	—	—	4·0
H(12)	−0·283	0·297	0·293	—	—	—	—	—	—	4·0
H′(12)	−0·208	0·275	0·203	—	—	—	—	—	—	4·0
H(13)	−0·029	0·283	0·290	—	—	—	—	—	—	4·0
H′(13)	−0·125	0·391	0·335	—	—	—	—	—	—	4·0
H(15)	0·191	0·385	0·286	—	—	—	—	—	—	4·0
H(18)	0·071	0·628	0·316	—	—	—	—	—	—	4·0
H′(18)	−0·008	0·603	0·406	—	—	—	—	—	—	4·0
H(19)	−0·158	0·524	0·328	—	—	—	—	—	—	4·0
H′(19)	−0·179	0·628	0·295	—	—	—	—	—	—	4·0
H(20)	0·291	0·238	0·083	—	—	—	—	—	—	4·0
H(21)	0·237	0·303	0·198	—	—	—	—	—	—	4·0
H(24)	−0·083	0·345	−0·019	—	—	—	—	—	—	4·0
H(25)	0·137	0·258	−0·038	—	—	—	—	—	—	4·0

[22] Rossmann, Jacobson, Hirshfeld, and Lipscomb, *Acta Cryst.*, 1959, **12**, 530.

cell. Volume of the unit cell = 2304 Å^3. Absorption coefficient for X-rays (λ = 1·542 Å), μ = 48·5 cm.$^{-1}$. Total number of electrons per unit cell = $F(000)$ = 1112.

Experimental Measurements.—Well-formed prisms elongated along b were obtained by crystallisation from aqueous alcohol. The cell dimensions were measured from precession photographs using Mo-$K\alpha$ radiation. The intensities were estimated visually from equi-inclination Weissenberg photographs taken with Cu-K_α radiation, the multiple-film technique being used to correlate strong and weak reflections.[23] For upper-layer data the film to film correlation factors given by Rossmann [24] were employed. The crystals were cut so that the cross-section perpendicular to the rotation axis was 0·2 × 0·2 mm. in each case, and absorption corrections were not applied. The intensity data consisted of the layer lines ($0kl$)—($8kl$), ($h0l$), and ($hk0$). 2116 independent structure factors were evaluated after the usual correction factors had been applied. This represents 66% of the total possible for Cu-$K\alpha$ radiation. The structure factors were later placed on an absolute scale by comparison with the calculated values.

Analysis of the Structure.—Approximate halogen positions had been obtained in the course of the preliminary two-dimensional work and these were confirmed by means of a sharpened three-dimensional Patterson synthesis. The first three-dimensional Fourier synthesis based on the bromine phases alone enabled us to assign co-ordinates to 15 of the 28 atoms (C, N, and O). Successive three-dimensional Fourier syntheses were then carried out with the inclusion of further atoms in the phasing calculations as they became clearly defined. The course of the

TABLE 3.

Extra-molecular contacts (Å) and angles.

The letters associated with some of the atoms refer to the following positions:

a: $x+1, y, z$; b: $\bar{x}, \frac{1}{2}+y, \frac{1}{2}-z$; c: $\bar{x}-1, \frac{1}{2}+y, \frac{1}{2}-z$; d: $\frac{1}{2}+x, \frac{1}{2}-y, 1-z$; e: $\frac{1}{2}-x, 1-y, \frac{1}{2}+z$; f: $\bar{x}-\frac{1}{2}, 1-y, \frac{1}{2}+z$; g: $\frac{1}{2}+x, \frac{1}{2}-y, \bar{z}$.

Br(0)···N(16)	3·22	Br(0)···H$_2$O(29d)	3·92	C(19)···C(21b)	3·67
Br(0)···N(27f)	3·35			C(6)···C(13b)	3·70
Br(1a)···N(26)	3·42	H$_2$O(28)···H$_2$O(29)	3·15	C(19)···C(20b)	3·73
Br(0)···C(8f)	3·60			C(18)···C(21b)	3·76
Br(0)···C(18)	3·61	H$_2$O(28)···N(10)	2·89	C(2)···C(12b)	3·77
Br(1)···N(10)	3·62	H$_2$O(28)···C(12)	3·18	C(20)···C(24g)	3·78
Br(1)···C(19)	3·64	H$_2$O(29a)···C(15)	3·35	C(3)···C(13b)	3·85
Br(0)···C(17)	3·69	H$_2$O(28)···C(13)	3·50	C(17)···C(2e)	3·85
Br(1a)···C(3)	3·77	H$_2$O(28)···C(11)	3·52	C(17)···C(24e)	3·85
Br(1)···C(11)	3·80	H$_2$O(28)···C(7f)	3·64	N(26)···C(11a)	3·87
Br(0b)···C(7)	3·80	H$_2$O(29)···C(7f)	3·72	C(4)···C(12b)	3·91
Br(0)···C(13)	3·84	H$_2$O(29a)···C(21)	3·76		
Br(1a)···C(17)	3·88	H$_2$O(28)···C(19)	3·77	C(15)–N(16)···Br(0)	131°
Br(0)···C(11f)	3·90	H$_2$O(29a)···C(17)	3·81	C(18)–N(16)···Br(0)	94
Br(1)···C(8)	3·91	H$_2$O(29)···C(11)	3·87	C(17)–N(16)···Br(0)	96
Br(1)···C(11c)	3·96	H$_2$O(29)···C(2f)	3·88	C(23f)–N(27f)···Br(0)	144
Br(1)···C(24f)	3·96	H$_2$O(29c)···C(6)	3·89	C(8f)–N(27f)···Br(0)	88
Br(0b)···C(2)	3·97	H$_2$O(28)···C(6f)	3·95	C(4)–N(26)···Br(1a)	105
Br(0)···C(19)	3·98	H$_2$O(29a)···N(26)	3·97	C(15)–N(26)···Br(1a)	127
Br(0)···C(6f)	3·98			C(12)–N(10)···H$_2$O(28)	87
Br(1a)···C(4)	3·99	C(7)···C(13b)	3·49	C(8)–N(10)···H$_2$O(28)	135
		C(2)···C(13b)	3·56	C(11)–N(10)···H$_2$O(28)	102
Br(1)···H$_2$O(28)	3·35	C(17)···C(25e)	3·57	Br(0)···H$_2$O(28)···Br(1)	114
Br(0)···H$_2$O(28)	3·36	C(3)···C(12b)	3·61	Br(1)···H$_2$O(29c)···H$_2$O(28c)	107
Br(1)···H$_2$O(29c)	3·47	C(18)···C(20b)	3·67		

analysis is summarised in Table 1. After the 4th F_o synthesis, refinement was continued by the method of least squares, only the diagonal terms of the normal equations being used. At this stage, eighteen hydrogen atoms whose positions could be derived unambiguously were included in the structure-factor calculations but their co-ordinates were not refined. In all these calculations the atomic scattering factors of Berghuis *et al.*[25] were used for carbon and

[23] Robertson, *J. Sci. Instr.*, 1943, **20**, 175.
[24] Rossmann, *Acta Cryst.*, 1956, **9**, 819.
[25] Berghuis, Haanappel, Potters, Loopstra, MacGillavry, and Veenendaal, *Acta Cryst.*, 1955, **8**, 478.

