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Isotope and Bonding Effects in Ammonium Oxalate Monohydrate, Determined by the Combined Use of Neutron and X-ray Diffraction Analyses

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Three-dimensional neutron and X-ray diffraction structural analyses have been carried out on protonated, deuterated and null-matrix (63% H, 37% D) crystals of ammonium oxalate monohydrate. The null-matrix composition is such that the neutron hydrogen and deuterium scattering lengths cancel ($b_H = -0{\cdot}378 \times 10^{-12}$ cm; $b_D = 0{\cdot}65 \times 10^{-12}$ cm). Absorption and extinction corrections were applied and refinement was by full-matrix least-squares calculations with anisotropic temperature factors. It was shown statistically that different sets of coordinates and temperature factors resulted if extinction corrections were not applied. In the null-matrix crystal the deuterium atoms were found to show a preference for the ammonium ion sites and the protons for the water sites, with a separation factor $(H/D)_{water}/(H/D)_{ammonium}$ of 1·07 (2). There was no significant expansion of the hydrogen bonds on deuteration. X–N maps for the deuterated and protonated forms (difference syntheses derived from neutron parameters and X-ray data) revealed deviations from spherical symmetry of the atoms, consistent with bonding and lone-pair electron distributions. These features were not observed with the X-ray data for the null-matrix crystals because of excessive extinction and would not have been seen for the deuterated and protonated crystals had extinction corrections not been applied.

Introduction

Recent work has shown that X-ray and neutron-diffraction data for crystals containing lighter atoms can be combined to give information on the bonding electron distribution. The basis of the method is to determine atomic positions and temperature factors by neutron-diffraction methods and to use these to calculate X-ray structure factors with spherical-atom form factors. Systematic differences between these and the observed X-ray structure factors, as revealed in a difference synthesis, the 'X–N map', are due to distortions from spherical-atom symmetry caused by electron redistribution due to bonding. Coppens (1967) first applied this method to reveal bonding and lone-pair electron density in *sym*-triazine. Coppens, Sabine, Delaplane & Ibers (1969), hereafter CSDI (1969), have also found bonding electron and lone-pair density effects in α-oxalic acid dihydrate. Duckworth, Willis & Pawley (1970) detected lone-pair density on the nitrogen atom in hexamethylenetetramine. Bonding effects in the X-ray data for a number of published structures have been discussed by O'Connell, Rae & Maslen (1966). Distortions from spherical symmetry in the X-ray charge density of bonded atoms also gives rise to systematic differences between X-ray and neutron-diffraction positions: the 'asphericity shift' (Coulson, 1970).

Coppens (1970) found that the H and D atoms in a null-matrix crystal (63% H, 37% D) of α-oxalic acid dihydrate, for which the combined H and D neutron-scattering lengths cancelled, were not distributed randomly over the H,D sites. Instead, non-zero effective scattering lengths were measured at these sites and they indicated a preference of H for the stronger hydroxylic hydrogen bond. Full deuteration in this crystal also lengthens the hydrogen bonds by about 0·02 Å (Delaplane & Ibers, 1969).

Ammonium oxalate monohydrate was selected for further study of these bonding and isotope effects for the following reasons:

(*a*) it has not as yet been subjected to a full three-dimensional neutron-diffraction analysis, having been the subject of a two-dimensional neutron study by Padmanabhan, Srikantha & Mehdi Ali (1965) and a three-dimensional X-ray study at 30°K (Robertson, 1965);

(*b*) because it consists of first row atoms, these effects should be readily detectable;

(*c*) because it is an oxalate, the effects should be comparable with, and provide additional evidence for, the effects observed in α-oxalic acid dihydrate.

Experimental

Protonated, deuterated and null-matrix crystals were grown from water, 99·8% D_2O (three recrystallizations), and from a solution of null-matrix composition respectively.

The three forms are designated HOX, DOX and NOX. The basic crystal data are listed in Table 1. The cell dimensions of NOX and DOX were determined by least-squares refinements of powder data (using 60 and 67 powder line positions respectively) with the program of Mueller, Heaton & Miller (1960) (Philips 114·6 mm diameter camera, Co $K\alpha$ radiation).

Three-dimensional neutron-diffraction data to $\sin\theta/\lambda \sim 0{\cdot}66$ with $\lambda = 1{\cdot}17$ Å were collected in one octant of reciprocal space at room temperature for the neutron crystals, labelled HOX(*N*), DOX(*N*) and NOX(*N*). The automatic neutron diffractometer (Pryor, Ellis & Dullow, 1968) on HIFAR, the A.A.E.C. research reactor, was used. The statistical uncertainty in the recorded intensity was less than 1·4% for DOX(N), 2% for NOX(N) and 3% for HOX(N) for most of the reflexions.

X-ray diffraction data were obtained with the Sydney automatic diffractometer (Freeman, Guss, Nockolds, Page & Webster, 1970) using Ni-filtered Cu $K\alpha$ radiation and a constant scan speed. As this machine has Weissenberg geometry, two crystals of each isotopic form mounted about different axes were used. The two sets of data for each form were not combined as they did not have the same extinction characteristics, but were refined separately. All reflexions in each data set were measured in at least two octants and the results were averaged. The X-ray crystals studied were labelled HOX1(XR), HOX2(XR), NOX1(XR), NOX2(XR), DOX1(XR) and DOX2(XR).

Analysis of data

(*a*) *Data reduction and absorption corrections*

The X-ray intensities were corrected for absorption, and the intensities were then converted to $F_o(hkl)$ values with associated standard deviations using the Sydney University Chemistry School series of computer programs for data reduction.

The neutron–diffraction data sets were corrected for absorption and reduced to $F_o(hkl)$ values (with corresponding standard deviations) by the program *DRACULA* (written by Drs B. M. Craven, Margaret Elcombe & G. W. Cox at this Establishment). The neutron transmission factors were obtained by direct measurement.

(*b*) *Extinction corrections*

Extinction corrections were included in the least-squares refinements of the diffraction data by using the program *LINUS*, which is a modification of the crystallographic least-squares program *ORFLS* (Busing, Martin & Levy, 1962*a*) by Coppens & Hamilton (1970). The program is based on the Zachariasen (1967) formulae and distinguishes two kinds of crystals: type 1 in which the extinction is controlled by the misorientation of the mosaic blocks; and type 2 in which the major cause of the extinction effect is the size of the mosaic blocks. Extinction may be anisotropic (anisotropic Gaussian mosaic spread distribution for type 1, or ellipsoidal rather than spherical average particle shape for type 2), for which case the extinction coefficients are the six independent components of a symmetrical second order tensor. From the extinction coefficient g' (isotropic), an equivalent mosaic spread parameter η and spherical domain radius r can be obtained; from the anisotropic extinction coefficients Z_{ij} (type 1) equivalent mosaic spread parameters η_{ij} can be obtained, while from the anisotropic coefficients W_{ij} (type 2), equivalent r_{ij} may be obtained. The relevant equations are given in Coppens & Hamilton (1970).

(*c*) *Refinements*

The refinements listed in Table 2 were carried out using *LINUS*. The least-squares weights were $w_{hkl}=[\sigma(F_{hkl})]^{-2}$ and the function minimized was $\sum w(F_o-|F_c|)^2$. In the X-ray refinements, the hydrogen coordinates were refined but the hydrogen atoms were given fixed isotropic Debye–Waller B factors of 4·0 Å^2. The X-ray scattering curves for C, N, O and H were taken for neutral atoms from *International Tables for X-ray Crystallography* (1962), and were not corrected for anomalous dispersion. The neutron scattering lengths used were $b_{\rm C}=0{\cdot}66$, $b_{\rm N}=0{\cdot}94$, $b_{\rm O}=0{\cdot}58$, $b_{\rm H}=-0{\cdot}378$, in units of 10^{-12} cm (Neutron Diffraction Commission, 1969). For deuterium $b_{\rm D}$ was chosen as 0·65 since crystallographic work (Coppens & Sabine, 1969) indicates that the Commission value of 0·62 is too low. In the case of NOX(N), the (H,D) atoms were assumed to have the same positional and thermal parameters as in DOX(N), and the 'effective' scattering factors at the five (H,D) sites were equal to mb where the occupancy factors m were refined and b was arbitrarily given the value $0{\cdot}65\times10^{-12}$ cm. In the DOX(N) refinement, the neutron-scattering factor for D was multiplied by 0·95 (an assumed percentage of 98·5% D by weight), to allow for incomplete deuteration.

