1181. *The Structure of Hunterburnine: X-Ray Analysis of Hunterburnine β-Methiodide* [1]

By J. D. M. Asher, J. Monteath Robertson, and G. A. Sim

X-Ray analysis of hunterburnine β-methiodide, a quaternary alkaloid isolated from *Hunteria eburnea* Pichon, has defined the constitution and stereochemistry as (II) and revealed hunterburnine as the first example of a new class of indole alkaloid. Crystals of the methiodide are orthorhombic, space group $P2_12_12_1$, with four molecules of $C_{20}H_{27}IN_2O_2$ in a unit cell of dimensions $a = 10{\cdot}96$, $b = 18{\cdot}83$, $c = 9{\cdot}20$ Å. The crystal structure was solved by the heavy-atom method and the atomic co-ordinates were refined by Fourier and least-squares calculations. The final value of R over 1622 independent reflexions was 15·4%.

A number of alkaloids, both tertiary [2] and quaternary,[3] have been isolated from *Hunteria eburnea* Pichon. By degradative studies and partial syntheses three of the quaternary alkaloids were shown to be akuammicine methochloride, yohimbol methochloride and dihydrocorynantheol methochloride (I).

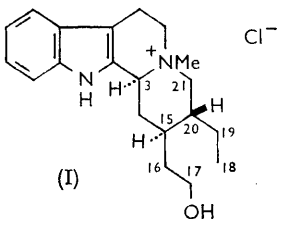
[1] For a preliminary communication see J. D. M. Asher, J. M. Robertson, G. A. Sim, M. F. Bartlett, R. Sklar, and W. I. Taylor, *Proc. Chem. Soc.*, 1962, 72.

[2] M. F. Bartlett and W. I. Taylor, *J. Amer. Chem. Soc.*, 1960, **82**, 5941; M. F. Bartlett, R. Sklar, A. F. Smith, and W. I. Taylor, *J. Org. Chem.*, 1963, **28**, 2197.

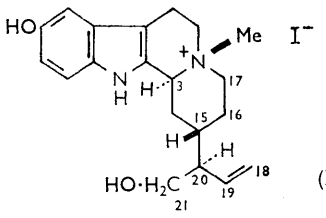
[3] M. F. Bartlett, B. Korzun, R. Sklar, A. F. Smith, and W. I. Taylor, *J. Org. Chem.*, 1963, **28**, 1445.

Among the quaternary bases is a group of isomeric compounds $C_{20}H_{27}ClN_2O_2$, all of which contain a 5-hydroxyindole chromophore, an isolated double bond and an aliphatic hydroxyl group which is readily acetylated. Because of lack of material detailed structural investigation by degradative methods was not feasible and the *X*-ray analysis approach was deemed imperative.

One of the compounds, hunterburnine methochloride, m. p. 307—308°, was converted into a crystalline methiodide, m. p. 277—280°, by Dr. W. I. Taylor and his colleagues of the CIBA Pharmaceutical Company, whom we thank. The crystal structure of the methiodide was elucidated by the usual heavy-atom method.[4] Several three-dimensional electron-density distributions were evaluated, yielding approximate positions for all atoms, other than hydrogen, in the unit cell. The atomic co-ordinates and temperature factors were then further refined by least-squares calculations and the average discrepancy between calculated and measured structure amplitudes decreased to a final value of 15·4% over 1622 independent reflexions.



(I)



(II)

Our final results establish the constitution and relative stereochemistry of the methiodide to be as in (II). The assignment of the double bond between C(18) and C(19) was based on the bond length and was confirmed by a nuclear magnetic resonance study by the CIBA group. Subsequent *X*-ray analysis of a further member of the series of isomeric quaternary alkaloids showed it to be the *N*(*b*)-epimer of (II);[5] it became necessary, therefore, to describe (II) as hunterburnine β-methiodide and the *N*(*b*)-epimer as the α-methiodide. Though synthetic *N*(*b*)-epimeric yohimban methiodides are known,[6] it is noteworthy that hunterburnine α- and β-methiodides constitute the first recognised pair of naturally occurring *N*(*b*)-epimeric quaternary alkaloids. There is an interesting pharmacological difference between the compounds, the α-methiodide inducing a marked lowering of blood pressure in the anaesthetised dog.[7]

Structure (II) is novel and hunterburnine β-methiodide is the first representative of a new class of indole alkaloid. The biogenetic relationship to other indole alkaloids becomes clear when it is realised that the skeleton of hunterburnine methiodide can be derived from that of dihydrocorynantheol methochloride (I) (also isolated from *Hunteria eburnea*) by an appropriate scission and recyclisation as indicated by the numbering in the formulæ. The absolute stereochemistry shown is based on the assumption that the rule [8] of uniform absolute stereochemistry of the C(15) equivalent of yohimbine [C(15) in (I) and (II)] is valid in this case. ψ-Akuammicine [9] appears to be an exception to this rule.

The final electron-density distribution for hunterburnine β-methiodide is shown in Figure 1 as superimposed contour sections drawn parallel to (001) and covering the region of one molecule. The atomic arrangement corresponding to this electron-density distribution is explained in Figure 2. The final atomic co-ordinates are listed in Table 1 and the

[4] J. M. Robertson and I. Woodward, *J.*, 1937, 219; 1940, 36; G. A. Sim, in "Computing Methods and the Phase Problem in *X*-ray Crystal Analysis," ed. R. Pepinsky, J. M. Robertson, and J. C. Speakman, Pergamon Press, Oxford, 1961, p. 227.

[5] C. C. Scott, G. A. Sim, and J. M. Robertson, *Proc. Chem. Soc.*, 1962, 355.

[6] B. Witkop and S. Goodwin, *J. Amer. Chem. Soc.*, 1953, **75**, 3371.

[7] Personal communication from Dr. W. I. Taylor.

[8] E. Wenkert and N. V. Bringi, *J. Amer. Chem. Soc.*, 1959, **81**, 1474, 6535.

[9] P. N. Edwards and G. F. Smith, *Proc. Chem. Soc.*, 1960, 215.

interatomic distances and valency angles in Table 2. The standard deviations of the final atomic co-ordinates were estimated in the usual manner from the least-squares residuals (see Experimental section) and are given in Table 3; from the results the average estimated standard deviation (e.s.d.) of a carbon–carbon or carbon–oxygen bond length is about 0·05 Å and the average e.s.d. of a valency angle about 3°.

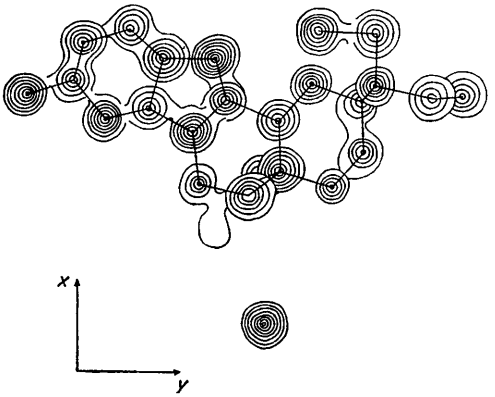


FIGURE 1. Final three-dimensional electron-density distribution for hunterburnine β-methiodide, shown by means of superimposed contour sections drawn parallel to (001). Contours are at an interval of 7e/Å³ for the iodide ion and 1e/Å³ for the other atoms

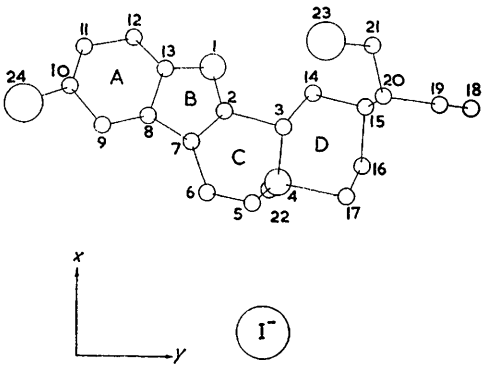


FIGURE 2. Atomic arrangement corresponding to Figure 1

TABLE 1

Atomic co-ordinates

(Origin of co-ordinates as in " International Tables " *)

Atom	*x/a*	*y/b*	*z/c*	Atom	*x/a*	*y/b*	*z/c*
N(1)	0·8259	−0·0522	0·1763	C(13)	0·8274	−0·1312	0·1623
C(2)	0·7180	−0·0311	0·1392	C(14)	0·7663	0·1015	0·2492
C(3)	0·6798	0·0541	0·1397	C(15)	0·7237	0·1764	0·2116
N(4)	0·5446	0·0560	0·1726	C(16)	0·5833	0·1809	0·2387
C(5)	0·4756	0·0107	0·0597	C(17)	0·5051	0·1360	0·1667
C(6)	0·4951	−0·0678	0·0867	C(18)	0·7490	0·3298	0·1722
C(7)	0·6347	−0·0776	0·1195	C(19)	0·7482	0·2923	0·0660
C(8)	0·7008	−0·1499	0·1147	C(20)	0·7679	0·2065	0·0582
C(9)	0·6679	−0·2153	0·1034	C(21)	0·9061	0·1945	0·0238
C(10)	0·7605	−0·2675	0·1277	C(22)	0·5176	0·0317	0·3401
C(11)	0·8765	−0·2466	0·1508	O(23)	0·9134	0·1184	−0·0264
C(12)	0·9158	−0·1793	0·1878	O(24)	0·7213	−0·3397	0·1020
				I	0·1278	0·0357	0·1841

* " International Tables for X-Ray Crystallography," The Kynoch Press, Birmingham, 1952. vol. I.