TABLE 4.

Observed and calculated structure factors.

h	k	l	$\|F_o\|$	$\|F_c\|$	α°
0	0	2	117	142	180
		4	100	118	0
		6	92	97	0
		8	53	44	0
		10	120	122	0
		12	30	27	0
		14	75	67	0
		18	27	26	0
		20	25	22	0
0	1	1	11	0	270
		2	23	37	270
		3	88	97	270
		4	50	59	270
		5	71	61	90
		6	24	34	90
		7	86	81	270
		8	60	56	270
		10	22	19	90
		11	10	8	270
		12	10	10	270
		13	19	18	270
		14	52	49	270
		15	22	20	270
		16	46	45	90
		17	35	35	270
		18	43	37	270
0	2	0	154	189	0
		1	24	32	180
		2	58	59	180
		3	17	9	0
		4	60	54	0
		6	45	56	0
		7	65	57	180
		8	71	63	0
		9	58	57	0
		10	65	58	0
		11	43	44	180
		12	29	28	0
		13	47	53	0
		14	21	19	0
		15	14	13	180
		16	17	16	0
		17	17	16	180
		18	22	19	0
		20	9	7	0
		21	9	10	180
0	3	1	90	87	270
		2	74	64	90
		3	176	185	270
		4	145	138	90
		5	5	4	270
		6	86	84	270
		7	150	151	270
		8	53	55	90
		9	42	39	270
		10	17	17	270
		11	61	54	270
		13	40	36	270
		14	14	15	90
		15	30	23	270
		17	52	45	270
		18	10	12	90
		21	7	8	270
0	4	0	129	161	180
		1	56	55	180
		2	78	80	0
		3	42	34	0
		4	21	20	180
		5	45	40	0
		6	13	20	180
		7	76	61	180
		8	26	29	0
		9	21	19	180
		10	75	68	180
		12	42	42	0
		13	21	19	180
		14	9	14	180
		15	21	22	180
		17	11	10	180
		20	13	13	180
		21	5	6	180
0	5	1	85	82	270
		2	10	7	270
		3	20	25	270
		4	48	48	90
		5	176	156	270
		6	114	100	270
		7	65	58	270
		8	36	36	90
		9	61	56	270
		11	35	37	270
		12	9	8	90
		13	27	24	270
		14	20	20	90
		15	21	24	270
		16	18	16	270
		17	17	12	270
		18	16	12	90
		19	21	20	270
		20	11	12	270
0	6	0	80	82	180
		1	78	79	0
		2	32	34	180
		3	35	36	180
		4	24	31	180
		6	78	68	180
		8	40	39	180
		9	16	19	180
		10	79	65	180
		11	46	45	0
		12	19	21	180
		13	33	34	180
		14	10	12	180
		15	14	13	0
		16	15	14	180
		18	19	17	180
		20	17	14	180
0	7	1	37	39	270
		2	16	20	90
		3	29	32	90
		4	13	14	270
		5	85	77	270
		6	41	38	90
		7	80	73	90
		8	12	15	270
		9	32	31	270
		12	10	14	90
		15	23	22	270
		16	20	19	90
		19	14	14	270
0	8	0	10	10	180
		2	117	105	180
		3	18	19	180
		4	53	53	180
		5	19	21	180
		6	57	52	180
		7	37	32	0
		8	45	49	180
		11	15	14	0
		12	57	60	180
		14	10	8	180
		16	29	27	180
		18	22	20	180
0	9	1	38	38	90
		2	21	23	90
		3	18	19	90
		4	16	19	270
		5	32	38	90
		6	21	23	90
		7	9	9	90
		8	9	8	90
		10	9	11	270
		11	29	28	90
		14	10	10	270
		15	16	16	90
		16	13	14	90
		17	11	12	0
0	10	0	11	12	0
		1	44	43	180
		2	39	40	180
		3	47	45	0
		4	20	20	180
		5	21	22	180
		6	20	25	180
		7	36	32	0
		10	19	20	180
		11	27	32	180
		12	10	11	180
		13	22	23	0
		17	7	5	0
		18	10	10	180
0	11	1	50	52	90
		4	18	23	90
		5	66	71	90
		6	41	41	270
		8	21	23	90
		11	18	23	90
		13	19	15	270
		14	42	36	90
		15	18	16	90
		16	13	13	90
		18	21	21	90
		19	6	4	90
0	12	2	15	16	0
		5	15	14	180
		7	10	10	0
		8	20	22	0
		10	24	21	180
		11	9	10	180
		12	18	19	0
		13	17	15	0
		15	7	6	180
		16	5	6	0
0	13	1	12	15	90
		3	9	11	90
		4	10	12	90
		5	17	17	90
		6	30	32	270
		7	22	25	90
		8	18	18	90
		10	11	10	270
		11	18	16	90
		13	9	11	90
		14	8	4	270
0	14	0	22	24	0
		1	20	24	0
		2	13	18	0
		3	14	13	180
		4	15	16	0
		6	21	17	0
		8	22	21	0
		10	13	13	0
		11	14	15	0
		12	8	11	0
		13	6	8	180
		15	12	15	90
		16	14	13	270
		17	10	10	90
		18	7	7	270
0	16	0	34	34	0
		1	11	11	0
		2	10	11	0
0	17	3	13	12	270
		4	8	9	270
		6	10	10	90
		7	6	8	270
0	18	0	6	11	0
		1	8	12	180
1	0	1	69	89	90
		2	11	24	180
		3	94	99	90
		4	9	9	180
		5	5	10	270
		6	20	22	180
		7	24	27	270
		9	42	48	90
		11	67	63	270
		12	9	6	0
		13	37	39	90
		14	12	18	0
		17	17	14	270
1	1	0	76	74	90
		1	29	26	71
		2	46	42	68
		3	130	131	356
		4	112	112	129
		5	52	40	47
		6	99	105	34
		7	122	119	347
		8	86	81	150
		9	13	12	354
		10	66	68	87
		11	26	29	347
		12	16	16	110
		13	42	44	355
		14	43	41	159
		15	32	33	326
		16	13	17	45
		17	15	18	9
		18	28	27	114
		20	13	15	60
		21	13	16	347
1	2	0	145	187	270
		1	76	72	74
		2	68	80	18
		3	107	109	342
		4	169	164	202
		6	48	47	302
		7	48	45	288
		8	81	82	350
		9	21	19	8
		10	88	92	265
		11	38	38	336
		12	9	14	78
		13	23	22	345
		14	45	48	286
		15	9	11	4
		16	10	13	21
		17	11	13	314
		19	12	12	334
		20	22	21	284
		21	8	9	315
1	3	0	22	24	90
		1	140	145	25
		2	88	88	71
		3	61	57	97
		4	122	107	194
		5	55	58	323
		6	93	98	38
		7	23	20	271
		8	41	38	193
		9	28	34	2
		10	8	4	56
		11	12	13	294
		12	13	14	90
		13	13	16	71
		14	31	28	153
		15	21	23	347
		16	10	15	21
		17	10	12	205
		18	15	19	137
		19	10	14	1
		21	7	7	272
1	4	0	26	30	270
		1	87	90	64
		2	101	99	298
		3	142	152	323
		4	73	82	249
		5	26	23	72
		6	13	15	221
		7	69	65	27
		8	51	49	283
		9	46	48	360
		10	43	40	264
		11	63	56	63
		12	31	28	255
		13	47	51	302
		14	29	32	247
		15	29	32	31
		16	37	30	290
		17	23	21	25
		18	14	13	245
		19	9	8	337
		20	17	16	280
		21	8	10	6
1	5	0	110	117	270
		1	15	20	181
		2	57	57	224
		3	87	94	180
		4	27	31	321
		5	30	31	238
		6	37	39	176
		7	56	61	177
		8	54	50	336
		9	21	23	347
		10	32	33	229
		11	9	14	188
		12	14	17	262
		13	33	37	159
		14	21	25	312
		15	8	10	277
		16	13	20	180
		17	21	21	205
		20	9	11	215
1	6	0	51	56	90
		1	50	52	34
		2	98	109	270
		3	48	44	12
		4	7	6	224
		5	21	24	40
		6	28	28	226
		7	28	28	30
		8	68	66	273
		9	34	32	10
		10	15	15	131
		11	28	26	52
		12	40	45	203
		13	19	19	319
		14	12	14	138
		15	20	18	33
		16	25	22	270
		17	11	7	81
		18	11	9	270