Table 1. *Crystal data for ammonium oxalate monohydrate, deuterate and null-matrix crystals at* 21 °C

Space group $P2_12_12$.

	HOX, $(NH_4)_2C_2O_4.H_2O$	DOX, $(ND_4)_2C_2O_4.D_2O$	NOX, $N[(H,D)_4]_2.C_2O_4.(H,D)_2O$
F.W.	142·11	152·03	145·73
	Orthorhombic	Orthorhombic	Orthorhombic
a (Å)	8·035 (4)	8·027 (2)	8·025 (2)
b	10·309 (4)	10·298 (2)	10·300 (2)
c	3·795 (2)*	3·816 (1)	3·799 (1)
U (Å^3)	314·4	315·4	314·0
D_m (g.cm^{-3})	1·50*		
D_x	1·501	1·601	1·541
Z	2	2	2
μ(Cu $K\alpha$) (cm^{-1})	13		
μ (neutron)	3·0	0·65	2·2

* Robertson (1965).

Table 2. *Final unweighted and weighted R values, R_U and R_W, together with χ values*

Refinement	Extinction	R_U	R_W	χ
HOX1(XR)	Iso	0·070	0·035	2·4
HOX2(XR)	Iso	0·040	0·017	1·5
HOX(N)	Iso	0·045	0·043	1·5
HOX(N)	Aniso 1	0·039	0·034	1·2
HOX(N)	Aniso 2	0·045	0·042	1·5
NOX1(XR)	Iso	0·035	0·035	12·8
NOX2(XR)	Iso	0·025	0·018	5·1
NOX(N)	Iso	0·034	0·044	1·5
NOX(N)	Aniso 1	0·034	0·042	1·4
NOX(N)	Aniso 2	0·037	0·044	1·5
DOX1(XR)	Iso	0·027	0·019	4·0
DOX2(XR)	Iso	0·024	0·021	7·4
DOX(N)	Iso	0·030	0·037	2·5
DOX(N)	Aniso 1	0·027	0·032	2·2
DOX(N)	Aniso 2	0·028	0·033	2·3

(*d*) *Results*

The final unweighted (R_U) and weighted (R_W) agreement indices (Busing *et al.*, 1962*a*) are given in Table 2. The quantity $\chi=[\sum w(F_o-|F_c|)^2/(\mathrm{NO}-\mathrm{NV})]^{1/2}$, where NO is the number of observations and NV the number of variables, is also given for each refinement; this should be unity for correspondence between theory and experiment. The R values were very low. The χ values were close to those expected for this kind of experiment, except perhaps in the case of DOX2(XR), and more especially NOX1(XR) ($\chi=12·8$). The latter

Table 3. *Extinction parameters, with corresponding mosaic spread and domain radius values*

(*a*) Isotropic

Crystal	g'	Mosaic spread, η (sec)	Spherical domain radius, r (μ)
HOX1(XR)	0·36 (5)	16·2	0·6
HOX2(XR)	0·39 (2)	14·9	0·6
HOX(N)	6·3 (4)	0·9	7·4
NOX1(XR)	10·2 (1)	0·6	15·7
NOX2(XR)	0·69 (2)	8·4	1·1
NOX(N)	0·43 (2)	13·5	0·5
DOX1(XR)	5·3 (1)	1·1	8·2
DOX2(XR)	0·55 (2)	10·6	0·8
DOX(N)	2·0 (1)	2·9	2·3

Table 3 (*cont.*)

(*b*) Anisotropic type 1

Crystal	Z'_{11}	Z'_{22}	Z'_{33}	Z'_{12}	Z'_{13}	Z'_{23}	$\langle\eta_1\rangle$	$\langle\eta_2\rangle$	$\langle\eta_3\rangle$
HOX(N)	22 (4)	12 (2)	58 (7)	0 (2)	−5 (4)	−8 (4)	1·2	1·7	0·8
NOX(N)	0·19 (6)	0·22 (7)	0·40 (5)	−0·07 (3)	0·10 (7)	−0·14 (8)	13·2	12·4	9·2
DOX(N)	2·3 (3)	3·2 (3)	5·6 (4)	0·6 (2)	0·6 (3)	0·7 (4)	3·8	3·3	2·5

(*c*) Anisotropic type 2

Crystal	W'_{11}	W'_{22}	W'_{33}	W'_{12}	W'_{13}	W'_{23}	$\langle r_1\rangle$	$\langle r_2\rangle$	$\langle r_3\rangle$
HOX(N)	0·04 (1)	0·08 (1)	0·47 (4)	0·01 (1)	−0·06 (2)	0·09 (1)	5·9	4·2	1·7
NOX(N)	2·2 (2)	0·8 (1)	2·1 (8)	0·0 (1)	1·8 (3)	0·7 (3)	0·8	1·3	0·8
DOX(N)	0·18 (2)	0·18 (1)	0·33 (6)	0·07 (1)	−0·15 (4)	0·26 (2)	2·8	2·8	2·0

Table 4. *Positional parameters* ($\times 10^4$)

HOX(XR) denotes the average of HOX1(XR) and HOX2(XR) and similarly for NOX(XR) and DOX(XR). In the X-ray case, hydrogen coordinates have been averaged over all refinements. Refinements are with isotropic extinction corrections, except for HOX(N), which was anisotropic type 1.

	Crystal	x	y	z
C	HOX(XR)	917 (4)	230 (4)	707 (11)
	HOX(N)	923 (2)	233 (2)	705 (5)
	NOX(XR)	927 (2)	244 (4)	702 (4)
	NOX(N)	921 (1)	232 (1)	701 (3)
	DOX(XR)	929 (2)	244 (2)	710 (3)
	DOX (N)	922 (1)	237 (1)	703 (3)
O(1)	HOX(XR)	2021 (3)	−591 (2)	1416 (9)
	HOX(N)	2017 (2)	−580 (2)	1439 (7)
	NOX(XR)	2017 (1)	−587 (1)	1410 (3)
	NOX(N)	2018 (2)	−587 (2)	1423 (4)
	DOX(XR)	2015 (1)	−589 (1)	1410 (3)
	DOX(N)	2017 (2)	−586 (2)	1410 (5)
O(2)	HOX(XR)	1178 (3)	1406 (2)	20 (10)
	HOX(N)	1177 (3)	1406 (2)	19 (7)
	NOX(XR)	1182 (1)	1408 (2)	15 (4)
	NOX(N)	1176 (2)	1410 (2)	12 (4)
	DOX(XR)	1178 (1)	1408 (1)	18 (3)
	DOX(N)	1176 (2)	1406 (1)	22 (5)
N	HOX(XR)	3876 (5)	2264 (3)	4249 (10)
	HOX(N)	3868 (2)	2265 (1)	4245 (4)
	NOX(XR)	3869 (1)	2262 (3)	4253 (4)
	NOX(N)	3867 (1)	2266 (1)	4251 (2)
	DOX(XR)	3867 (1)	2265 (1)	4259 (4)
	DOX(N)	3868 (1)	2266 (1)	4251 (2)
O(W)	HOX(XR)	0	$\frac{1}{2}$	1860 (13)
	HOX(N)			1879 (9)
	NOX(XR)			1860 (4)
	NOX(N)			1855 (5)
	DOX(XR)			1877 (5)
	DOX(N)			1835 (6)
H(W)	*	841 (103)	4829 (18)	467 (54)
	HOX(N)	968 (6)	4821 (4)	375 (13)
	DOX(N)	957 (2)	4811 (1)	382 (4)
H(1)	*	4749 (77)	2657 (40)	2902 (112)
	HOX(N)	4747 (5)	2677 (4)	2614 (11)
	DOX(N)	4750 (2)	2678 (1)	2638 (4)
H(2)	*	4248 (54)	1519 (44)	5425 (141)
	HOX(N)	4327 (6)	1492 (5)	5608 (12)
	DOX(N)	4335 (2)	1475 (1)	5587 (4)
H(3)	*	2912 (123)	2007 (36)	2882 (147)
	HOX(N)	2891 (6)	1981 (4)	2745 (12)
	DOX(N)	2874 (2)	1970 (2)	2723 (4)
H(4)	*	3553 (58)	2935 (54)	5810 (146)
	HOX(N)	3492 (6)	2969 (5)	6034 (13)
	DOX(N)	3501 (2)	2970 (2)	6021 (4)