The average carbon–carbon bond length in the benzene ring, 1·38 Å, and the average carbon–carbon single bond length, 1·56 Å, do not differ significantly from the values of 1·397 Å in benzene and 1·545 Å in diamond, respectively.[10] The C(2)–C(7) double bond length, 1·28 Å, is not significantly shorter than the value of 1·334 Å in ethylene.[11] The C(18)–C(19) double bond length of 1·21 Å is very short, but the adjacent single bond C(19)–C(20) is rather long, 1·63 Å, and it is possible that C(19) has been misplaced by about 0·1 Å from its true position by the least-squares refinement procedure.

[10] L. E. Sutton *et al.*, " Tables of Interatomic Distances and Configuration in Molecules and Ions," *Chem. Soc. Special Publ.* No. 11, 1958.

[11] L. S. Bartell and R. A. Bonham, *J. Chem. Phys.*, 1957, **27**, 1414.

TABLE 2

Interatomic distances (Å) and angles

Intramolecular bonded distances

N(1)–C(2)	1·29	N(4)–C(17)	1·57	C(9)–C(10)	1·43	C(15)–C(20)	1·60
N(1)–C(13)	1·49	N(4)–C(22)	1·64	C(10)–C(11)	1·35	C(16)–C(17)	1·37
C(2)–C(3)	1·66	C(5)–C(6)	1·51	C(10)–O(24)	1·45	C(18)–C(19)	1·21
C(2)–C(7)	1·28	C(6)–C(7)	1·57	C(11)–C(12)	1·38	C(19)–C(20)	1·63
C(3)–N(4)	1·51	C(7)–C(8)	1·54	C(12)–C(13)	1·35	C(20)–C(21)	1·56
C(3)–C(14)	1·65	C(8)–C(9)	1·29	C(14)–C(15)	1·53	C(21)–O(23)	1·51
N(4)–C(5)	1·54	C(8)–C(13)	1·50	C(15)–C(16)	1·56		

Intramolecular non-bonded distances

N(1) · · · N(4) ...	3·70	C(3) · · · C(8) ...	3·86	C(6) · · · C(13) ...	3·90	C(14) · · · C(21)...	3·12
N(1) · · · C(6) ...	3·73	C(3) · · · C(13) ...	3·85	C(6) · · · C(17) ...	3·90	C(14) · · · C(22)...	3·14
N(1) · · · C(9) ...	3·59	C(3) · · · C(16) ...	2·76	C(6) · · · C(22) ...	3·00	C(14) · · · O(23)...	3·02
N(1) · · · C(11) ...	3·71	C(3) · · · C(20) ...	3·12	C(7) · · · C(10) ...	3·83	C(15) · · · C(18)...	2·92
N(1) · · · C(14) ...	3·04	C(3) · · · C(21) ...	3·78	C(7) · · · C(12) ...	3·68	C(15) · · · C(22)...	3·73
N(1) · · · O(23) ...	3·84	C(3) · · · O(23) ...	3·22	C(7) · · · C(14) ...	3·86	C(15) · · · O(23)...	3·21
C(2) · · · C(5)	2·87	N(4) · · · C(7) ...	2·75	C(7) · · · C(22) ...	3·16	C(16) · · · C(18)...	3·40
C(2) · · · C(9)	3·53	N(4) · · · C(15) ...	3·02	C(8) · · · C(11) ...	2·67	C(16) · · · C(19)...	3·19
C(2) · · · C(12) ...	3·56	N(4) · · · C(20) ...	3·89	C(8) · · · O(24) ...	3·58	C(16) · · · C(22)...	3·05
C(2) · · · C(15) ...	3·97	C(5) · · · C(8) ...	3·94	C(9) · · · C(12) ...	2·91	C(17) · · · C(20)...	3·31
C(2) · · · C(17) ...	3·92	C(5) · · · C(16) ...	3·79	C(10) · · · C(13)...	2·69	C(18) · · · C(21)...	3·37
C(2) · · · C(22) ...	3·11	C(6) · · · C(9) ...	3·37	C(12) · · · O(24)...	3·78	C(19) · · · O(23)...	3·84
C(2) · · · O(23) ...	3·85			C(14) · · · C(17)...	3·05		
C(3) · · · C(6)	3·10			C(14) · · · C(19)...	3·97		

Intermolecular distances (<4 Å)

C(22) · · · O(23)$_{I}$	3·17	C(22) · · · O(24)$_{II}$	3·61	C(6) · · · O(23)$_{I}$	3·82	C(17) · · · C(10)$_{II}$	3·95
C(17) · · · O(24)$_{II}$	3·34	C(16) · · · O(24)$_{II}$	3·67	C(14) · · · C(2)$_{I}$	3·83	C(14) · · · C(9)$_{I}$	3·97
C(7) · · · O(23)$_{I}$	3·39	C(16) · · · C(9)$_{II}$	3·68	C(15) · · · C(8)$_{I}$	3·84	C(9) · · · O(23)$_{I}$	3·97
O(23) · · · I$_{III}$	3·42	N(1) · · · I$_{III}$	3·70	C(21) · · · C(19)$_{V}$	3·85	C(9) · · · C(21)$_{I}$	3·97
I · · · O(24)$_{II}$	3·48	I · · · C(5)$_{IV}$	3·74	C(15) · · · C(9)$_{I}$	3·87	C(18) · · · O(24)$_{I}$	3·97
C(14) · · · C(8)$_{I}$	3·50	C(19) · · · C(17)$_{V}$	3·76	O(24) · · · C(6)$_{VI}$	3·88	C(18) · · · C(6)$_{II}$	3·98
C(22) · · · N(1)$_{I}$	3·56	C(22) · · · I$_{IV}$	3·77	C(21) · · · C(16)$_{V}$	3·89	C(14) · · · C(13)$_{I}$	3·98
C(8) · · · O(23)$_{I}$	3·58	C(21) · · · C(17)$_{V}$	3·77	C(22) · · · C(13)$_{I}$	3·90	I · · · C(6)$_{IV}$	3·99
C(14) · · · C(7)$_{I}$	3·61	C(2) · · · O(23)$_{I}$	3·78	C(13) · · · O(23)$_{I}$	3·90		

The subscripts refer to the following positions:

I	$1\frac{1}{2} - x, -y, \frac{1}{2} + z$	IV	$\frac{1}{2} - x, -y, \frac{1}{2} + z$
II	$1 - x, \frac{1}{2} + y, \frac{1}{2} - z$	V	$\frac{1}{2} + x, \frac{1}{2} - y, -z$
III	$1 + x, y, z$	VI	$\frac{1}{2} + x, -\frac{1}{2} - y, -z$

Interbond angles

C(2)N(1)C(13)	107°	C(5)N(4)C(17)	110°	C(8)C(9)C(10)	117°	C(14)C(15)C(16)	108°
N(1)C(2)C(3)	122	C(5)N(4)C(22)	113	C(9)C(10)C(11)	120	C(14)C(15)C(20)	116
N(1)C(2)C(7)	119	C(17)N(4)C(22)	107	C(9)C(10)O(24)	114	C(16)C(15)C(20)	115
C(3)C(2)C(7)	119	N(4)C(5)C(6)	111	C(11)C(10)O(24)	126	C(15)C(16)C(17)	119
C(2)C(3)N(4)	106	C(5)C(6)C(7)	107	C(10)C(11)C(12)	127	N(4)C(17)C(16)	112
C(2)C(3)C(14)	115	C(2)C(7)C(6)	130	C(11)C(12)C(13)	110	C(18)C(19)C(20)	128
N(4)C(3)C(14)	112	C(2)C(7)C(8)	106	N(1)C(13)C(8)	104	C(15)C(20)C(19)	106
C(3)N(4)C(5)	109	C(6)C(7)C(8)	124	N(1)C(13)C(12)	131	C(15)C(20)C(21)	115
C(3)N(4)C(17)	106	C(7)C(8)C(9)	135	C(8)C(13)C(12)	124	C(19)C(20)C(21)	106
C(3)N(4)C(22)	111	C(7)C(8)C(13)	103	C(3)C(14)C(15)	101	C(20)C(21)O(23)	105
		C(9)C(8)C(13)	121				

The carbon–nitrogen bonds fall into two categories, C(sp^2)–N and C(sp^3)–N^+. The average C(sp^2)–N bond length of 1·39 Å is in good agreement with values reported for such bonds in, *e.g.*, *p*-nitroaniline (1·371 Å),[12] 2-chloro-4-nitroaniline (1·386 Å),[13] and macusine-A iodide (1·33 Å).[14] The average C(sp^3)–N^+ bond length, 1·57 Å, is rather longer than might have been expected, but long C(sp^3)–N^+ bonds have been reported in a number of alkaloid salts; for a discussion see Hamilton *et al.*[15]

[12] K. N. Trueblood, E. Goldish, and J. Donohue, *Acta Cryst.*, 1961, **14**, 1009.
[13] A. T. McPhail and G. A. Sim, *J.*, 1965, 227.
[14] A. T. McPhail, J. M. Robertson, and G. A. Sim, *J.*, 1963, 1832.
[15] J. A. Hamilton, T. A. Hamor, J. M. Robertson, and G. A. Sim, *J.*, 1962, 5061.