		19	9	8	67
		20	8	8	163
1	7	0	42	47	270
		1	54	56	171
		2	40	45	240
		3	28	28	180
		4	56	63	329
		5	77	89	164
		6	43	43	192
		8	54	55	290
		9	55	56	192
		10	23	25	239
		11	21	22	213
		12	25	25	235
		13	19	20	157
		14	20	20	315
		15	24	22	189
		16	24	23	229
		18	12	12	319
		19	22	21	184
		20	9	10	251
1	8	1	58	62	258
		2	13	15	31
		3	54	56	113
		4	19	22	91
		5	13	14	264
		6	18	18	262
		7	32	36	256
		8	14	15	7
		9	35	37	85
		10	24	24	147
		11	14	18	242
		13	22	26	99
		16	7	8	89
		19	6	7	66
1	9	0	37	37	270
		1	36	40	193
		2	34	38	301
		4	27	30	228
		5	67	70	185
		6	20	21	328
		7	25	26	35
		8	46	50	242
		9	58	61	176
		10	30	28	341
		11	11	11	185
		12	14	19	283
		13	8	10	144
		14	8	9	247
		15	23	23	166
		16	14	15	320
		17	9	9	32
		19	13	9	180
1	10	0	24	24	270
		1	18	22	226
		2	50	57	87
		3	14	17	132
		4	20	20	104
		5	25	30	176
		6	9	11	75
		7	36	40	203
		8	44	50	93
		9	19	23	126
		11	19	16	243
		12	28	30	90
		15	10	10	177
		16	13	13	87
		17	8	8	164
1	11	2	12	18	329
		3	19	21	184
		4	21	21	171
		6	21	25	344
		8	14	17	176
		10	19	19	8
		16	14	13	344
		17	7	9	162
1	12	0	16	16	90
		1	35	40	119
		2	30	32	83
		3	35	34	232
		4	22	24	94
		5	14	17	152
		6	8	10	79
		7	12	16	144
		8	17	20	90
		9	13	17	220
		10	14	13	48
		11	7	10	147
		12	12	12	82
		13	19	21	243
		14	11	11	70
		15	10	11	142
		16	6	7	58
1	13	0	11	11	90
		1	21	24	39
		2	11	13	107
		3	8	9	209
		4	11	13	34
		5	20	29	25
		8	12	13	54
		9	14	17	0
		10	11	11	102
		14	8	8	81
		15	13	17	12
1	14	0	19	24	90
		1	22	19	105
		2	8	9	292
		3	13	14	233
		4	11	12	65
		5	11	7	224
		6	10	10	120
		7	12	12	152
		10	11	14	61
		13	6	7	199
1	15	0	8	6	90
		1	8	10	40
		2	21	20	124
1	16	2	10	11	255
		3	11	10	69
		4	10	8	342
		5	9	9	299
1	17	3	12	14	5
		4	7	8	86
		7	10	14	347
2	0	0	113	129	0
		1	133	132	90
		2	71	54	180
		3	119	124	90
		4	45	45	0
		5	151	156	90
		6	84	72	0
		7	101	92	90
		8	27	30	180
		10	57	59	0
		11	67	62	90
		12	46	48	180
		13	64	60	90
		14	18	24	0
		15	32	29	90
		16	9	12	180
		17	37	31	90
		19	12	13	90
		20	13	15	0
		21	9	12	90
2	1	0	54	56	0
		1	82	65	21
		2	63	63	65
		3	68	56	271
		4	116	108	292
		5	77	68	94
		6	46	51	72
		7	39	39	312
		8	40	40	285
		9	18	19	43
		10	32	35	43
		12	19	19	315
		13	9	12	267
		14	33	33	311
		15	13	17	61
		16	17	20	51
		17	23	21	303
		18	12	13	306
		20	10	12	13
2	2	0	22	24	0
		1	115	114	186
		2	8	7	129
		3	160	129	50
		4	69	62	262
		5	95	82	101
		6	29	23	51
		7	53	48	118
		8	13	9	37
		9	52	59	38
		11	61	62	134
		12	16	15	277
		13	32	38	24
		14	9	11	7
		15	22	27	137
		17	27	26	70
		18	10	12	270
		19	9	8	58
		21	18	19	138
2	3	0	79	95	0
		1	33	35	105
		2	146	131	351
		3	92	82	288
		4	121	99	26
		5	62	58	104
		6	101	96	326
		7	58	63	238
		8	76	76	46
		9	16	19	31
		10	55	65	352
		11	57	53	8
		14	28	28	27
		15	13	16	102
		16	36	35	310
		17	21	21	282
		18	21	18	44
		20	17	17	340
2	4	0	176	179	180
		1	11	9	181
		2	114	98	322
		3	10	12	112
		4	40	42	128
		5	25	26	305
		7	34	32	196
		8	40	44	351
		9	15	15	55
		10	50	52	198
		11	18	19	177
		12	22	26	295
		13	13	13	293
		14	12	12	251
		15	17	16	188
		16	17	16	337
		20	14	15	223
2	5	0	46	46	0
		1	75	67	244
		2	95	92	347
		3	58	65	23
		4	41	46	62
		5	42	41	275
		6	105	102	330
		7	31	31	130
		8	67	71	35
		9	23	22	291
		10	32	32	314
		11	21	21	227
		12	61	61	0
		14	28	26	75
		16	36	33	313
		18	23	24	49
2	6	0	37	38	180
		1	49	61	303
		2	47	46	317
		3	68	69	219
		4	49	46	104
		5	80	78	289
		6	29	32	265
		7	51	51	275
		8	17	20	95
		9	45	49	221
		10	9	9	340
		11	46	50	323
		12	25	26	189
		13	36	43	222
		14	10	11	64
		15	20	19	293
		17	14	14	274
		19	9	8	194
		20	9	9	230
2	7	0	20	21	0
		1	30	30	259
		2	26	28	346
		3	60	63	85
		4	36	41	280
		5	51	52	252
		6	31	33	88
		7	25	29	109
		8	9	11	308
		9	8	11	255
		10	12	12	93
		11	11	14	188
		12	20	19	336
		13	11	11	158
		15	14	12	242
		16	15	16	30
		17	17	16	215
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		2	54	57	187
		3	30	35	279
		4	12	11	41
		5	63	69	275
		6	12	10	241
		7	36	34	286
		8	23	27	194
		9	35	39	255
		10	33	35	9
		11	29	28	290
		12	29	30	162
		13	34	28	261
		15	37	32	297
		16	10	11	176
		17	11	12	267
		19	10	12	260
2	9	0	10	13	180
		1	17	17	52
		2	41	42	156
		3	12	15	228
		4	30	32	244
		5	20	24	84
		6	38	39	117
		7	12	13	252
		8	14	20	209
		9	11	13	112
		10	16	18	120
		11	9	11	332
		12	18	19	174
		16	18	18	127
		18	7	8	212
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		2	28	29	112
		3	21	25	344
		4	23	23	236
		6	15	15	90
		7	12	16	231
		8	13	14	213
		9	26	31	327
		11	26	28	199
		13	21	20	340
		14	15	13	254
		16	7	7	82
		17	6	5	180
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		3	38	44	176
		4	23	26	298
		5	21	27	168
		6	21	27	106
		7	36	39	209
		8	12	18	270
		9	28	30	156
		10	17	20	66
		11	14	19	185
		12	23	24	182
		14	10	13	147
		15	7	8	93
		16	14	15	206
2	12	0	23	26	180
		1	21	24	19
		3	14	14	10
		4	14	16	234
		5	11	16	96
		6	11	12	38
		7	16	15	225
		10	21	20	170
		12	9	11	36
2	13	0	12	8	6
		2	9	13	180
		3	2	4	201
		4	11	17	53
		5	4	17	21	119
		6	23	27	224
		7	12	11	111
		8	15	15	174
		9	12	11	350
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		12	9	9	192
		13	9	8	7
		14	7	7	144
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TABLE 4. (*Continued*).