* Average over all X-ray refinements.

crystal suffered from very severe extinction; refinements with anisotropic extinction parameters (assuming the symmetry of the anisotropy was that of the crystal) gave no significant improvement according to the R-value ratio test of Hamilton (1964). R-value ratio tests on the results in Table 2 showed that at the 95% confidence level there was no improvement in going from isotropic to anisotropic extinction, except in the case of HOX(N), which was of type 1 (extinction dominated by mosaic spread effect). Anisotropic refinements were also carried out for the X-ray data sets; these gave results which were not significantly different from the isotropic refinements and are not given in Table 2. Thus, in this crystal, there was little to gain in going from isotropic to anisotropic extinction corrections.

The crystals were probably of type 1 (extinction dominated by mosaic spread). There were large variations in crystal perfection in the crystals studied; the crystals had either low or very high extinction parameters. The crystals with high g' values were clear needles, *e.g.* HOX(N), DOX(N), DOX1(XR) and NOX1(XR), whereas the crystals with low g' were opaque squat pillars, *e.g.* NOX(N) and DOX2(XR). This variation is also seen in the mosaic spreads and particle sizes in Table 3. A Lang topograph (Michell & Smith, unpublished work) of HOX(N) showed pendellösung fringes characteristic of a perfect crystal.

The extinction parameters and corresponding mosaic spread and domain radius parameters are in Table 3. The final positional and thermal parameters are given in Tables 4 and 5 and interatomic distances and angles uncorrected for thermal motion, as given by the program *ORFFE* (Busing, Martin & Levy, 1962*b*), in Table 6. The final observed and calculated structure factors for the nine crystals are given in Table 7.

Table 5. *Thermal parameters* ($\times 10^4$)

The temperature factor expression is $\exp[-(\beta_{11}h^2+2\beta_{12}hk\ldots)]$. HOX(XR) is the average of HOX1(XR) and HOX2(XR), and similarly for NOX(XR) and DOX(XR). Refinements are with isotropic extinction corrections except for HOX(N), which was anisotropic type 1.

	Crystal	β_{11}	β_{22}	β_{33}	β_{12}	β_{13}	β_{23}
C	HOX(XR)	60 (9)	52 (4)	284 (34)	−10 (4)	−28 (14)	−3 (9)
	NOX(XR)	57 (2)	40 (5)	269 (10)	−8 (2)	−7 (3)	0 (3)
	DOX(XR)	59 (2)	38 (4)	262 (11)	−3 (2)	−11 (4)	2 (4)
	HOX(N)	40 (3)	31 (2)	227 (12)	−3 (1)	−16 (4)	15 (3)
	NOX(N)	49 (1)	35 (1)	310 (5)	−5 (1)	−6 (2)	8 (2)
	DOX(N)	43 (2)	31 (1)	261 (7)	−5 (1)	−11 (3)	11 (2)
O(1)	HOX(XR)	60 (5)	48 (2)	628 (29)	11 (3)	0 (12)	54 (7)
	NOX(XR)	60 (1)	47 (2)	636 (9)	4 (1)	−2 (3)	52 (3)
	DOX(XR)	58 (1)	42 (2)	624 (9)	5 (1)	−2 (4)	53 (3)
	HOX(N)	41 (3)	45 (2)	554 (19)	6 (2)	−5 (6)	50 (5)
	NOX(N)	58 (2)	51 (2)	593 (9)	4 (1)	−2 (3)	46 (3)
	DOX(N)	48 (2)	44 (1)	600 (12)	3 (1)	−4 (4)	55 (3)
O(2)	HOX(XR)	74 (5)	38 (2)	643 (30)	−13 (3)	−37 (11)	34 (7)
	NOX(XR)	72 (1)	39 (4)	599 (10)	−15 (1)	−44 (3)	46 (3)
	DOX(XR)	70 (1)	24 (2)	642 (10)	−12 (1)	−50 (4)	39 (4)
	HOX(N)	62 (3)	31 (2)	532 (19)	−10 (2)	−46 (6)	38 (5)
	NOX(N)	65 (2)	43 (1)	579 (10)	−13 (1)	−46 (3)	34 (3)
	DOX(N)	63 (2)	33 (1)	568 (12)	−13 (1)	−44 (4)	40 (4)
N	HOX(XR)	85 (8)	49 (4)	377 (37)	−13 (4)	5 (16)	−11 (8)
	NOX(XR)	80 (1)	42 (3)	377 (10)	−10 (1)	1 (4)	−2 (3)
	DOX(XR)	80 (1)	40 (2)	389 (10)	−12 (1)	4 (4)	−6 (4)
	HOX(N)	70 (2)	38 (2)	284 (10)	−8 (1)	8 (4)	−5 (3)
	NOX(N)	80 (1)	48 (1)	344 (4)	−12 (1)	0 (2)	−5 (1)
	DOX(N)	77 (1)	43 (1)	317 (6)	−9 (1)	−1 (2)	−6 (2)
O(*W*)	HOX(XR)	73 (10)	60 (4)	516 (41)	−6 (5)	0	0
	NOX(XR)	66 (1)	58 (3)	528 (10)	0 (1)		
	DOX(XR)	66 (2)	58 (3)	518 (12)	1 (1)		
	HOX(N)	45 (4)	49 (3)	387 (20)	4 (3)		
	NOX(N)	64 (2)	62 (2)	447 (10)	1 (2)		
	DOX(N)	61 (3)	47 (2)	450 (13)	−5 (2)		
H(*W*)	HOX(N)	83 (6)	80 (5)	612 (35)	−3 (4)	25 (12)	−37 (11)
	DOX(N)	65 (2)	57 (2)	523 (10)	−1 (1)	35 (4)	−5 (4)
H(1)	HOX(N)	87 (7)	71 (4)	516 (27)	−12 (4)	32 (11)	4 (9)
	DOX(N)	86 (2)	69 (2)	442 (9)	−18 (1)	41 (4)	3 (3)
H(2)	HOX(N)	156 (8)	67 (5)	450 (28)	1 (5)	−13 (13)	28 (9)
	DOX(N)	126 (2)	48 (1)	451 (10)	4 (1)	−21 (4)	30 (4)
H(3)	HOX(N)	110 (8)	77 (4)	513 (29)	−17 (4)	−40 (12)	−1 (9)
	DOX(N)	84 (2)	66 (2)	470 (10)	−22 (1)	−32 (4)	−8 (4)
H(4)	HOX(N)	125 (7)	69 (4)	530 (30)	−2 (4)	43 (13)	−60 (10)
	DOX(N)	121 (2)	54 (1)	410 (9)	0 (1)	33 (4)	−35 (4)

Table 6. *Interatomic distances and angles, uncorrected for thermal motion*

The X-ray values were averaged over the X-ray refinements (isotropic extinction corrections, except for NOX1(XR) and NOX2(XR) which were anisotropic type 1). The neutron results were averaged over the three forms (anisotropic type 1 refinements).