TABLE 3

Standard deviations of the final atomic co-ordinates (Å)

Atom	$\sigma(x)$	$\sigma(y)$	$\sigma(z)$	Atom	$\sigma(x)$	$\sigma(y)$	$\sigma(z)$
N(1)	0·031	0·034	0·030	C(13)	0·041	0·034	0·045
C(2)	0·029	0·025	0·034	C(14)	0·029	0·036	0·033
C(3)	0·024	0·026	0·037	C(15)	0·038	0·029	0·037
N(4)	0·023	0·025	0·034	C(16)	0·044	0·031	0·034
C(5)	0·037	0·039	0·042	C(17)	0·033	0·033	0·041
C(6)	0·032	0·028	0·041	C(18)	0·030	0·039	0·036
C(7)	0·048	0·045	0·057	C(19)	0·046	0·037	0·038
C(8)	0·043	0·038	0·037	C(20)	0·038	0·046	0·033
C(9)	0·034	0·032	0·044	C(21)	0·040	0·034	0·034
C(10)	0·038	0·042	0·042	C(22)	0·045	0·033	0·037
C(11)	0·037	0·032	0·034	O(23)	0·044	0·036	0·041
C(12)	0·041	0·043	0·048	O(24)	0·042	0·038	0·040
				I	0·009	0·002	0·003

The arrangement of the alkaloid cations and the iodide anions in the crystal as viewed in projections along the *a* axis is shown in Figure 3. In addition to the normal ionic and van der Waals forces, the crystal lattice is stabilised by a system of hydrogen bonds involving the hydroxyl groups O(23) and O(24), the indole nitrogen atom N(1) and the iodide ion. The OH · · · I distances, 3·42 and 3·48 Å, are similar to hydrogen-bonded distances in macusine-A iodide, 3·43 Å,[14] and (+)-de(oxymethylene)lycoctonine hydriodide monohydrate, 3·52—3·62 Å,[16] and the angles C(2)O(23)I and C(10)O(24)I, 107° and 114°, respectively, are close to tetrahedral. The angles C(2)N(1)I and C(13)N(1)I are 133 and 116°, respectively, and the N(1)H · · · I distance is 3·70 Å.

The closest contacts between an iodide ion and carbon atoms, 3·74, 3·77, and 3·99 Å (see Table 2) are similar to the minimum C · · · I distances of 3·93, 3·95 Å in macusine-A iodide,[14] 3·81 Å in (+)-de(oxymethylene)lycoctonine hydriodide monohydrate,[16] and 3·96 Å in isocryptopleurine methiodide.[17] Of the carbon · · · carbon intermolecular contacts sixteen are less than 4 Å, ranging from 3·50 to 3·98 Å. There are three short van der Waals contacts between carbon and oxygen of 3·17, 3·34, and 3·39 Å; this appears to be a normal occurrence, as is evidenced by values of 3·18 Å in calycanthine dihydrobromide dihydrate,[18] 3·2 Å in (+)-demethanolaconinone hydriodide trihydrate [19] and 3·2 Å in (−)-aspidospermine methiodide.[20]

EXPERIMENTAL

Crystal Data.—Hunterburnine β-methiodide, $C_{20}H_{27}IN_2O_2$; $M = 454·3$; m. p. 277—280°. Orthorhombic, $a = 10·96$, $b = 18·83$, $c = 9·20$ Å, $U = 1899$ Å^3, $Z = 4$, $D_c = 1·589$, $D_m = 1·602$ g. cm.$^{-3}$, $F(000) = 920$, space group $P2_12_12_1$ (D_2^4). Absorption coefficient for *X*-rays ($\lambda = 1·542$ Å), $\mu = 136·5$ cm.$^{-1}$.

Crystallographic Measurements.—Rotation, oscillation, and Weissenberg photographs were taken with copper K_α ($\lambda = 1·542$ Å) radiation; precession photographs were taken with molybdenum K_α ($\lambda = 0·7107$ Å) radiation. The unit-cell dimensions were obtained from precession photographs. The space group was uniquely assigned from the systematic halvings in the *X*-ray spectra. For the intensity measurements small crystals of about 0·1 mg. were employed completely bathed in a uniform *X*-ray beam, and no corrections for absorption were applied. The *hk*0–*hk*7 spectra were recorded photographically by means of an equi-inclination Weissenberg camera and the 0*kl* spectra were obtained with a precession camera. The intensities were estimated visually and were corrected by the usual factors (Lorentz, polarisation, and the rotation factor appropriate to upper layers [21]). The various layers were placed on the same relative

[16] M. Przybylska, *Acta Cryst.*, 1961, **14**, 424.
[17] J. Fridrichsons and A. McL. Mathieson, *Acta Cryst.*, 1955, **8**, 761.
[18] T. A. Hamor and J. M. Robertson, *J.*, 1962, 194.
[19] M. Przybylska, *Acta Cryst.*, 1961, **14**, 429.
[20] J. F. D. Mills and S. C. Nyburg, *J.*, 1960, 1458.
[21] G. Tunell, *Amer. Min.*, 1939, **24**, 448.

TABLE 4

Measured and calculated values of the structure facors.