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	9	11	14	11	3
	10	8	9	9	173
	11	16	17	17	49
	13	8	9	9	137
2	15	3	13	15	99
	5	9	11	11	251
	7	17	16	16	69
	9	11	10	11	305
	0	14	13	14	0
2	16	3	9	14	122
	5	10	10	10	59
	7	15	15	15	97
	8	7	7	7	198
2	17	2	8	9	16
3	0	1	107	103	270
		3	73	65	90
		4	39	41	0
		5	34	27	270
		6	61	61	180
		7	33	28	270
		8	27	29	0
		9	81	82	90
		10	10	10	0
		11	42	44	270
		13	27	31	90
		14	13	15	0
		15	10	13	270
		17	9	8	270
		18	17	15	0
		19	8	10	90
		20	6	8	180
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		1	88	83	208
		2	85	75	35
		3	43	35	101
3	1	4	110	101	122
		5	124	121	184
		6	35	35	33
		7	37	32	354
		8	56	51	119
		9	80	79	192
		10	86	83	83
		11	16	16	235
		12	16	15	60
		13	13	14	168
		14	30	33	112
		15	30	33	200
		16	25	23	47
		17	11	10	317
		18	18	18	136
		19	17	19	185
		20	13	15	56
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		1	82	69	341
		2	133	121	95
		3	66	63	1
		4	44	39	255
		5	16	18	27
		6	48	44	72
		7	90	83	355
		8	69	65	81
		9	38	42	19
		10	40	42	326
		11	18	20	326
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		13	29	31	2
		15	11	9	22
		16	14	14	72
		17	23	24	3
		18	23	19	69
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		1	41	45	270
		2	42	37	60
		3	61	52	152
		4	87	77	112
		5	47	45	321
		6	23	25	346
		7	42	43	185
		8	51	49	142
		9	16	20	33
		10	8	12	306
		11	26	25	249
		12	39	41	54
		13	24	31	154
		14	24	27	181
		15	21	18	266
		16	17	14	32
		17	9	10	207
		18	15	12	143
		20	7	10	24
3	4	0	27	32	90
		1	84	78	24
		2	80	76	74
		3	108	96	317
		4	62	59	70
		5	30	31	10
		6	70	68	50
		7	69	71	25
		8	41	42	135
		9	53	58	311
		10	35	36	43
		11	46	47	53
		12	35	32	137
		13	53	51	304
		14	16	20	101
		15	22	19	27
		17	25	25	3
		18	18	19	123
		19	13	14	325
		20	5	8	51
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		1	55	54	359
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		3	45	42	164
		4	59	55	357
		5	90	88	350
		6	65	64	233
		7	30	31	203
		8	36	39	323
		9	32	37	1
		10	32	34	237
		11	11	9	287
		12	11	12	30
		13	9	13	355
		14	16	20	330
		15	16	16	331
		16	20	21	249
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		1	37	39	319
		2	23	25	279
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		6	24	27	97
		7	17	17	46
		8	21	22	208
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		13	39	38	51
		14	16	16	222
		15	10	11	325
		16	30	26	107
		17	14	14	345
		18	16	10	9
		19	8	7	3
		20	19	7	8
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		1	15	13	161
		2	80	74	257
		3	55	59	353
		4	23	28	335
		5	22	24	356
		6	67	67	235
		7	31	33	16
		8	38	44	310
		9	9	10	334
		10	22	24	230
		11	15	16	59
		12	29	30	249
		13	33	31	319
		14	24	23	334
		15	12	9	27
		16	33	30	230
		17	24	23	354
		1	27	30	256
		2	46	46	288
		3	48	49	121
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		5	17	17	302
		6	23	22	202
		7	20	23	219
		8	7	8	255
		9	14	18	126
		10	16	16	281
		12	12	15	235
		13	20	20	33
		16	11	12	173
		1	13	17	303
		2	48	51	277
		3	49	54	353
		4	27	30	217
		5	14	15	78
		6	44	47	296
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		8	29	33	229
		10	11	13	286
		11	14	11	36
		12	19	19	246
		13	17	17	15
		14	18	16	248
		16	10	11	290
		17	13	13	32
		0	47	53	270
		1	25	29	181
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		3	19	21	189
		4	17	22	234
		5	19	24	175
		6	18	22	263
		7	9	12	234
		8	36	31	181
		10	26	25	277
		11	17	16	189
		12	14	14	195
		14	20	20	272
		15	11	12	179
		0	11	12	270
		1	18	21	299
		2	16	18	347
		3	16	15	159
		4	25	26	188
		5	11	11	24
		6	36	31	3
		7	13	9	99
		8	14	8	9
		0	15	20	270
		1	30	33	132
		2	12	10	269
		3	31	32	230
		4	13	18	242
		5	17	21	149
		6	9	10	6
		7	21	21	138
		8	11	9	297
		9	10	12	200
		10	19	21	282
		11	19	21	137
		13	13	13	233
		14	12	11	293
		2	14	13	103
		3	22	23	174
		4	19	19	91
		7	22	24	161
		8	11	13	47
		10	9	10	140
		12	9	10	83
		13	12	12	194
3	14	0	9	4	90
		1	11	11	118
		2	13	15	274
		3	17	18	203
		5	10	8	335
		7	22	21	165
		8	15	16	264
		10	8	9	55
		11	9	10	200
3	15	0	12	14	90
		1	12	11	150
		4	23	22	51
		5	11	10	187
		6	9	11	161
		8	11	11	64
		10	10	11	120
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		3	14	12	91
		5	10	9	336
		6	7	8	102
3	17	0	12	11	90
		1	11	9	222
		2	7	5	21
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		3	61	57	90
		4	28	34	180
		5	8	1	270
		6	101	95	180
		7	41	43	90
		8	68	76	180
		9	28	23	270
		10	31	30	180
		11	27	32	90
		12	42	41	180
		14	26	28	180
		16	27	26	180
		17	15	15	90
		18	30	26	180
		20	7	8	180
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		1	66	56	68
		2	26	19	229
		3	37	26	307
		4	41	34	340
		5	94	86	118
		6	37	37	87
		7	38	40	330
		8	15	18	229
		9	44	44	82
		10	20	21	337
		11	20	15	47
		12	26	22	137
		13	12	11	82
		14	21	22	345
		15	18	16	93
		16	21	24	111
		17	9	8	303
		18	7	11	278
		19	12	14	83
		20	8	8	50
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		1	94	89	161
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		3	25	21	53
		4	98	75	236
		5	45	35	339
		6	50	48	130
		7	21	20	130
		8	45	44	216
		9	39	40	348
		10	46	46	183
		11	44	47	137
		12	8	9	139
		13	39	41	11
		14	26	25	207
		15	7	12	184
		16	18	18	169
		17	9	9	344
		18	14	15	215
		19	6	4	42
		20	9	9	156
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		1	93	100	107
		2	22	20	208
		3	68	63	40
		4	85	82	47
		5	130	126	105
		6	39	39	291
		7	12	12	108
		8	13	19	57
		9	82	75	83
		10	19	19	330
		11	29	31	99
		12	11	14	160
		14	24	26	69
		15	29	30	23
		16	33	32	91
		17	9	9	251
		18	10	8	75
		19	6	6	74
		20	18	20	85
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		1	16	14	160
		2	60	58	22
		3	11	14	216
		4	17	12	234
		5	19	17	48
		6	18	14	211
		7	34	33	293
		8	15	15	29
		9	24	20	75
		10	30	27	223
		12	36	37	0
		14	20	19	193
		15	9	9	50
		16	12	13	320
		17	11	11	317
		18	10	8	309
		19	9	8	64
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		1	62	63	133
		2	42	43	331
		3	63	63	79
		4	43	42	76