	X-rays (average)	Neutrons (average)
C——C	1·565 (14) Å	1·557 (2) Å
C——O(1)	1·254 (5)	1·248 (2)
C——O(2)	1·248 (9)	1·254 (3)
N——H(1)	0·966 (57)	1·030 (3)
N——H(2)	0·943 (49)	1·026 (4)
N——H(3)	0·974 (106)	1·024 (3)
N——H(4)	0·951 (42)	1·035 (3)
O(*W*)-H(*W*)	0·887 (58)	0·976 (4)
O(*W*)-H(*W*)··O(1)	2·767 (4)	2·767 (4)
N——H(2)···O(2)	3·172 (4)	3·172 (4)
N——H(3)···O(*W*)	2·905 (4)	2·910 (4)
N——H(4)···O(2)	2·835 (6)	2·836 (4)
N——H(5)···O(1)	2·850 (3)	2·849 (4)
O(1)—C——O(2)	125·9 (4)°	125·6 (2)°
O(1)—C——C	116·7 (12)	117·5 (4)
O(2)—C——C	117·4 (5)	116·9 (2)
O(*W*)-H(*W*)-O(1)	163·4 (49)	166·9 (4)
N——H(1)—O(2)	169·7 (38)	172·5 (3)
N——H(2)—O(*W*)	175·2 (34)	175·2 (4)
N——H(3)—O(2)	175·1 (43)	177·8 (4)
N——H(4)—O(1)	172·4 (42)	169·6 (4)
H(*W*)-O(*W*)-H(*W*)	104·0 (80)	109·3 (6)
H(2)—N——H(1)	111·3 (27)	112·0 (3)
H(2)—N——H(4)	112·7 (35)	109·1 (4)
H(2)—N——H(3)	106·6 (10)	109·4 (3)
H(4)—N——H(3)	109·6 (29)	110·4 (3)
H(3)—N——H(1)	113·5 (50)	108·2 (3)
H(1)—N——H(4)	103·0 (54)	107·5 (3)

Discussion

(*a*) *Effects of extinction on coordinates and thermal parameters*

The effects of extinction corrections on the accuracy of a refinement are illustrated at this point using the DOX(N) results, for which $g' = 2\cdot0(1)$. The DOX(N) results with and without extinction corrections are given in Table 8. The extinction corrections reduced the R indices and χ values by a factor of 3 and the coordinate and temperature factor standard deviations by a factor of ~5. The effect on the coordinates themselves did not appear large, but the correction increased all the temperature factors by a factor of ~1·3. Results before and after correction were compared statistically. First, the positional parameters were considered.

The Δ_1 values in Table 8 were converted to Δ_1/σ where Δ_1 is the absolute difference between a corrected and uncorrected result, and σ is the standard deviation of the difference. The total number of Δ_1/σ values was 28. For statistically identical data sets the number of values in ranges of Δ_1/σ should be predictable from the normal distribution. A χ^2 test (Hamilton, 1964) was used to see whether this hypothesis was true at the 99·5% confidence level. With the data for $\sigma > 1\cdot25$ taken as a single group

$$\chi^2 = \sum \left[\frac{(N_{\text{obs}} - N_{\text{expected}})^2}{N_{\text{expected}}} \right]$$

was 26, whereas for six groups (five degrees of freedom) $\chi_{5,\,0.005} = 16\cdot8$. Thus it was extremely unlikely the two coordinate sets were the same. The same result was obtained with smaller numbers of groups. The χ^2 test also failed with the two temperature-factor sets.

Thus, while it is generally recognized that the lack of an extinction correction gives artificially low temperature factors, the possibility that the positional parameters are also affected should not be overlooked. The worst affected reflexion in the DOX(N) data was 021, for which the intensity was reduced by a factor of seven.

(*b*) *Isotope distribution in null-matrix crystal NOX(N)*

The measured scattering lengths at the various H, D sites in NOX(N) are listed as b(H, D) values in Table 9. These values showed the crystal differed slightly from the null-matrix composition (59·9 ± 0·3 atomic % instead of 63·2% for a null-matrix). The scattering length residual at the water site, $0\cdot021(3) \times 10^{-12}$ cm was lower than the residuals at the ammonium sites, suggesting a preference of hydrogen for the water site and deuterium for the ammonium sites. Defining a separation factor as

$$S = \frac{(\text{H/D})_{\text{water}}}{(\text{H/D})_{\text{ammonium ion}}},$$

it was found that $S = 1\cdot07 \pm 0\cdot02$, and the isotope separation effect was small but significant. Coppens (1970) has carried out a similar analysis on a null-matrix crystal of α-oxalic acid dihydrate; for his results we find $S = 1\cdot42$ (3) for that compound, a larger effect. In α-oxalic acid dihydrate, the hydrogen atoms prefer the hydroxyl site, which has a shorter and presumably stronger hydrogen bond than the water sites. There is no marked differentiation of hydrogen-bond distances in ammonium oxalate monohydrate. The isotope effect in the present analysis was not detectable before extinction corrections were introduced.

(*c*) *Asphericity shifts*

Dawson (1964) noted that the use of spherically averaged f curves in least-squares refinements of X-ray data gave shifts (~0·02 Å) and apparent thermal anisotropy, which were actually a compensation for an incorrectly assumed atom shape.

Coulson (1970) calculated that the true centroid of charge is displaced 0·04 Å towards the lone-pair electrons for the nitrogen atom in ammonia. For the oxygen atom in a nitro group, the displacement towards N due to the σ and π bonds was 0·023 Å and the displacement away due to the lone-pair effects was 0·039 Å giving a net displacement of 0·016 Å towards the lone-pair. Duckworth *et al.* (1970) found a displacement of the nitrogen atom in hexamethylene

Table 7. *Observed and calculated structure factors*

FO and FC refer to the moduli of the structure factor and the FO are extinction-corrected. Asterisks denote unobservably small reflexions.