h	k	l	$\|F_o\|$	$\|F_c\|$	α°	h	k	l	$\|F_o\|$	$\|F_c\|$	α°	h	k	l	$\|F_o\|$	$\|F_c\|$	α°	h	k	l	$\|F_o\|$	$\|F_c\|$	α°
0	1	2	113	251	90			7	40	41	180			5	32	40	270			5	27	27	128
		3	44	40	90	0	11	1	76	65	90			6	31	54	180			6	35	24	202
		4	96	156	270			2	88	75	270			7	31	49	90			7	37	33	280
		5	72	68	90			3	34	27	270	1	1	0	57	4	90	1	10	0	61	61	90
		6	32	48	90			4	74	71	90			1	46	54	299			1	54	48	226
		7	19	21	270			5	19	21	90			2	46	47	269			2	52	48	273
0	2	0	135	143	0			6	7	7	270			3	120	87	213			3	52	57	10
		1	73	97	180			7	15	12	270			4	23	23	181			4	28	23	153
		2	141	126	180	0	12	0	120	93	180			5	75	100	32			5	35	34	171
		3	89	79	0			1	45	46	180			6	22	38	89			6	42	37	40
		4	25	31	180			2	39	35	0			7	7	3	234			7	22	20	308
		5	6	2	0			3	21	20	0	1	2	0	34	32	90	1	11	0	51	49	270
		6	54	68	0			4	14	5	0			1	59	66	124			1	37	34	238
		7	20	16	180			5	13	10	180			2	144	137	163			2	58	53	131
0	3	1	37	35	90			6	45	47	180			3	153	113	325			3	40	43	39
		2	121	110	90	0	13	1	14	6	270			4	57	64	9			4	56	53	342
		3	94	92	270			2	61	59	270			5	23	28	222			5	38	35	145
		4	90	75	270			3	11	11	270			6	28	43	174			6	19	19	215
		5	90	67	90			4	39	60	90			7	21	23	110			7	26	27	286
		6	33	29	90			5	12	15	90	1	3	0	113	121	90	1	12	0	24	28	90
		7	6	0	270			6	19	21	270			1	87	73	266			1	44	47	258
0	4	0	121	103	0	0	14	0	81	75	180			2	120	82	231			2	44	45	336
		1	125	122	180			2	67	72	0			3	128	104	181			3	31	32	26
		2	54	49	180			4	11	11	180			4	72	59	33			4	59	56	177
		3	53	49	0			5	13	10	180			5	66	47	15			5	12	16	157
		4	35	29	180			6	39	35	180			6	32	28	107			6	27	26	46
		5	95	68	0	0	15	1	14	17	270			7	36	42	261			7	23	18	278
		6	42	37	0			2	18	19	270	1	4	0	78	95	90	1	13	0	30	28	270
		7	69	62	180			3	19	16	90			1	99	91	127			1	47	41	185
0	5	1	38	32	90			4	81	56	90			2	96	80	238			2	34	38	104
		2	42	41	90			5	7	2	270			3	94	70	343			3	48	46	339
		3	94	94	270			6	32	27	270			4	90	67	358			4	21	21	9
		4	36	25	270	0	16	0	46	46	180			5	82	53	195			5	30	32	170
		5	97	64	90			1	33	30	0			6	37	25	150			6	21	23	268
		6	57	49	90			2	30	26	0			7	34	28	70			7	12	10	309
		7	25	22	270			3	7	4	180	1	5	0	67	68	90	1	14	1	57	55	268
0	6	0	75	70	0			4	8	10	0			1	123	124	281			2	50	43	338
		1	69	140	180			5	16	9	180			2	98	92	207			4	17	24	197
		2	35	33	180	0	17	1	13	6	270			3	87	74	138			5	19	19	95
		3	36	35	0			2	26	25	270			4	83	73	355			6	10	15	5
		4	18	11	0			3	29	30	90			5	58	44	55			7	23	23	250
		5	50	40	0			4	15	15	90			6	43	36	149	1	15	0	58	57	270
		6	18	20	0			5	18	17	270			7	48	37	270			1	21	24	187
		7	81	71	180	0	18	0	23	20	180	1	6	0	84	81	90			2	19	19	117
0	7	1	72	65	90			1	29	31	0			1	47	51	163			3	46	47	357
		2	41	35	270			2	16	12	0			2	55	54	265			4	17	12	93
		3	126	116	270			3	7	9	180			3	68	86	1			5	22	23	189
		4	8	13	270			5	12	15	180			4	28	23	304			6	23	27	277
		5	80	81	90	0	19	1	13	17	270			5	68	55	186			7	20	13	52
		6	6	6	90			2	14	10	270			6	42	31	78	1	16	1	36	32	273
		7	16	19	270			3	23	25	90			7	26	18	48			2	20	19	24
0	8	0	95	87	180			4	15	18	90	1	7	0	45	40	90			3	19	17	123
		1	104	127	180			5	16	16	270			1	78	80	278			4	38	39	178
		2	43	41	0	0	20	0	13	5	180			2	77	75	178			5	17	19	16
		3	14	13	0			1	35	30	0			3	32	40	109			6	18	19	336
		5	33	29	0			2	5	12	0			4	64	60	351			7	9	11	271
		6	12	10	180			3	13	9	180			5	31	27	96	1	17	0	21	26	270
		7	64	50	180			5	6	11	180			6	34	24	195			1	15	22	118
0	9	1	14	18	90	0	21	2	8	2	90			7	41	30	276			2	30	34	58
		2	62	61	270			3	20	27	90	1	8	0	23	30	90			3	24	23	5
		3	56	66	270			5	8	21	270			1	56	40	182			4	17	21	196
		4	64	64	90	0	22	1	14	16	0			2	66	57	255			5	13	16	200
		5	60	57	90			2	10	15	180			3	89	84	359			6	18	17	288
		6	22	22	270			3	11	13	180			4	26	12	157			7	8	11	72
		7	23	15	270	0	23	2	7	14	90			5	67	59	184	1	18	0	26	31	270
0	10	0	107	95	180			3	7	10	90			6	31	28	117			1	17	22	298
		1	69	76	180	0	24	0	8	14	0			7	33	21	268			2	31	27	46
		2	80	71	0			1	8	10	0	1	9	0	41	33	270			3	18	21	174
		3	23	15	0	1	0	1	26	44	90			1	117	115	267			4	9	20	182
		4	57	57	0			2	116	126	0			2	29	33	169			5	13	11	22
		5	43	39	0			3	79	77	270			3	40	33	19			6	11	12	301
		6	55	56	180			4	52	57	0			4	76	66	0	1	19	1	21	24	103

TABLE 4 (*Continued*)

h	*k*	*l*	$\|F_o\|$	$\|F_c\|$	α°
		2	20	18	21
		3	17	15	320
		4	21	19	180
		5	12	15	234
		6	10	16	2
1	20	0	18	18	270
		1	7	11	352
		2	10	11	65
		3	21	20	170
		5	10	13	8
		6	4	10	280
1	21	0	7	2	270
		1	23	20	81
		2	15	15	16
		3	5	1	257
		4	13	15	202
1	22	0	19	20	270
		1	11	12	9
		2	10	12	116
		3	16	16	174
1	23	1	16	17	90
		2	11	15	347
		3	5	7	228
1	24	0	13	17	270
		1	6	8	83
2	0	0	67	98	180
		1	45	52	90
		2	22	26	0
		3	111	161	270
		4	35	34	0
		5	52	54	90
		6	15	12	0
		7	20	24	270
2	1	0	23	47	180
		1	140	183	173
		2	101	102	358
		3	38	32	330
		4	35	24	70
		5	67	52	345
		6	35	29	165
		7	51	57	162
2	2	0	80	92	0
		1	84	87	103
		2	68	84	294
		3	120	134	269
		4	103	84	108
		5	116	92	66
		6	47	37	257
		7	18	14	180
2	3	0	100	109	180
		1	124	133	171
		2	85	85	334
		3	32	27	352
		4	36	33	65
		5	70	62	24
		6	67	47	172
		7	64	58	186
2	4	0	9	1	0
		1	50	45	111
		2	119	104	258
		3	60	70	275
		4	50	82	86
		5	88	64	79
		6	50	37	259
		7	20	11	227
2	5	0	145	178	180
		1	65	69	202
		2	83	77	3
		3	51	42	349
		4	27	21	29
		5	33	28	23
		6	95	64	175
		7	19	13	180
2	6	0	19	13	180
		1	7	15	140
		2	94	93	271
		3	33	24	285
		4	125	124	86
		5	26	18	118
		6	55	41	265
		7	13	8	238
2	7	0	161	167	180
		1	23	22	331
		2	100	108	4
		3	27	24	51
		4	15	3	117
		5	17	19	214
		6	64	53	178
2	8	0	17	17	0
		1	49	46	226
		2	116	106	275
		3	45	36	90
		4	108	96	87
		5	18	11	321
		6	39	37	278
2	9	0	108	123	180
		1	65	65	37
		2	61	58	353
		3	29	21	207
		4	20	16	349
		5	24	22	193
		6	50	45	180
		7	19	14	342
2	10	1	55	44	247
		2	49	47	280
		3	52	48	106
		4	55	55	106
		5	39	36	276
		6	29	26	272
2	11	0	30	34	180
		1	14	68	355
		2	45	44	13
		5	30	27	164
		6	12	17	175
		7	31	33	3
2	12	1	25	26	287
		2	20	23	296
		3	65	72	106
		4	23	24	92
		5	41	38	273
		6	12	15	238
2	13	0	18	20	180
		1	13	65	4
		2	26	24	67
		3	38	32	139
		4	8	9	154
		5	25	28	227
		7	28	34	356
2	14	1	13	19	303
		2	23	18	136
		3	47	47	94
		5	37	42	273
		7	10	11	107
2	15	0	45	42	0
		1	48	53	4
		2	24	19	174
		3	16	18	210
		4	24	19	195
		5	16	16	185
		7	23	27	3
2	16	0	23	25	0
		1	35	34	251
		2	32	34	86
		3	42	43	92
		4	27	21	251
		5	24	28	268
		6	11	8	120
2	17	0	26	27	0
		1	35	36	357
		2	21	18	136
		3	7	7	172
		5	14	16	151
		6	19	18	357
2	18	1	7	5	260
		2	20	18	83
		3	19	18	79
		4	25	22	272
		5	10	15	249
2	19	0	35	31	0
		1	13	15	6
		2	19	21	183
2	20	2	19	21	96
		3	6	9	71
		4	23	25	263
2	21	0	28	28	0
		2	17	17	175
2	22	2	13	10	77
2	23	0	11	10	0
		2	11	11	199
3	0	1	121	153	270
		2	108	137	180
		3	52	52	90
		4	49	50	0
		5	52	46	90
		6	30	29	180
		7	74	71	270
3	1	0	67	81	90
		1	67	79	212
		2	87	87	289
		3	75	93	8
		5	78	73	181
		6	33	24	85
		7	37	35	308
3	2	0	56	65	270
		1	108	128	271
		2	62	59	151
		3	54	56	22
		4	74	78	5
		5	32	24	110
		6	36	37	205
		7	49	45	287
3	3	0	136	161	90
		1	69	62	235
		2	48	56	314
		3	46	59	22
		4	57	55	183
		5	51	46	174
		6	39	41	65
		7	27	26	337
3	4	0	65	77	270
		1	76	74	250
		2	84	82	124
		3	14	45	1
		4	46	44	2
		5	46	40	159
		6	37	43	251
		7	31	24	270
3	5	0	11	20	90
		1	73	66	259
		2	28	34	1
		3	11	17	32
		4	68	64	186
		5	16	24	148
		6	41	41	25
		7	30	22	285
3	6	0	105	112	270
		1	54	47	190
		2	67	67	131
		3	75	65	4
		4	24	25	21
		5	49	47	171
		6	41	43	268
		7	17	15	298
3	7	0	15	14	90
		1	71	64	257
		2	72	67	354
		3	31	30	72
		4	58	54	188
		5	12	34	82
		6	35	27	359
		7	50	38	263
3	8	0	83	77	270
		1	27	30	176
		2	49	55	84
		3	58	49	346
		4	17	17	163
		5	43	47	177
		6	41	45	278
		7	19	18	32
3	9	0	15	11	270
		1	45	40	298
		2	51	46	2
		3	43	44	136
		4	47	46	180
		5	19	23	37
		6	32	29	334
		7	32	32	265
3	10	0	93	90	270
		1	60	55	102
		2	46	45	48
		3	48	42	337
		4	33	34	164
		5	36	36	227
		6	21	19	274
		7	29	28	83
3	11	0	49	52	270
		1	42	45	292
		2	47	46	40
		3	55	54	180
		4	30	26	162
		5	40	39	347
		6	25	29	311
		7	12	11	265
3	12	0	37	36	270
		1	64	60	91
		2	41	43	20
		3	32	30	315
		4	33	32	181
		5	35	35	234
		6	21	19	325
		7	29	30	73
3	13	0	54	56	270
		1	37	30	316
		2	36	38	77
		3	49	49	158
		4	23	19	122
		5	34	32	10
		6	17	19	290
3	14	0	17	14	270
		1	45	47	104
		2	26	28	11
		3	9	11	262
		4	40	40	171
		5	18	20	265
		6	9	12	351
		7	13	17	80
3	15	0	32	37	270
		1	10	17	55