		5	7	6	156
		6	41	41	286
		7	75	72	97
		8	22	22	25
		9	28	29	51
		11	34	31	108
		12	22	21	358
		13	37	34	89
		15	25	21	134
		16	20	17	294
		17	28	26	86
		19	5	5	97
4	6	0	45	48	0
		1	52	50	347
		2	70	68	334
		3	40	39	193
		4	50	55	25
		6	32	36	315
		7	20	21	291
		8	33	36	12
		9	31	32	197
		10	35	34	353
		11	28	29	354
		12	25	26	358
		13	25	26	198
		16	20	21	326
		18	10	12	32
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		1	10	12	3
		2	23	29	9
		3	43	44	113
		4	35	35	244
		5	22	23	243
		6	32	30	52
		7	38	42	61
		8	33	33	283
		9	9	10	203
		10	14	13	198
		11	7	6	124
		12	12	11	309
		13	18	14	156
		14	13	14	247
		17	11	11	88
		18	11	14	299
4	8	0	92	105	0
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		2	13	13	34
		3	9	9	1
		4	45	52	357
		5	25	26	233
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		7	23	24	52
		8	16	17	351
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		2	6	7	121
		3	33	35	239
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		8	18	21	261
		9	15	17	219
		10	14	15	22
		11	10	10	313
		12	14	14	155
		13	20	20	222
		16	13	13	81
		17	13	16	272
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		1	30	33	200
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		3	29	29	19
		4	38	40	311
		5	29	26	221
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		12	8	3	340
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		14	11	12	349
		16	7	5	8
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		3	42	50	289
		4	7	9	124
		5	9	12	262
		6	13	14	222
		7	47	44	263
		8	13	13	134
		9	19	17	272
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		11	10	9	280
		12	12	12	157
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		15	11	9	221
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		7	13	12	239
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		11	7	8	144
		12	16	14	12
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		5	21	20	240
		6	19	17	276
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		8	11	10	111
		9	18	15	316
		10	6	7	216
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		12	6	3	138
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		2	11	10	208
		3	12	11	174
		4	15	15	162
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		6	11	13	198
		8	11	10	151
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		6	6	7	97
		7	9	9	92
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		2	10	15	179
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		4	5	8	162
		6	12	9	201
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		18	10	12	180
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		4	36	32	0
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		6	36	36	180
		7	15	18	270
		8	31	29	0
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		11	45	50	270
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		15	13	16	270
		17	9	7	90
		18	10	11	0
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		20	4	8	180
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		1	108	91	183
		2	84	71	279
		3	18	20	78
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		5	105	105	183
		6	37	35	328
		7	15	18	177
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		9	50	51	152
		10	20	18	42
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		3	12	7	134
		4	23	20	95
		5	49	51	181
		6	36	39	90
		7	13	10	27
		8	43	47	82
		9	29	28	171
		10	7	9	196
		12	43	44	92
		13	22	26	99
		14	16	17	98
		15	25	25	197
		16	8	14	78
		18	19	17	89
		19	9	9	146
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		1	33	31	275
		2	37	33	34
		3	34	34	161
		4	40	41	206
		5	8	9	316
		6	47	45	359
		7	62	62	188
		8	28	29	204
		9	10	12	44
		10	25	27	312
		11	17	17	247
		12	15	12	354
		13	12	12	160
		14	7	12	205
		16	17	15	11
		17	18	20	176
5	4	0	55	55	90
		1	45	46	104
		2	57	57	68
		3	58	53	244
		4	60	59	123
		5	34	30	144
		6	67	64	57
		7	30	31	151
		8	38	42	113
		9	47	45	249
		10	18	18	47
		11	23	23	138
		12	44	38	96
		13	23	22	243
		14	23	23	113
		15	15	15	145
		16	17	18	32
		17	9	7	188
		18	14	14	114
		19	7	9	242
5	5	0	10	4	270
		1	34	35	33
		2	33	32	109
		3	28	25	292
		4	12	10	328
		5	57	57	357
		6	26	26	95
		7	21	20	136
		8	45	41	40
		9	30	33	347
		10	20	18	300
		11	26	26	17
		12	22	22	72
		13	14	12	257
		14	10	10	6
		15	18	19	13
		17	8	7	237
		18	11	11	36
		19	8	12	357
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		1	6	3	257
		2	12	12	7
		3	48	51	201
		4	52	49	99
		5	24	22	56
		6	29	32	99
		7	39	43	176
		8	10	11	154
		9	21	19	323
		10	35	40	81
		11	11	11	203
		13	14	13	196
		14	22	21	80
		15	8	6	341
		16	8	8	73
		17	15	15	175
		18	6	6	158
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		2	38	40	49
		3	23	22	152
		5	56	55	339
		6	34	36	31
		7	20	20	39
		8	31	29	172
		9	18	17	310
		10	32	35	115
		11	32	36	12
		13	27	26	334
		14	16	14	49
		15	8	9	28
		16	6	8	166
		17	16	15	347
		18	11	13	49
5	8	0	8	8	90
		1	37	39	262
		2	14	16	245
		3	16	15	91
		4	29	32	336
		5	19	18	12
		6	29	28	232
		7	15	14	261
		8	9	9	242
		9	7	9	18
		10	11	9	127
		11	14	13	285
		13	9	9	102
		15	12	10	296
		16	9	10	165
		17	6	4	236
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		4	17	19	118
		5	16	19	4
		6	8	11	91
		7	28	31	339
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		9	11	12	344
		10	24	24	92
		11	17	16	6
		12	12	9	204
		13	22	20	13
		14	11	11	121
		15	7	6	293
		16	5	2	158
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		1	16	22	354
		4	20	25	267
		6	28	30	268
		7	27	28	341
		8	7	8	70
		9	9	9	189
		10	31	28	268
		11	12	11	309
		13	13	15	5
		14	22	23	281
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		1	16	15	309
		3	18	17	109
		4	9	9	188
		5	16	14	343
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		10	14	13	332
		11	7	6	303
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		13	13	13	97
		14	8	8	199
		15	5	7	308
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TABLE 4. (*Continued*).