H	K	FO	FC
HOX 1 L - 0			
2	0	6.86	6.49
4	0	22.34	22.96
6	0	4.52	4.78
8	0	8.96	7.98
2	1	9.65	9.70
3	1	35.88	35.62
4	1	5.63	5.66
5	1	6.69	6.92
6	1	5.52	4.49
7	1	10.39	11.34
8	1	* 1.87	1.86
9	1	4.20	3.18
1	2	8.93	9.00
2	2	* 1.43	0.27
3	2	30.74	31.33
4	2	19.81	19.14
5	2	6.40	6.49
6	2	5.37	4.19
7	2	12.15	12.48
8	2	5.25	5.03
9	2	3.64	3.56
1	3	7.47	7.67
2	3	20.88	21.63
3	3	9.97	9.93
4	3	19.10	19.03
5	3	* 2.27	1.75
6	3	16.17	18.05
7	3	11.95	12.31
8	3	* 2.50	0.89
9	3	2.49	1.81
0	4	21.02	21.38
1	4	28.33	28.73
2	4	10.92	12.05
3	4	4.16	3.77
4	4	6.73	7.29
5	4	* 1.83	2.75
6	4	* 1.98	0.72
7	4	* 1.87	2.33
8	4	* 1.79	1.23
9	4	* 2.31	1.84
1	5	14.15	13.54
2	5	24.27	25.68
3	5	* 1.76	2.33
4	5	10.65	12.75
5	5	5.88	5.29
6	5	4.33	3.18
7	5	5.27	5.46
8	5	4.23	4.35
0	6	5.63	5.78
1	6	10.64	10.33
2	6	14.52	14.29
3	6	7.67	7.61
4	6	* 2.23	1.21
5	6	* 1.93	2.45
6	6	* 1.98	1.81
7	6	3.98	3.89
8	6	5.23	5.23
1	7	8.27	7.83
2	7	4.59	5.39
3	7	7.51	7.77
4	7	9.16	8.33
5	7	21.28	22.12
6	7	5.69	5.12
7	7	* 2.21	2.81
8	7	* 2.21	1.44
0	8	8.26	7.33
1	8	* 2.37	1.26
2	8	12.62	12.53
3	8	5.13	5.74
4	8	7.77	7.67
5	8	* 1.79	0.68
6	8	* 1.72	1.38
7	8	* 2.12	2.26
1	9	8.99	9.28
2	9	11.13	11.64
3	9	4.26	4.28
4	9	* 3.36	2.68
5	9	3.67	3.93
6	9	3.92	3.51
0	10	7.43	7.61
1	10	10.72	12.74
2	10	5.82	5.93
3	10	8.41	7.69
4	10	5.05	4.52
5	10	4.62	3.26
1	11	3.97	3.68
2	11	4.28	3.67
3	11	* 1.68	0.62
4	11	* 1.57	1.84
0	12	* 1.43	2.26
1	12	* 1.57	0.03
2	12	* 1.54	2.26
HOX 1 L - 1			
2	0	2.86	2.75
3	0	* 1.51	0.74
4	0	6.30	3.82
5	0	13.15	13.67
6	0	* 1.95	1.67
7	0	4.09	3.76
8	0	* 1.84	0.17
9	0	* 1.62	2.83
2	1	40.13	39.66
3	1	22.86	22.44
4	1	11.63	11.36
5	1	8.71	8.85
6	1	14.91	15.49
7	1	6.39	6.87
8	1	6.78	6.99
9	1	5.32	4.91
1	2	27.73	27.55
2	2	7.95	7.55
3	2	16.75	16.45
4	2	11.03	10.68
5	2	8.05	8.13
6	2	6.23	5.73
7	2	7.24	7.06
8	2	* 1.88	1.84
9	2	6.66	6.13
0	3	* 1.84	1.62
1	3	22.09	21.09
2	3	12.67	12.29
3	3	5.26	5.25
4	3	9.17	8.99
5	3	5.97	6.59
6	3	5.61	6.21
7	3	7.24	7.71
8	3	4.93	5.11
9	3	* 2.51	3.97
0	4	14.33	13.64
1	4	10.29	10.17
2	4	4.65	4.22
3	4	11.57	11.25
4	4	17.76	17.51
5	4	5.19	5.35
6	4	6.97	7.23
7	4	4.25	3.96
8	4	5.38	5.57
0	5	* 1.66	1.61
1	5	7.01	6.53
2	5	6.83	7.12
3	5	10.33	10.41
4	5	12.29	11.54
5	5	10.10	8.91
6	5	5.89	6.62
7	5	4.97	4.63
8	5	* 1.66	0.56
0	6	14.81	14.11
1	6	16.55	16.06
2	6	10.78	10.58
3	6	7.54	7.59
4	6	11.83	11.52
5	6	5.85	5.96
6	6	5.72	5.47
7	6	5.19	5.07
8	6	6.01	6.50
0	7	12.68	12.78
1	7	4.50	4.36
2	7	* 2.80	1.62
3	7	* 2.87	3.84
4	7	6.38	5.64
5	7	8.47	8.96
6	7	* 2.95	3.81
7	7	* 2.10	2.13
0	8	3.85	2.47
1	8	8.57	9.23
2	8	7.52	7.18
3	8	14.18	14.36
4	8	5.15	5.36
5	8	* 1.91	3.39
6	8	3.22	3.44
7	8	8.87	8.09
0	9	5.59	5.47
1	9	* 1.99	2.34
2	9	11.52	12.32
3	9	* 1.91	2.64
4	9	5.80	6.18
5	9	6.35	6.90
6	9	5.93	5.99
0	10	5.60	5.90
1	10	* 3.34	3.83
2	10	* 1.91	2.54
3	10	4.64	4.47
4	10	* 3.28	3.23
5	10	* 1.58	1.27
0	11	5.55	5.78
1	11	7.77	7.88
2	11	* 1.77	3.03
3	11	4.84	4.28
4	11	* 1.47	1.37
0	12	* 1.51	0.77
1	12	3.55	3.17
HOX 1 L - 2			
2	0	2.89	2.30
3	0	8.89	8.88
5	0	* 1.91	1.30
6	0	9.57	9.22
7	0	3.85	4.12
8	0	4.77	4.71
2	1	11.75	11.95
3	1	* 1.58	2.13
4	1	* 1.76	2.80
5	1	5.92	5.97
6	1	4.55	5.08
7	1	* 1.91	3.39
8	1	7.64	5.61
0	2	9.02	9.30
1	2	13.64	13.46
3	2	10.84	10.78
4	2	7.33	7.54
5	2	9.82	9.22
6	2	6.05	6.52
7	2	6.81	6.76
8	2	3.92	3.10
0	3	9.19	9.36
1	3	4.47	4.29
2	3	14.46	14.32
3	3	15.68	15.34
4	3	6.64	6.55
5	3	* 1.94	2.28
6	3	7.83	7.39
7	3	5.08	6.09
8	3	3.44	3.61
0	4	6.79	7.16
2	4	7.63	7.09
3	4	5.17	4.28
4	4	8.11	8.07
5	4	4.35	4.24
6	4	5.56	5.53
7	4	* 2.28	2.72
8	4	4.14	3.68
0	5	7.42	6.97
1	5	9.82	10.14
2	5	7.66	7.65
3	5	6.76	6.27
4	5	* 1.95	2.74
5	5	3.55	3.36
6	5	7.99	7.68
7	5	* 1.72	0.68
0	6	* 1.83	2.56
2	6	5.21	4.24
3	6	5.58	7.11
4	6	8.72	8.88
5	6	3.95	3.31
6	6	7.31	7.34
7	6	* 2.09	1.74
0	7	* 1.87	1.70
1	7	11.41	11.27
3	7	8.11	8.35
4	7	8.30	7.95
5	7	5.35	5.24
6	7	5.12	5.56
0	8	5.74	5.68
1	8	4.20	4.63
2	8	* 1.91	2.27
3	8	5.10	5.49
4	8	6.80	6.66
5	8	5.29	4.71
6	8	* 1.54	2.28
0	9	9.45	9.78
1	9	* 1.83	2.09
2	9	7.61	7.18