TABLE 4 (*Continued*)

h	k	l	$\|F_o\|$	$\|F_c\|$	α°
		2	33	32	62
		3	23	26	187
		4	14	10	307
		5	24	24	333
		6	16	17	245
		7	9	10	119
3	16	1	23	22	71
		2	31	37	344
		3	26	27	232
		4	22	26	175
		6	8	12	41
		7	10	20	82
3	17	0	35	35	270
		1	16	17	73
		2	21	27	134
		3	9	22	213
		4	7	14	10
		5	17	22	358
		6	14	14	261
		7	7	11	120
3	18	0	25	29	90
		1	15	23	66
		2	18	16	296
		3	14	12	195
		4	16	19	193
		5	14	15	325
3	19	0	9	3	270
		1	17	19	76
		2	19	17	166
		3	6	8	218
		4	17	18	14
		5	7	11	323
3	20	0	26	24	90
		2	15	15	275
		3	20	22	181
		5	12	18	11
3	21	1	14	16	99
		2	15	17	181
		4	14	18	353
3	22	0	14	23	90
		2	11	12	267
		3	12	10	175
3	23	2	9	13	186
4	0	0	55	74	180
		1	64	60	270
		2	82	84	0
		4	12	4	180
		5	20	13	90
		6	68	64	180
4	1	0	25	23	180
		1	6	7	172
		2	105	103	276
		3	26	18	356
		4	108	119	84
		5	12	6	247
		6	40	36	298
		7	12	6	179
4	2	0	99	123	180
		1	16	16	0
		2	74	73	0
		3	25	19	190
		4	15	15	319
		5	27	20	154
		6	48	40	183
		7	25	22	344
4	3	0	23	23	0
		1	54	55	275
		2	55	49	274
		3	65	63	84
		4	50	84	90
		5	47	46	278
		6	52	33	280
		7	12	15	106
4	4	0	75	80	180
		1	83	92	2
		2	70	82	13
		3	28	26	133
		4	9	12	278
		5	39	28	186
		6	43	30	188
		7	41	35	1
4	5	0	19	17	0
		1	44	46	305
		2	47	53	257
		3	113	111	87
		4	50	43	100
		5	68	68	270
		6	28	22	250
		7	15	14	29
4	6	0	8	8	0
		1	113	113	349
		2	16	18	351
		3	47	42	156
		4	24	22	190
		5	28	25	187
		6	7	9	89
		7	47	43	5
4	7	0	12	15	180
		1	53	46	269
		2	23	16	111
		3	103	97	95
		4	11	7	343
		5	63	52	275
		6	8	8	260
		7	10	2	291
4	8	0	25	24	0
		1	105	101	0
		2	25	29	130
		3	29	28	199
		5	34	31	189
		6	11	14	28
		7	30	30	6
4	9	0	16	14	0
		1	43	45	278
		2	40	39	106
		3	83	80	90
		4	34	30	283
		5	42	45	268
		6	16	18	80
		7	14	14	94
4	10	0	54	51	0
		1	59	55	0
		2	38	36	186
		3	28	30	185
		4	12	17	295
		5	19	21	204
		6	20	19	14
		7	27	21	7
4	11	0	14	11	0
		1	13	10	273
		2	45	43	84
		3	55	55	95
		4	45	45	272
		5	42	44	268
		6	25	22	69
		7	23	15	343
4	12	0	69	65	0
		1	34	34	359
		2	46	41	174
		5	13	22	185
		6	25	26	358
		7	11	16	11
		0	8	9	180
		1	12	13	274
		2	36	37	77
		6	23	33	106
4	14	0	61	64	0
		2	27	29	198
		4	7	8	115
		5	11	11	100
		6	24	25	10
4	15	0	14	16	180
		1	16	13	125
		2	53	46	95
		3	5	4	14
		4	33	42	282
		5	8	9	356
		6	11	18	92
4	16	0	26	28	0
		1	16	16	161
		2	25	27	192
		3	15	11	258
		5	7	10	335
		6	13	12	13
4	17	0	11	6	0
		1	15	15	102
		2	19	19	73
		3	27	30	272
		4	19	19	262
		5	12	14	84
		6	13	13	105
4	18	0	33	28	0
		1	28	25	183
		2	15	17	175
		5	12	19	16
4	19	1	12	11	117
		2	9	9	76
		3	27	22	274
		4	8	11	266
		5	16	20	86
4	20	1	23	24	178
4	21	0	7	1	0
		1	11	12	102
4	22	1	15	19	178
5	0	1	97	117	270
		2	68	73	0
		3	11	11	270
		4	76	73	180
		5	46	49	90
		6	43	43	0
		7	30	36	270
5	1	0	71	90	270
		1	89	84	137
		2	61	76	95
		3	55	58	356
		4	26	29	210
		5	36	38	182
		6	38	41	280
		7	10	17	68
5	2	0	56	62	270
		1	75	80	303
		2	31	36	23
		3	49	42	163
		4	86	75	178
		5	27	29	43
		6	43	40	306
		7	28	26	264
5	3	0	75	83	270
		1	87	77	106
		2	67	66	46
		3	53	52	329
		4	62	59	151
		5	33	34	244
		6	38	41	284
		7	26	21	62
5	4	0	71	83	270
		1	65	59	297
		2	70	66	56
		3	70	72	172
		4	48	42	162
		5	54	50	15
		6	40	34	300
		7	28	25	261
5	5	0	37	33	270
		1	100	84	102
		2	58	58	31
		3	42	37	343
		4	50	42	171
		5	36	38	229
		6	26	29	316
		7	32	34	84
5	6	0	56	57	270
		1	51	50	315
		2	57	58	54
		3	78	70	182
		4	25	25	185
		5	44	40	7
		6	34	32	281
		7	14	14	220
5	7	0	8	6	270
		1	77	77	89
		2	37	38	2
		3	31	22	261
		4	51	44	169
		5	22	25	260
		6	30	33	358
		7	34	38	83
5	8	0	111	86	270
		1	23	22	40
		2	46	50	115
		3	49	52	177
		4	15	15	18
		5	43	46	354
		6	25	30	236
		7	12	18	151
5	9	0	37	31	90
		1	42	45	83
		2	40	39	327
		3	41	37	228
		4	44	41	191
		5	31	32	290
		6	19	29	25
5	10	0	44	44	270
		1	52	52	67
		2	41	40	135
		3	44	40	176
		4	29	30	347
		5	31	33	329
		6	21	25	257
		7	8	8	87
5	11	0	40	34	90
		1	38	38	56
		2	27	28	324
		3	39	38	180
		4	20	19	195
		5	8	9	7
		6	19	23	69
5	12	0	16	18	270
		1	33	32	96
		2	26	30	189
		3	13	13	260
		4	24	22	6
		5	22	23	307
		6	12	15	190
5	13	0	56	65	90
		1	24	21	15
		2	27	27	254
		3	27	34	216