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		8	7	8	235
		10	9	12	280
		11	6	8	55
		12	13	11	241
		13	9	7	22
5	13	0	26	23	270
		2	12	12	137
		4	11	11	161
		5	20	21	180
		6	13	13	328
		7	6	8	197
		8	16	17	176
		10	12	11	260
		11	7	8	218
		12	4	4	212
5	14	2	17	19	270
		6	9	9	313
		8	13	10	260
		9	6	7	2
		10	4	3	24
5	15	1	11	11	171
		2	8	7	212
		4	8	8	342
		5	11	11	178
		6	7	10	214
		7	7	9	142
		8	6	8	322
5	16	0	6	8	90
		1	6	6	285
		3	10	8	153
6	0	0	27	28	0
		1	78	67	270
		2	55	46	180
		3	25	25	270
		4	7	9	180
		5	88	83	270
		6	5	5	180
		7	23	23	270
		8	42	38	180
		9	57	59	270
		10	7	8	0
		11	37	37	270
		12	15	11	180
		13	8	12	270
		14	13	9	180
		15	31	31	270
		16	17	18	180
		18	5	6	180
		19	15	21	270
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		2	38	37	186
		3	42	35	195
		4	28	27	277
		5	38	32	62
		6	57	47	321
		7	18	20	309
		8	25	28	202
		9	34	32	133
		10	16	13	73
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		12	27	26	187
		13	14	14	187
		15	15	13	46
		16	17	16	107
		17	8	8	255
		18	9	7	193
		19	7	9	129
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		2	40	37	117
		3	57	50	319
		4	53	53	260
		5	32	24	306
		6	47	41	111
		7	21	23	216
		8	36	35	236
		9	44	39	342
		10	25	23	97
		11	35	36	249
		13	26	24	318
		14	16	16	207
		15	17	17	275
		16	10	12	95
		17	11	7	307
		18	10	10	268
		19	7	9	300
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		2	78	77	195
		3	11	12	352
		4	53	53	159
		5	27	22	69
		6	37	38	181
		7	29	28	184
		8	53	53	173
		9	42	46	65
		10	10	10	134
		12	38	36	181
		13	19	22	12
		14	19	20	123
		15	16	15	125
		16	24	23	193
		17	6	6	323
		18	14	14	164
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		1	9	10	78
		2	39	40	26
		3	20	20	234
		4	26	22	192
		5	38	34	39
		6	26	22	65
		7	36	35	227
		8	15	14	290
		9	37	31	87
		10	31	27	139
		11	18	13	345
		12	15	16	346
		13	8	8	288
		14	11	10	189
		15	16	15	62
		16	13	12	50
		17	10	11	291
		18	3	3	21
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		1	37	35	173
		2	35	37	210
		3	74	58	37
		4	59	53	154
		5	28	24	281
		6	23	19	223
		7	44	38	128
		8	34	26	204
		9	13	11	39
		10	35	30	183
		11	18	19	144
		12	15	12	195
		13	19	18	42
		14	29	27	163
		16	25	25	236
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		2	48	44	330
		3	38	39	125
		4	37	37	83
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		6	36	34	311
		7	33	30	64
		9	21	22	145
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		11	31	26	36
		13	15	18	134
		15	29	25	60
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		17	7	7	123
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		3	22	22	120
		4	26	26	242
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		7	22	23	80
		8	26	24	268
		9	15	13	268
		10	21	22	187
		11	12	13	13
		12	15	14	349
		13	7	8	108
		14	13	10	209
		15	8	8	307
		16	11	10	283
		17	7	8	51
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		1	12	15	90
		2	14	15	301
		3	15	18	87
		4	35	31	18
		5	17	16	109
		6	12	12	288
		7	49	53	76
		8	8	9	107
		9	7	9	114
		10	36	27	358
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		13	21	20	93
		14	12	12	8
		15	8	7	88
		16	5	6	1
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		1	24	22	347
		2	20	20	89
		3	28	24	205
		4	40	39	307
		5	16	17	66
		6	20	23	68
		7	16	16	272
		8	16	14	299
		9	9	9	180
		10	16	17	5
		12	5	6	41
		13	7	9	224
		14	10	11	338
		15	8	9	52
6	10	1	21	19	157
		2	11	10	68
		3	19	17	58
		4	11	11	319
		5	13	11	157
		6	11	12	79
		7	10	12	117
		8	12	13	268
		9	11	12	50
		10	9	8	323
		11	17	16	169
		12	9	5	297
		13	7	8	64
		14	9	8	310
		15	4	6	147
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		1	7	8	241
		2	15	14	323
		3	31	24	278
		4	31	30	13
		5	11	10	63
		6	15	15	4
		7	13	15	260
		8	11	13	14
		9	6	3	273
		10	24	23	352
		11	5	7	239
		12	10	7	13
		13	4	6	271
6	12	0	16	16	180
		1	7	5	86
		2	15	13	356
		3	14	12	270
		4	11	10	216
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		7	16	13	267
		8	6	6	359
		9	9	8	91
		10	10	10	174
		11	10	9	220
		12	11	12	5
		0	10	9	0
		2	18	19	324
		3	10	10	302
		4	12	10	81
		6	9	10	308
		7	9	8	197
		8	11	12	40
		9	8	7	310
		11	6	10	205
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		7	9	10	287
		9	6	7	233
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		2	9	9	342
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		4	8	7	187
		6	8	9	53
		8	5	3	73
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		3	8	8	91
		4	11	11	282
		5	12	13	343
		6	14	15	198
		7	16	15	286
		8	11	14	14
		9	18	14	251
7	2	0	56	56	270
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		2	9	11	62
		3	5	8	146
		4	33	38	278
		5	69	58	181
		6	5	4	181
		7	18	17	3
		8	10	10	157
		9	41	46	184
		10	25	28	261
		11	25	24	188
		12	13	13	120
		14	12	13	268
		15	18	19	181
		16	9	7	211
7	3	0	29	27	270
		1	16	16	301
		2	11	11	3
		3	26	24	135
		4	31	30	228
		5	53	47	358
		6	20	19	330
		7	13	10	159
		8	7	5	326
		9	20	23	25
		10	33	34	272