3	9	* 3.83	2.38
4	9	* 1.76	1.43
5	9	3.90	4.07
1	10	4.17	4.67
2	10	6.44	6.57
3	10	6.58	6.32
0	11	* 1.54	0.35
1	11	4.62	3.66
HOX 1 L - 3			
2	0	* 1.52	2.30
3	0	* 1.62	1.13
4	0	9.03	9.52
5	0	* 1.81	0.54
6	0	4.54	4.28
7	0	5.78	5.56
1	1	6.26	6.71
2	1	7.10	7.44
3	1	7.72	7.70
4	1	* 2.17	1.96
5	1	5.22	5.29
6	1	3.58	3.84
7	1	6.90	6.46
0	2	6.82	7.04
1	2	10.76	11.37
2	2	* 1.88	1.61
3	2	5.78	6.28
4	2	5.51	5.66
5	2	7.64	7.59
6	2	4.08	3.51
7	2	4.35	3.27
0	3	3.43	3.03
1	3	4.68	5.20
2	3	5.08	5.45
3	3	7.08	6.97
4	3	* 1.81	3.55
5	3	3.65	3.56
6	3	* 2.09	1.88
0	4	8.26	8.25
1	4	4.43	4.32
2	4	4.11	4.08
3	4	* 2.38	3.17
4	4	4.03	4.59
5	4	* 1.81	1.49
6	4	4.03	3.71
0	5	6.27	6.90
1	5	* 3.40	3.53
2	5	4.84	4.24
3	5	7.01	6.77
4	5	5.85	6.03
5	5	* 2.09	2.34
6	5	6.08	5.81
0	6	3.54	3.58
1	6	3.85	3.69
2	6	* 3.59	3.57
3	6	5.26	4.77
4	6	* 3.68	4.69
5	6	6.84	7.29
0	7	3.46	2.92
1	7	5.92	5.83
2	7	* 1.81	2.91
3	7	4.05	3.48
4	7	* 1.70	1.98
0	8	4.15	4.59
1	8	* 2.24	3.75
2	8	8.20	7.95
3	8	* 2.02	2.91
0	9	3.43	2.69
1	9	4.41	4.16
HOX 2 L - 0			
1	2	8.92	8.93
2	2	* 1.35	0.22
3	2	32.53	32.25
4	2	19.37	19.35
5	2	6.09	6.40
1	3	7.23	6.93
2	3	22.88	22.70
3	3	10.15	10.23
4	3	18.72	18.86
5	3	* 1.46	0.99
0	4	20.98	21.19
1	4	28.09	27.71
2	4	12.38	12.26
3	4	4.09	4.09
4	4	6.34	6.17
5	4	3.07	3.03
1	5	13.68	13.67
2	5	26.29	25.74
3	5	2.33	2.13
4	5	10.40	10.27
5	5	4.61	4.68
0	6	6.07	5.65
1	6	10.03	10.05
2	6	13.98	14.42
3	6	8.84	8.28
4	6	* 1.39	1.64
5	6	2.86	3.12
1	7	8.09	8.04
2	7	5.04	4.94
3	7	7.74	7.68
4	7	9.11	8.50
5	7	21.03	21.30
0	8	7.35	7.23
1	8	* 1.39	1.72
2	8	12.75	12.26
3	8	5.62	5.96
4	8	7.50	7.78
5	8	* 1.05	0.51
1	9	8.70	9.20
2	9	11.06	11.20
3	9	4.67	4.16
4	9	* 3.03	2.80
5	9	3.52	3.65
0	10	7.41	7.66
1	10	10.57	10.60
2	10	5.78	5.71
3	10	8.32	7.98
4	10	4.92	4.55
5	10	2.62	1.91
1	11	3.48	3.92
2	11	4.08	3.50
3	11	* 1.27	0.92
4	11	* 1.46	1.78
0	12	2.39	2.49
1	12	* 1.14	0.26
2	12	1.45	1.86
HOX 2 L - 1			
0	0	38.44	38.88
1	0	15.24	15.18
2	0	2.42	2.29
3	0	* 1.05	0.37
4	0	3.73	3.70
5	0	12.78	13.89
0	1	14.68	14.76
1	1	23.62	23.92
2	1	39.73	39.75
3	1	22.68	22.62
4	1	11.22	11.17
5	1	8.28	8.45
1	2	27.40	27.50
2	2	7.59	7.58
3	2	16.36	16.56
4	2	11.00	10.81
5	2	7.70	7.70
0	3	2.45	1.72
1	3	21.92	21.64
2	3	12.33	12.35
3	3	4.40	4.76
4	3	9.01	8.74
5	3	6.23	6.39
0	4	13.94	13.82
1	4	9.91	9.71
2	4	4.34	3.96
3	4	11.36	11.30
4	4	17.35	17.33
5	4	4.82	5.03
0	5	* 1.23	1.77
1	5	6.90	6.75
2	5	6.73	6.59
3	5	10.31	10.17
4	5	11.48	11.70
5	5	8.12	8.44
0	6	13.80	14.06
1	6	16.12	16.00
2	6	10.28	10.21
3	6	7.15	7.26
4	6	11.45	11.72
5	6	5.98	6.25
0	7	12.18	12.50
1	7	4.32	4.32
2	7	* 1.74	1.68
3	7	3.28	3.33
4	7	6.07	5.77
5	7	8.48	8.55
0	8	* 1.01	2.56
1	8	8.75	9.12
2	8	7.45	7.30
3	8	14.26	14.15
4	8	5.10	5.58
5	8	* 1.85	2.75
0	9	4.72	5.34
1	9	3.53	2.30
2	9	11.91	12.02
3	9	* 1.81	2.10
4	9	6.08	6.03
5	9	6.87	6.61
0	10	5.57	5.94
1	10	3.55	3.87
2	10	* 1.01	2.35
3	10	4.25	4.33
4	10	2.50	3.10
5	10	* 1.10	1.28
0	11	5.59	5.62
1	11	7.29	7.04
2	11	2.93	2.99
3	11	4.45	4.28
0	12	* 1.12	1.14
1	12	3.32	3.06
HOX 2 L - 2			
0	0	28.09	28.46
1	0	23.49	23.76
2	0	2.45	2.35
3	0	0.07	0.50
4	0	21.76	21.31
5	0	* 1.39	1.02
0	1	12.87	12.91
1	1	16.69	16.85
2	1	11.83	11.74
3	1	* 1.74	2.14
4	1	2.77	2.54
5	1	5.87	5.89
0	2	9.01	9.28
1	2	13.26	13.42
2	2	* 1.42	1.32
3	2	10.66	10.39
4	2	7.26	7.47
5	2	8.97	8.72
0	3	9.65	9.45
1	3	3.89	4.07
2	3	14.05	14.21
3	3	15.43	15.06
4	3	6.85	6.65
5	3	2.57	2.21
0	4	6.55	6.88
1	4	13.23	13.42
2	4	7.51	7.64
3	4	4.55	4.46
4	4	7.80	7.90
5	4	3.66	4.09
0	5	6.65	6.85
1	5	9.53	9.75
2	5	7.32	7.43
3	5	6.14	5.82
4	5	* 1.81	2.54
5	5	3.65	3.18
0	6	3.29	2.53
1	6	* 1.43	1.56
2	6	4.71	4.59
3	6	6.17	6.61
4	6	8.65	8.68
5	6	3.62	3.15
0	7	* 1.45	1.26
1	7	11.10	11.12
2	7	5.37	5.53
3	7	8.25	8.03
4	7	7.84	7.85
5	7	4.58	5.16
0	8	5.51	5.51
1	8	4.80	4.32
2	8	* 2.31	2.20
3	8	5.62	5.24
4	8	6.16	6.36
5	8	4.44	4.57
0	9	9.42	9.61
1	9	* 1.39	1.93
2	9	7.13	7.17
3	9	* 2.68	2.25
4	9	* 2.38	2.03
5	9	3.80	3.82
0	10	3.80	3.61
1	10	4.04	4.43
2	10	6.83	6.42
3	10	6.58	6.30
0	11	* 1.12	0.36