TABLE 4 (*Continued*)

h	k	l	$\|F_o\|$	$\|F_c\|$	α°
		4	14	17	319
		5	23	25	357
		6	13	20	70
		7	16	4	240
5	14	0	11	11	90
		1	37	35	97
		2	30	34	187
		3	17	9	273
		4	33	36	354
		5	20	20	248
		6	21	18	136
		7	9	22	96
5	15	0	60	42	90
		1	16	15	306
		2	32	32	247
		3	36	31	162
		4	8	6	345
		5	20	19	19
		6	12	14	86
5	16	0	17	15	90
		1	26	26	95
		2	31	29	192
		3	21	15	330
		4	18	25	352
		5	10	13	240
		6	11	8	150
5	17	0	26	26	90
		1	22	18	284
		2	16	14	243
		3	19	14	151
		4	14	17	342
		5	7	12	25
		6	8	15	114
5	18	0	15	16	90
		1	15	18	139
		2	15	19	235
		3	17	16	339
		4	6	9	355
5	19	0	17	14	90
		1	20	20	289
		2	14	14	209
		4	6	15	346
		5	9	12	27
5	20	0	17	16	90
		2	13	8	248
5	21	1	16	15	255
		2	10	11	185
5	22	2	10	11	274
6	0	0	10	4	0
		1	23	32	270
		2	3	14	180
		3	63	67	90
		4	27	34	180
		5	49	61	270
		6	10	15	0
		7	20	21	90
6	1	0	27	33	0
		1	80	94	344
		2	45	41	102
		3	24	27	204
		4	13	10	208
		5	16	12	195
		6	7	6	354
		7	37	39	5
6	2	0	68	65	0
		1	45	47	281
		2	26	24	103
		3	70	79	88
		4	35	28	256
		5	53	54	282
		6	12	17	51
		7	20	19	146
6	3	0	36	43	0
		1	70	68	0
		2	37	37	139
		3	32	33	170
		4	7	7	337
		5	20	16	175
		6	19	23	32
		7	12	33	350
6	4	0	14	13	0
		1	33	34	262
		2	47	44	94
		3	47	45	100
		4	56	58	282
		5	38	31	258
		6	29	28	90
		7	13	15	88
6	5	0	78	71	0
		1	32	29	5
		2	51	56	187
		3	12	6	197
		4	11	7	38
		5	7	6	127
		6	39	38	351
6	6	0	19	17	180
		2	68	65	85
		3	17	15	53
		4	63	64	273
		5	20	20	307
		6	28	26	97
6	7	0	92	82	0
		2	45	42	187
		3	6	3	211
		4	15	12	186
		6	28	32	19
6	8	0	7	5	0
		1	28	23	167
		2	56	52	97
		3	20	15	262
		4	58	57	279
		6	20	19	89
6	9	0	70	83	0
		1	31	28	132
		2	39	40	187
		3	8	10	342
		4	13	14	159
		5	19	16	359
		6	28	29	9
6	10	0	27	31	180
		1	12	9	191
		2	60	59	81
		3	23	23	273
		4	41	42	262
		5	30	33	104
		6	19	17	106
6	11	0	45	49	0
		1	35	35	195
		2	33	39	216
		3	28	26	332
		4	10	15	67
		6	12	16	1
		7	24	28	184
6	12	0	18	19	180
		1	22	25	111
		2	35	33	52
		3	49	46	258
		4	20	27	296
		5	34	36	72
		6	14	15	131
		7	9	9	224
6	13	0	26	24	0
		1	48	47	162
		2	16	15	164
		3	18	23	4
		4	8	10	124
		5	14	13	355
		6	8	10	334
		7	18	24	187
6	14	0	8	11	0
		1	9	10	102
		3	36	38	271
		5	32	33	94
		7	7	10	212
6	15	1	34	34	186
		4	7	5	319
		5	13	11	25
		7	15	18	180
6	16	1	16	12	88
		3	26	29	283
		4	13	15	111
		5	25	28	90
6	17	0	23	22	180
		1	26	28	181
		2	10	10	3
		3	8	10	352
		5	6	6	347
6	18	2	21	19	281
		3	15	17	255
		4	14	16	97
6	19	0	28	29	180
		1	11	8	177
		2	20	20	17
		3	7	8	49
6	20	1	5	6	40
		2	20	20	252
6	21	0	17	15	180
		2	11	14	358
7	0	1	76	68	90
		2	47	42	0
		3	26	22	270
		5	48	32	270
		6	12	16	0
		7	30	26	90
7	1	0	75	78	270
		1	22	19	13
		2	50	52	95
		3	64	70	183
		4	32	29	52
		5	25	44	342
		6	46	41	258
		7	9	5	101
7	2	0	39	32	90
		1	51	57	89
		2	54	59	335
		3	32	29	209
		4	49	51	186
		5	31	27	304
		6	22	20	16
		7	32	30	96
7	3	0	49	44	270
		1	51	51	41
		2	55	50	135
		3	52	53	177
		4	35	29	343
		5	30	32	351
		6	28	28	235
		7	20	18	94
7	4	0	48	50	90
		1	53	50	54
		2	28	24	289
		3	35	39	211
		4	32	30	202
		5	31	26	312
		6	22	23	63
		7	19	16	103
7	5	0	24	20	270
		1	67	62	80
		2	57	56	166
		3	29	31	194
		4	39	41	7
		5	36	37	301
		6	10	11	190
		7	20	20	85
7	6	0	71	67	90
		1	33	33	48
		2	48	45	266
		3	43	48	179
		5	31	27	356
		6	24	26	102
7	7	1	49	45	81
		2	42	38	182
		3	17	19	266
		4	66	61	354
		5	27	23	252
		6	29	25	176
		7	20	19	105
7	8	0	54	53	90
		1	25	22	332
		2	36	39	249
		3	41	39	163
		5	24	24	10
		6	20	19	66
7	9	0	41	39	90
		1	45	43	109
		2	18	23	233
		3	22	19	333
		4	29	32	355
		5	22	17	210
		6	27	27	151
		7	9	15	74
7	10	1	43	39	270
		2	38	37	214
		3	16	17	193
		4	22	24	8
		5	24	25	26
		6	16	19	138
		7	9	14	224
7	11	0	62	60	90
		1	17	22	155
		2	27	28	236
		3	42	38	335
		4	25	24	338
		5	16	19	212
		6	19	16	110
		7	9	12	88
7	12	0	16	13	90
		1	32	28	274
		2	40	40	205
		3	9	17	148
		4	29	29	5
		5	12	18	67
		6	7	12	134
		7	12	15	258
7	13	0	21	24	90
		1	18	20	162
		2	21	19	258
		3	33	36	8
		4	16	12	326
		5	15	15	185
		6	17	16	126
7	14	1	43	38	267
		2	15	13	168
		3	7	4	16
		4	7	11	359
		5	10	12	60
		6	7	9	187
7	15	0	25	25	90

TABLE 4 (*Continued*)