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		12	14	12	55
		13	24	23	99
		14	8	11	232
		16	11	11	343
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		3	24	24	214
		4	40	41	214
		5	34	31	175
		6	47	44	14
		7	41	37	157
		8	23	22	244
		9	27	30	219
		10	13	17	277
		11	32	35	148
		13	22	22	229
		14	8	8	266
		16	6	8	343
		17	13	14	176
7	5	0	12	10	270
		1	42	43	87
		2	36	33	89
		3	39	37	210
		4	17	17	22
		5	23	22	349
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		16	5	3	348
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		2	11	12	122
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		4	19	22	85
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		6	20	29	114
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		8	17	18	70
		9	20	19	226
		10	21	24	100
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		14	17	19	58
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		16	7	14	128
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		2	12	14	242
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		4	22	24	24
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		6	13	19	166
		7	8	6	273
		8	19	20	10
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		10	11	10	143
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		15	9	10	273
7	9	0	50	59	90
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		2	12	13	343
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		5	14	15	198
		6	20	20	75
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		8	9	11	124
		9	7	9	137
		10	24	24	92
		11	11	9	262
		14	12	15	83
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		3	18	24	6
		4	6	7	321
		5	9	8	212
		6	7	5	162
		7	24	27	358
		8	10	11	66
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		12	9	11	75
		13	5	9	4
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		3	7	9	83
		5	7	7	18
		6	10	7	36
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		3	11	13	338
		4	14	15	159
		5	6	9	335
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		8	13	14	140
		9	6	8	330
		11	3	9	18
7	13	0	19	21	270
		2	7	8	164
		4	11	11	300
		6	13	13	224
		8	4	6	353
		9	2	6	343
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		15	16	17	270
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		2	37	34	138
		3	42	41	183
		4	27	27	258
		5	11	8	133
		6	22	20	103
		7	33	39	320
		8	22	22	224
		9	28	22	187
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		13	23	22	212
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		3	20	20	304
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		6	32	29	68
		7	12	13	213
		8	12	14	324
		9	20	20	319
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		12	10	11	6
		13	8	10	322
		14	14	14	331
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		5	14	14	238
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		16	7	9	191
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		2	12	12	337
		3	33	33	257
		4	8	9	169
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		4	32	30	154
		5	29	28	272
		6	14	14	231
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		8	9	9	121
		9	23	25	300
		10	18	17	188
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		13	17	19	325
		14	13	13	159
		15	11	14	242
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		2	15	18	238
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		11	12	12	31
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		13	7	9	142
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		3	8	7	149
		4	15	14	210
		5	19	16	296
		6	7	8	97
		10	14	13	180
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		14	7	9	170
8	8	0	12	13	180
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		2	15	20	178
		3	21	25	86
		4	16	20	161
		5	7	8	303
		6	17	19	198
		7	19	24	80
		8	17	20	173
		10	8	10	219
		11	6	6	51
		12	14	18	187
		13	8	11	102
8	9	1	8	9	90
		3	8	9	142
		4	17	16	326
		5	14	16	71
		6	7	9	77
		7	8	10	15
		8	6	4	326
		9	15	17	117
		11	11	14	16
8	10	0	7	8	180
		1	10	13	149
		2	8	11	125
		3	7	7	74
		4	20	23	243
		6	9	13	117
		7	7	6	105
		8	9	9	251
		9	8	8	47
		10	7	6	173
8	11	0	13	16	0
		1	10	14	123
		3	13	12	60
		4	16	18	18
		5	16	16	99
		7	12	17	82
		10	8	12	358
8	12	2	8	9	38
		3	7	11	280
		4	8	9	268
		5	6	8	49
		7	6	9	238
		8	5	6	335
		9	5	4	180
		10	17	16	142
		11	18	14	270
8	13	0	5	4	180
		1	8	12	142
		2	6	9	347
		3	6	8	74
		6	5	8	303
8	14	0	4	9	0
9	0	2	23	20	180
		4	39	39	0
		5	9	10	90
		6	38	33	180
		7	12	8	270
		8	8	8	0
		10	17	16	180
		11	18	14	270
9	1	0	10	10	90
9	2	0	25	29	270
9	3	0	17	20	270
9	4	0	13	13	270
9	5	0	16	15	270
9	7	0	17	17	90
9	9	0	15	14	90
9	11	0	8	7	270
10	0	0	33	33	0
		3	42	39	90
		4	24	23	0
		6	11	7	0
		7	33	32	90
		8	7	8	0
		10	10	12	0
		12	6	6	0
		14	7	7	0
10	1	0	25	24	0
10	2	0	16	18	0
10	3	0	31	33	0
10	7	0	13	12	180
11	0	2	10	10	180
		4	20	18	0
		6	24	21	180
		8	20	20	0
11	1	0	23	30	90
11	3	0	10	13	270
11	5	0	20	27	270
12	0	0	9	6	180
		2	7	9	180
		3	19	20	90
		4	9	9	180
12	1	0	13	13	0
12	3	0	18	21	0