1	11	3.32	3.42
HOX 2 L - 3			
0	0	* 1.45	1.79
1	0	* 1.43	0.48
2	0	* 1.49	2.10
3	0	* 1.56	0.87
4	0	9.44	9.25
5	0	* 1.46	0.58
0	1	10.57	10.51
1	1	7.01	6.92
2	1	7.20	7.02
3	1	7.42	7.47
4	1	* 1.53	1.98
5	1	5.10	5.01
0	2	6.84	6.96
1	2	10.93	11.04
2	2	* 1.74	1.46
3	2	5.83	5.98
4	2	5.03	5.21
5	2	7.35	7.24
0	3	3.98	3.62
1	3	4.47	5.00
2	3	5.25	5.38
3	3	6.81	6.75
4	3	* 2.34	2.86
5	3	3.84	3.57
0	4	7.99	8.04
1	4	3.84	3.95
2	4	4.22	4.01
3	4	* 2.66	3.28
4	4	4.56	4.50
5	4	* 1.56	1.17
0	5	6.90	7.22
1	5	3.58	3.55
2	5	4.12	4.00
3	5	6.83	6.66
4	5	6.08	5.62
5	5	2.67	2.10
0	6	3.49	3.24
1	6	3.97	3.67
2	6	3.68	3.59
3	6	4.76	4.77
4	6	4.55	4.16
5	6	6.76	6.59
0	7	3.31	2.75
1	7	5.90	5.60
2	7	3.23	2.69
3	7	3.70	3.21
4	7	* 1.21	1.65
0	8	4.33	4.18
1	8	* 2.89	3.51
2	8	7.63	7.63
3	8	2.97	2.87
0	9	3.03	2.68
1	9	3.94	3.91
2	9	4.68	4.26
HOX 2 L - 4			
0	0	* 1.30	1.83
1	0	3.99	3.93
2	0	* 1.59	1.75
3	0	2.84	3.06
4	0	2.07	2.06
0	1	4.00	4.06
1	1	* 1.36	1.54
2	1	5.87	5.78
3	1	* 1.81	2.29
4	1	* 1.21	0.73
0	2	* 1.70	2.10
1	2	4.00	3.88
2	2	* 2.72	2.61
3	2	1.27	0.66
4	2	1.42	1.26
0	3	5.40	5.56
1	3	4.19	4.70
2	3	3.60	3.89
3	3	4.67	4.05
4	3	3.30	2.91
0	4	4.04	4.35
1	4	3.06	3.21
2	4	* 1.42	0.72
3	4	4.09	3.77
0	5	* 1.41	1.09
1	5	* 1.21	0.69
2	5	* 1.99	2.07
0	6	3.31	2.99
HOX 1 L - 0			
2	0	6.67	6.76
4	0	22.43	23.35
6	0	4.54	4.83
8	0	7.27	7.27
2	1	10.72	10.58
3	1	32.47	36.12
4	1	5.52	5.56
5	1	6.74	6.83
6	1	4.98	4.96
7	1	11.00	11.24
8	1	1.43	1.73
9	1	3.72	3.65
1	2	9.96	9.39
2	2	* 0.32	0.35
3	2	30.41	31.74
4	2	20.02	19.42
5	2	6.27	6.73
6	2	3.94	3.91
7	2	12.60	12.20
8	2	5.15	5.47
9	2	3.54	3.71
1	3	7.72	6.92
2	3	23.22	22.99
3	3	11.17	10.79
4	3	20.04	19.06
5	3	1.11	1.46
6	3	10.87	10.51
7	3	12.17	11.98
8	3	1.50	1.15
9	3	1.57	1.55
0	4	20.02	21.75
1	4	26.34	27.69
2	4	13.04	12.30
3	4	3.96	4.09
4	4	6.42	6.20
5	4	3.85	3.24
6	4	* 0.26	0.62
7	4	* 0.36	0.44
8	4	0.54	0.59
9	4	2.12	2.09
1	5	13.93	13.79
2	5	26.47	25.61
3	5	2.18	1.07
4	5	11.06	10.61
5	5	4.65	4.06
6	5	2.37	2.62
7	5	4.73	5.12
8	5	3.28	3.59
0	6	6.07	6.36
1	6	9.97	9.74
2	6	14.85	14.00
3	6	8.54	8.50
4	6	0.88	1.17
5	6	2.83	2.91
6	6	1.51	1.35
7	6	4.26	4.39
8	6	4.46	4.49
1	7	8.14	7.95
2	7	5.16	5.19
3	7	8.06	8.13
4	7	9.44	9.19
5	7	22.04	21.70
6	7	5.82	5.82
7	7	3.12	3.12
8	7	2.58	2.55
0	8	7.48	7.23
1	8	1.31	1.42
2	8	12.72	12.27
3	8	6.19	5.99
4	8	7.59	7.58
5	8	0.93	0.56
6	8	1.05	1.21
7	8	2.62	2.65
1	9	8.87	9.25
2	9	11.36	11.24
3	9	4.51	4.34
4	9	2.94	3.03
5	9	3.49	3.57
6	9	3.81	3.94
0	10	7.50	7.61
1	10	10.70	10.70
2	10	5.71	5.65
3	10	8.54	8.36
4	10	4.78	4.78
5	10	2.23	2.48
1	11	3.66	3.81
2	11	3.87	3.85
3	11	0.95	0.75
4	11	1.20	1.19
0	12	2.39	2.66
1	12	0.75	0.17
2	12	2.23	2.10
HOX 1 L - 1			
2	0	2.17	2.02
3	0	0.09	0.45
4	0	3.47	3.69
5	0	13.60	13.70
6	0	1.88	1.94
7	0	3.38	3.84
8	0	0.43	0.89
9	0	3.03	2.81
2	1	36.20	39.70
3	1	22.32	23.09
4	1	11.55	11.31
5	1	8.67	8.70
6	1	15.39	15.43
7	1	6.44	6.84
8	1	6.70	6.77
9	1	5.13	5.08
1	2	26.53	27.53
2	2	7.70	7.58
3	2	16.71	16.60
4	2	11.42	11.08
5	2	8.05	8.00
6	2	5.62	5.76
7	2	7.22	7.09
8	2	1.70	1.75
9	2	5.97	5.96
0	3	2.03	2.07
1	3	21.39	21.53
2	3	12.75	12.22
3	3	4.50	4.50
4	3	9.23	9.03
5	3	6.43	6.44
6	3	5.97	5.97
7	3	7.41	7.76
8	3	4.60	4.88
9	3	3.79	3.81
0	4	13.65	13.27
1	4	9.98	9.77
2	4	4.30	4.11
3	4	11.76	11.52
4	4	18.31	17.57
5	4	5.26	5.40
6	4	7.05	7.08
7	4	4.28	4.18
8	4	6.12	6.05
0	5	1.00	1.42
1	5	6.84	6.78
2	5	6.64	6.69
3	5	10.38	10.22
4	5	12.18	12.02
5	5	8.52	8.75
6	5	5.76	6.04
7	5	4.85	4.97
8	5	0.60	0.45
0	6	14.14	14.16
1	6	16.05	15.92
2	6	10.25	10.11
3	6	7.33	7.20
4	6	12.11	11.92
5	6	6.24	6.37
6	6	4.99	5.12
7	6	4.68	4.75
8	6	6.34	6.20
0	7	12.09	12.32
1	7	4.24	4.11
2	7	1.65	1.52
3	7	3.10	3.40
4	7	5.98	5.97
5	7	8.60	8.51
6	7	3.46	3.48
7	7	2.40	2.39
0	8	2.64	2.51
1	8	8.76	9.14
2	8	7.23	7.22
3	8	14.28	14.42
4	8	5.25	5.39
5	8	2.81	2.98
6	8	3.21	3.29
7	8	9.03	8.51
0	9	4.76	5.25
1	9	2.17	2.24
2	9	12.11	12.16
3	9	2.44	2.50
4	9	6.34	6.28
5	9	6.80	6.81
6	9	6.23	6.18
0	10	5.54	5.82
1	10	3.71	3.69
2	10	2.59	2.49
3	10	4.56	4.55