h	*k*	*l*	$\|F_o\|$	$\|F_c\|$	α°
		1	18	20	203
		2	8	10	307
		3	27	29	343
		5	15	17	163
7	16	0	18	19	270
		1	24	24	248
		2	16	15	148
		3	6	9	19
		5	11	13	120
7	17	0	7	12	90
		1	16	10	245
		2	13	13	324
		3	15	18	12
		4	15	15	180
		5	10	10	168
7	18	1	14	11	256
		3	12	12	33
		5	7	11	140
7	19	1	15	13	248
		2	3	5	347
7	20	1	4	7	206
		2	10	9	77
8	0	0	88	85	0
		1	13	5	90
		2	41	41	180
		3	7	0	270
		6	24	28	0
8	1	1	23	17	135
		2	41	40	101
		3	15	17	299
		4	39	42	271
		5	11	16	63
		6	21	25	100
		7	11	7	241
8	2	0	55	57	0
		1	41	36	197
		2	44	42	191
		3	10	17	328
		4	9	9	89
		5	12	14	43
		6	17	23	7
		7	8	17	190
8	3	0	32	32	180
		1	15	9	111
		2	50	48	73
		3	40	43	291
		4	35	41	283
		5	28	31	74
		6	13	21	114
		7	4	4	190
8	4	0	56	54	0
		1	46	44	176
		2	27	32	216
		3	31	31	341
		4	19	19	104
		5	14	12	6
		6	19	26	351
		7	10	30	179
8	5	1	14	11	109
		2	27	21	42
		3	33	30	253
		4	25	27	274
		5	37	40	87
		6	17	20	122
		7	25	19	242
8	6	0	11	10	0
		1	50	52	193
		2	12	18	230
		3	20	22	21
		5	13	16	23
		6	14	15	350
8	7	0	12	18	180
9	1	0	58	61	90
		1	32	34	71
		3	48	52	279
		4	18	18	341
		5	32	40	91
		6	7	11	168
		7	25	10	289
8	8	0	9	6	180
		1	58	56	189
		2	12	11	255
		3	10	14	26
		5	18	18	353
		6	9	6	105
		7	6	30	183
8	9	0	27	20	180
		1	22	21	78
		2	15	18	323
		3	45	44	273
		4	17	15	38
		5	20	26	101
		7	12	11	319
8	10	0	21	20	180
		1	48	52	170
		2	29	21	330
		3	10	4	36
		4	15	13.	62
		5	17	18	346
		6	16	18	181
8	11	0	8	3	180
		1	27	24	80
		2	16	15	277
		3	26	27	265
		4	28	30	97
		5	12	12	83
		6	10	17	254
8	12	0	30	32	180
		1	24	22	208
		2	23	24	10
		3	12	7	352
		6	10	17	175
8	13	0	8	7	0
		2	33	26	270
		4	29	30	89
		6	9	15	289
8	14	0	37	37	180
		2	29	29	20
		6	16	20	182
8	15	2	18	18	278
		4	28	26	100
8	16	0	32	28	180
		1	13	11	357
		2	21	19	0
		4	5	3	171
8	17	1	12	10	294
		2	18	19	260
		3	14	16	81
		4	15	19	92
8	18	0	19	26	180
		1	18	20	355
		2	18	13	26
		3	1	5	202
		4	4	7	311
8	19	1	6	9	247
		2	11	13	261
8	20	1	11	19	4
9	0	1	26	27	90
		2	36	40	180
		3	16	15	270
		4	43	42	0
		5	15	5	270
		6	10	20	180
		7	9	12	90
9	1	0	58	61	90
		1	10	12	344
		2	29	29	246
		3	27	27	174
		5	25	23	14
		6	23	20	101
9	2	0	26	33	90
		1	29	27	115
		2	43	44	200
		3	25	25	307
		4	30	31	348
		5	14	14	215
		6	20	18	147
		7	9	13	94
9	3	0	32	30	90
		1	26	23	279
		2	47	44	207
		3	29	30	174
		4	27	29	1
		5	24	26	23
		6	10	9	96
9	4	0	49	51	90
		1	39	35	125
		2	31	33	233
		3	34	36	4
		4	25	22	327
		5	16	20	215
		6	21	18	130
9	5	0	34	32	90
		1	48	47	287
		2	34	34	209
		3	14	11	156
		4	34	31	355
		5	17	22	81
		6	14	14	160
9	6	0	44	44	90
		1	17	20	199
		2	28	27	228
		3	40	37	357
		4	12	11	303
		5	24	22	161
		6	17	18	92
9	7	0	8	6	90
		1	21	32	256
		2	37	36	177
		3	17	16	56
		4	19	28	357
9	8	0	32	28	90
		1	18	17	215
		2	24	24	285
		3	29	31	1
		4	14	12	217
		5	22	25	165
		6	15	20	82
9	9	0	21	18	270
		1	33	30	264
		2	30	25	140
		3	21	16	45
		4	26	26	1
9	10	0	21	17	90
		1	15	12	221
		2	26	23	322
		3	17	19	26
		4	15	13	185
		5	21	24	162
		6	29	29	270
9	11	0	29	29	270
		1	21	17	249
		2	17	14	99
		3	20	16	35
		4	14	15	34
		5	11	11	162
9	12	0	8	3	90
		1	22	20	239
		2	24	22	351
		3	14	12	1
		4	28	26	170
9	13	0	27	27	270
		1	16	11	232
		2	19	19	78
		3	17	10	1
		5	12	11	191
9	14	0	8	11	270
		1	20	14	270
		2	24	20	358
		4	19	22	162
		5	5	7	139
9	15	0	24	24	270
		1	6	5	151
		2	21	16	67
		3	17	14	6
		4	9	7	131
		5	5	10	199
9	16	0	11	8	270
		1	13	11	277
		2	15	14	28
		3	11	8	138
		4	11	17	175
9	17	0	7	7	270
		1	8	7	134
		2	14	11	47
		3	8	7	323
		4	3	9	189
9	18	1	4	4	333
		2	9	7	74
10	0	1	27	20	90
		2	10	8	0
		3	62	53	270
		4	8	2	0
		5	35	38	90
10	1	0	11	3	180
		1	50	53	184
		2	9	7	312
		3	12	18	10
		5	29	29	358
		7	22	27	176
10	2	1	23	22	76
		2	13	11	338
		3	48	43	269
		4	17	10	87
		5	25	26	84
10	3	0	18	15	180
		1	39	38	184
		2	21	17	319
		3	10	11	41
		4	13	16	126
		5	12	13	349
		7	10	14	174
10	4	0	16	10	180
		1	11	11	2
		2	16	15	270
		3	24	21	264
		4	28	26	83
		5	21	21	89
10	5	0	43	42	180
		1	15	19	190
		2	29	28	1
		3	19	17	24
		5	14	13	313
10	6	0	7	6	0
		1	7	12	35
		2	32	23	265
		3	20	18	257
		4	38	34	96
		5	7	12	129
10	7	0	34	33	180

TABLE 4 (*Continued*)

h	*k*	*l*	$\|F_o\|$	$\|F_c\|$	α°	*h*	*k*	*l*	$\|F_o\|$	$\|F_c\|$	α°	*h*	*k*	*l*	$\|F_o\|$	$\|F_c\|$	α°	*h*	*k*	*l*	$\|F_o\|$	$\|F_c\|$	α°
		1	7	9	245			4	27	19	16			1	24	19	98	12	9	1	5	9	264
		2	36	29	7			5	9	7	113			2	15	12	36			3	19	18	76
		3	11	14	87	11	3	0	28	17	90			3	5	3	10	12	10	1	17	15	1
10	8	2	31	25	267			1	19	15	241			4	10	8	192			2	7	8	179
		3	14	11	88			2	25	26	339	11	13	0	15	11	270			3	6	4	196
		4	8	25	95			3	22	20	10			1	11	8	322	12	11	1	4	5	256
10	9	0	47	37	180			4	22	21	181			2	12	8	76			2	2	10	92
		1	25	21	358			5	16	19	174			3	14	14	182			3	9	15	71
		2	30	23	2	11	4	0	35	30	270			4	4	3	179	13	0	1	15	10	270
		3	6	7	134			1	29	23	244	11	14	1	16	15	98			2	22	17	0
10	10	1	16	15	285			2	26	21	123			2	9	3	357			4	17	23	180
		2	18	18	254			3	18	17	10	11	15	1	6	5	22	13	1	0	35	23	270
		3	22	20	91			5	13	15	135	12	0	0	29	26	180			1	11	4	114
		4	25	24	86	11	5	0	10	7	90			1	6	5	270			2	18	18	90
10	11	0	21	22	180			1	24	19	244			2	25	20	0			3	11	10	1
		1	32	27	3			2	19	19	355			5	5	8	270	13	2	1	13	11	289
		2	16	15	358			3	11	13	38	12	1	1	6	8	348			2	11	11	18
		3	8	10	170	11	6	0	41	36	270			2	28	22	267			3	4	7	131
10	12	1	9	9	264			1	8	8	198			4	23	23	89			4	14	19	178
		2	7	9	274			2	21	17	94	12	2	0	42	35	180	13	3	0	16	13	270
		3	22	26	97			3	21	19	14			1	22	19	336			1	20	14	112
		4	16	13	89			4	23	5	63			2	25	18	352			2	12	13	74
10	13	1	32	29	3			5	13	14	173	12	3	1	14	11	311			3	3	7	3
		3	12	7	161	11	7	0	10	2	270			2	26	22	276	13	4	1	16	12	325
10	14	1	12	15	269			1	19	18	276			3	21	19	83			3	13	17	163
		3	17	21	87			2	30	26	2			4	22	21	82	13	5	0	8	9	270
10	15	1	20	21	0			3	10	4	49			5	13	18	277			1	22	18	89
		2	5	2	284	11	8	0	34	28	270	12	4	0	25	23	180			2	5	6	77
		3	8	4	232			1	8	9	173			1	27	21	357			3	4	6	324
10	16	1	9	9	244			2	27	20	76			2	21	15	2	13	6	0	5	3	270
		2	8	8	96			3	17	13	15			3	13	10	165			1	9	8	342
10	17	1	10	14	7			4	5	7	163			5	5	8	205			2	10	8	71
11	0	1	32	26	270			5	7	7	183	12	5	1	20	13	280			3	11	17	171
		2	25	23	180	11	9	1	17	13	295			2	12	10	261	13	7	1	23	20	91
		4	22	18	0			2	25	19	22			3	30	25	97			2	7	5	15
		5	5	3	90	11	10	0	33	25	270			4	12	12	82	13	8	0	17	14	270
11	1	0	26	26	90			1	17	12	123			5	10	17	273			1	21	7	358
		1	16	13	206			2	16	13	75	12	6	1	27	28	355			2	6	6	128
		2	23	18	307			3	14	10	354			3	5	8	175	13	9	0	8	8	90
		3	22	20	0			4	16	14	170			5	9	10	184			1	6	14	90
		4	9	7	167	11	11	0	6	8	270	12	7	1	13	10	266	14	0	1	14	15	270
		5	13	13	185			1	13	12	326			3	29	25	86	14	1	1	22	21	358
11	2	0	32	24	270			2	21	14	30			5	11	20	279	14	2	1	11	9	261
		1	29	23	263			3	14	14	165	12	8	1	25	24	356	14	3	1	13	12	350
		2	27	22	132			4	12	11	179			3	11	10	190			5	3	10	161
		3	16	12	68	11	12	0	17	12	279												

scale by comparison of common reflexions on different photographs; the absolute scale was obtained at a later stage by correlation with the calculated structure amplitudes, $|F_c|$. In all, 1622 independent non-zero structure amplitudes, $|F_o|$, were evaluated.