nitrogen and those of James and Brindley [26] for bromine and hydrogen. The peaks representing the oxygen atoms of the water molecules were rather lower than would have been expected in the various Fourier syntheses and corresponded in height approximately to the carbon atoms of the calycanthine molecule. It was accordingly assumed that only 75% of the water sites were occupied and carbon scattering factors were used for the oxygen contributions. During the Fourier refinement an isotropic temperature factor $B = 4{\cdot}0$ Å^2 was used for all the atoms. The least-squares programme refined six vibrational parameters for each atom, the anisotropic temperature factor being of the form [27]

$$t = 2^{-(\beta_{11}h^2 + \beta_{22}k^2 + \beta_{33}l^2 + \beta_{23}kl + \beta_{31}lh + \beta_{12}hk)}$$

The final co-ordinates, anisotropic temperature parameters, and equivalent isotropic temperature factors are listed in Table 2. The weighting scheme used for the least squares analysis was as follows:

$$\sqrt{w} = |F_o|/|F^*| \text{ if } |F_o| < |F^*|; \quad \sqrt{w} = |F^*|/|F_o| \text{ if } |F_o| > |F^*|$$

where $|F^*|$ is a constant. It was taken equal to the average value of $|F_o|$ (about 27).

58 Accidentally absent reflections, for which $|F_c| > 1\frac{1}{2}|F_{min.}|$, where $|F_{min.}|$ is the minimum observable value of the structure amplitude, were included in the last cycle. These reflections were given a weight $w = 1$. The final discrepancy, $\sum||F_o| - |F_c||/\sum|F_o|$, over the 2116 observed reflections is 9·6%. Of the 432 unobserved reflections only 29 are calculated to be greater than $1\frac{1}{2}|F_{min.}|$. The parameter shifts indicated by the third least-squares cycle were quite small and it was felt that further refinement would not be profitable with the data available.

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[26] James and Brindley, *Phil. Mag.*, 1932, **12**, 81.
[27] Rollett, "Computing Methods and the Phase Problem in *X*-Ray Crystal Analysis," ed. Pepinsky, Robertson, and Speakman, Pergamon Press, Oxford, 1961, p. 87.