The crystal density was determined by the flotation method in a mixture of chlorobenzene and bromobenzene.

Analysis of the Structure.—The position of the iodide ion was deduced initially from two-dimensional Patterson projections *P*(UV) and *P*(VW). These projections are shown in Figure 4.

An isotropic temperature factor *B* of 4·4 Å^2 being assumed, structure factors based on the iodide ion alone were calculated. The value of *R*, the average discrepancy between measured and calculated structure amplitudes, was 32%. With the measured $|F|$ values and the calculated phase angles we then evaluated a three-dimensional electron-density distribution. The contours were drawn on glass sheets which were stacked in a metal frame for study. Fourteen prominent electron-density peaks were chosen as atoms and included, weighted as carbon, in the second calculation of structure factors. The value of *R* was 29%.

In the subsequent three-dimensional electron-density distribution only three carbon atoms, C(14), C(19), and C(21) could not be assigned to peaks. The other atoms (each assigned its correct chemical type) were included in the third calculation of structure factors. The value of

TABLE 5

Anisotropic temperature factors ($b_{ij} \times 10^5$)

Atom	b_{11}	b_{22}	b_{33}	b_{12}	b_{23}	b_{13}
N(1)	876	349	3095	−62	−12	−2268
C(2)	856	407	2129	424	1137	1605
C(3)	518	296	2220	−404	221	−817
N(4)	665	499	1989	731	633	54
C(5)	1099	570	2055	715	−1099	−1203
C(6)	1389	508	2630	−135	1337	982
C(7)	945	399	1517	−179	701	1325
C(8)	1232	558	2091	−73	−514	1303
C(9)	1155	144	3515	−325	−1049	838
C(10)	1795	492	666	−538	−560	683
C(11)	1806	430	359	870	1185	−475
C(12)	1406	423	888	441	− 404	165
C(13)	672	714	826	−112	−653	−155
C(14)	1759	317	1143	−132	936	−1716
C(15)	2596	287	824	436	1045	−40
C(16)	2847	361	1133	877	1261	−1284
C(17)	1948	322	1825	529	368	550
C(18)	1995	628	793	−238	−1375	448
C(19)	1800	607	4071	137	686	−1778
C(20)	788	151	2570	113	179	941
C(21)	2401	337	1832	452	−378	1693
C(22)	1835	748	222	42	−482	1217
O(23)	2607	892	1760	354	1397	2067
O(24)	1649	296	4086	−289	−81	−616
I	1119	418	1887	−25	−137	−54
(B = 5 Å^2	1501	508	2130	—	—	—)

R decreased to 27·4%. In the subsequent F_o synthesis C(21) was clearly resolved and on the recalculation of structure factors the value of R was 24·3%.

In the fourth three-dimensional electron-density distribution the position of C(14) was unambiguously established but C(19) was not yet clear and the height of the peak allotted to

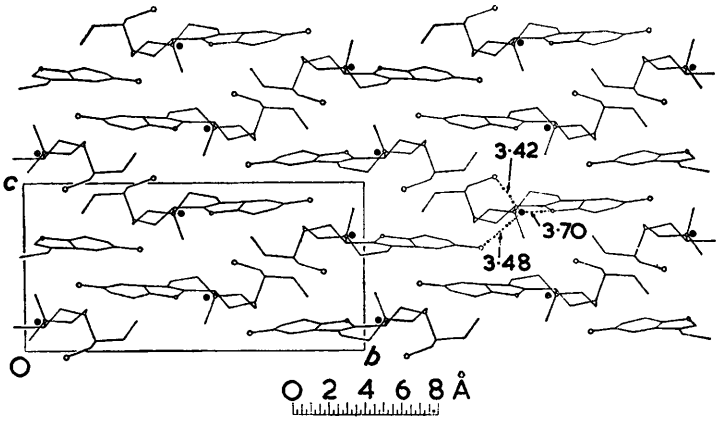


FIGURE 3. Projection of the crystal structure on (100)

C(18) was suspiciously low. Possible alternative sites for C(18) and C(19) were evident and these atoms were therefore excluded from the fifth calculation of structure factors. The value of R was 20·6%.

The fifth three-dimensional electron-density distribution unambiguously established structure (II) for hunterburnine β-methiodide. On the calculation of structure factors with all atoms, except hydrogen, included the value of R was 18·4%.

Two rounds of refinement by the Fourier method were then carried out. Both F_o and F_c syntheses were computed and atomic co-ordinates determined by numerical interpolation; errors due to termination of series were corrected and individual isotropic temperature factors obtained for the atoms. The value of R fell to 18·1%.

For the final stages of the analysis Rollett's least-squares programme [22] for DEUCE was used to refine positional and anisotropic thermal parameters for all atoms apart from hydrogen. The weighting scheme employed was

$$\sqrt{w} = |F_o|/15 \text{ if } |F_o| < 15,$$

$$\sqrt{w} = 15/|F_o| \text{ if } |F_o| > 15.$$

After six rounds of calculations the value of R decreased to 15·4% and the parameter shifts became very small; the refinement was therefore terminated.

For the structure-factor calculations the theoretical atomic scattering factors employed were those of Berghuis *et al.*[23] for carbon, nitrogen, and oxygen, and the Thomas–Fermi values [24] for iodine. The final calculated structure amplitudes and phase constants are listed with the

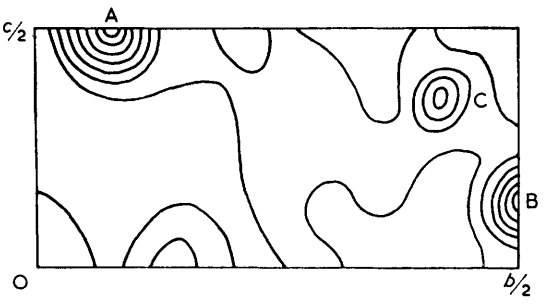


FIGURE 4*a*

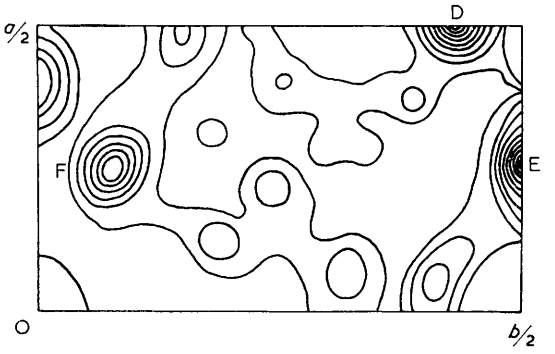


FIGURE 4*b*

FIGURE 4. Patterson projections along (*a*) the *a*-axis, (*b*) the *c*-axis. The iodine vector peaks shown are A ($2y$, $\frac{1}{2}$), B ($\frac{1}{2}$, $\frac{1}{2} - 2z$), C ($\frac{1}{2} - 2y$, $2z$), D ($\frac{1}{2}$, $\frac{1}{2} - 2y$), E ($2x$, $\frac{1}{2}$), F ($\frac{1}{2} - 2x$, $2y$). A, B, D, and E are double weight peaks, and C and F single weight peaks

measured values of the structure amplitudes in Table 4. With the measured structure amplitudes and the phase constants of Table 4 we calculated the final electron-density distribution which is shown in Figure 1 by means of superimposed contour sections drawn parallel to (001). The final atomic co-ordinates, molecular dimensions, and some non-bonded distances are given in Tables 1 and 2. The parameters defining the anisotropic thermal vibrations by means of the equation

$$\exp(-B \sin^2\theta/\lambda^2) = 2^{-(b_{11}h^2 + b_{22}k^2 + b_{33}l^2 + b_{12}hk + b_{23}kl + b_{31}hl)}$$

are shown in Table 5. The standard deviations of the final atomic co-ordinates were derived from the least-squares residuals by means of the equation

$$\sigma^2(x_i) = \sum w_j(\Delta F_j)^2/[(n - s)\sum w\,(\partial F_j/\partial x_i)^2]$$

The results are listed in Table 3.

The extensive calculations were performed on the Glasgow University DEUCE computer, with programmes devised by Dr. J. S. Rollett and Dr. J. G. Sime. J. D. M. A. is indebted to the Department of Scientific and Industrial Research for financial support.

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