4	10	3.16	3.13
5	10	1.15	1.15
0	11	5.91	5.86
1	11	7.99	8.01
2	11	3.11	3.19
3	11	4.54	4.50
4	11	0.72	0.74
0	12	1.02	1.34
1	12	3.35	3.34
HOX 1 L - 2			
2	0	2.32	2.30
3	0	8.98	8.92
4	0	23.70	21.87
6	0	9.59	9.12
7	0	4.32	4.36
8	0	4.78	4.82
1	1	17.12	16.63
2	1	12.29	11.95
3	1	2.11	2.11
4	1	2.84	2.91
5	1	5.70	5.95
6	1	4.56	4.90
7	1	3.11	3.16
8	1	5.14	5.34
0	2	8.87	8.74
1	2	13.63	13.37
2	2	1.08	1.32
3	2	11.16	10.83
4	2	7.40	7.56
5	2	9.21	9.40
6	2	6.52	6.51
7	2	6.79	6.80
8	2	3.43	3.29
0	3	9.88	9.75
1	3	3.99	4.03
2	3	14.60	14.32
3	3	16.01	15.18
4	3	6.77	6.69
5	3	2.11	2.32
6	3	6.96	7.01
7	3	6.42	6.38
8	3	3.34	3.51
0	4	6.76	6.78
1	4	13.43	12.87
2	4	7.59	7.65
3	4	4.36	4.36
4	4	8.15	7.89
5	4	3.83	4.07
6	4	5.32	5.37
7	4	2.40	2.42
8	4	3.38	3.55
0	5	6.57	6.59
1	5	9.64	9.64
2	5	7.29	7.48
3	5	5.79	5.89
4	5	2.28	2.48
5	5	3.37	3.34
6	5	7.84	7.59
7	5	1.14	1.15
0	6	2.92	2.59
1	6	1.56	1.60
2	6	4.46	4.58
3	6	6.37	6.59
4	6	8.55	8.72
5	6	2.93	3.00
6	6	7.48	7.33
7	6	1.59	1.61
0	7	0.74	1.10
1	7	11.05	11.02
2	7	5.41	5.53
3	7	8.18	8.26
4	7	8.86	8.01
5	7	5.15	5.21
6	7	4.93	5.20
0	8	5.43	5.61
1	8	4.42	4.37
2	8	2.23	2.14
3	8	5.24	5.36
4	8	6.56	6.65
5	8	4.33	4.49
0	9	9.83	9.77
1	9	1.99	1.88
2	9	7.01	7.12
3	9	2.46	2.52
4	9	1.81	1.98
5	9	2.64	3.85
0	10	3.22	3.96
1	10	3.84	4.63
2	10	5.66	6.61
3	10	4.46	6.49
0	11	* 0.20	0.35
HOX 1 L - 3			
2	0	2.13	2.13
3	0	0.93	0.91
4	0	9.76	9.39
5	0	0.67	0.75
6	0	4.24	4.48
7	0	5.63	5.41
1	1	6.67	6.81
2	1	7.42	7.36
3	1	7.72	7.56
4	1	1.87	2.11
5	1	5.11	5.14
6	1	3.82	3.91
7	1	6.50	6.23
0	2	6.26	6.29
1	2	11.81	10.92
2	2	1.34	1.52
3	2	6.11	6.23
4	2	5.24	5.00
5	2	7.76	7.43
6	2	3.28	3.37
0	3	3.44	3.56
1	3	4.71	4.95
2	3	5.19	5.29
3	3	7.01	6.94
4	3	2.87	2.98
5	3	3.75	3.61
6	3	1.73	1.69
0	4	7.98	8.11
1	4	3.56	3.82
2	4	3.87	3.98
3	4	2.93	3.01
4	4	4.45	4.53
5	4	1.21	1.13
6	4	3.95	3.91
0	5	7.05	7.10
1	5	3.42	3.32
2	5	3.93	3.89
3	5	6.79	6.68
4	5	5.81	5.77
5	5	2.13	2.24
0	6	3.45	3.47
1	6	3.62	3.66
2	6	3.40	3.30
3	6	4.60	4.55
4	6	4.40	4.43
5	6	6.95	6.72
0	7	2.84	2.77
1	7	5.84	5.73
2	7	2.76	2.68
3	7	3.25	3.33
4	7	1.76	1.75
0	8	4.11	4.13
1	8	3.45	3.59
2	8	7.91	7.78
3	8	2.05	2.98
0	9	2.44	2.63
1	9	3.36	3.88
HOX 2 L - 0			
2	0	6.46	6.58
4	0	23.37	23.20
6	0	4.66	4.74
8	0	7.21	7.43
2	1	10.06	10.20
3	1	37.27	36.73
4	1	5.43	5.49
5	1	6.76	6.85
6	1	5.11	5.18
7	1	10.49	10.64
8	1	1.69	1.58
9	1	3.79	3.61
2	2	* 0.75	0.61
3	2	32.15	32.09
4	2	19.93	19.33
5	2	6.30	6.57
6	2	4.19	4.24
7	2	12.07	12.32
8	2	5.06	5.24
9	2	3.66	3.71
2	3	23.31	22.03
3	3	10.52	10.49
4	3	19.11	19.04
5	3	2.21	1.47
6	3	10.23	10.48
7	3	11.44	11.69
8	3	1.34	1.14
9	3	1.54	1.60
2	4	12.48	12.43
3	4	4.07	4.13
4	4	6.66	6.46
5	4	3.19	3.03
6	4	* 0.44	0.54
7	4	* 0.43	0.30
8	4	0.77	0.59
9	4	2.21	2.10
2	5	26.53	25.63
3	5	2.27	1.02
4	5	10.71	10.05
5	5	4.66	5.02
6	5	2.60	2.49
7	5	4.69	4.95
8	5	3.34	3.33
HOX 2 L - 1			
1	0	15.62	15.46
2	0	2.28	2.16
3	0	* 0.38	0.62
4	0	3.56	4.05
5	0	13.63	13.64
6	0	2.01	1.79
7	0	3.46	3.67
8	0	* 0.44	0.13
9	0	3.08	2.00
0	1	14.79	15.19
1	1	23.76	24.02
2	1	39.13	40.06
3	1	23.24	23.25
4	1	11.60	11.44
5	1	8.70	8.04
6	1	15.20	15.37
7	1	6.45	6.56
8	1	6.66	6.63
9	1	5.19	5.04
1	2	26.76	27.25
2	2	7.76	7.66
3	2	16.93	16.72
4	2	11.57	11.05
5	2	8.23	8.16
6	2	5.71	5.55
7	2	7.18	7.14
8	2	1.70	1.67
9	2	5.99	5.93
0	3	2.20	2.37
1	3	21.30	21.94
2	3	12.74	12.55
3	3	4.65	4.67
4	3	9.34	9.20
5	3	6.67	6.57
6	3	6.15	6.09
7	3	7.36	7.50
8	3	4.61	4.71
9	3	4.04	3.85
0	4	13.70	13.84
1	4	9.85	9.86
2	4	4.42	4.30
3	4	11.62	11.61
4	4	17.70	17.71
5	4	5.39	5.42
6	4	7.05	7.09
7	4	4.39	4.30
8	4	6.11	6.03
0	5	1.12	1.19
1	5	6.99	6.07
2	5	6.79	6.67
3	5	10.48	10.48
4	5	11.93	12.24
5	5	8.52	9.15
6	5	5.05	5.99
7	5	5.03	5.03
8	5	* 0.40	0.29
HOX 2 L - 2			
0	0	20.77	20.98
1	0	23.68	23.61
2	0	2.78	2.50
3	0	8.96	8.77
4	0	22.73	22.22
5	0	1.68	0.97
6	0	9.63	9.20
7	0	4.56	4.32
8	0	5.00	4.01
0	1	12.97	13.15
1	1	16.77	17.01
2	1	12.06	12.17
3	1	2.11	2.05
4	1	3.59	3.03
5	1	5.89	5.97
6	1	4.83	4.91
7	1	3.11	3.20
8	1	5.17	5.23
0	2	9.30	9.36
1	2	13.70	13.71
2	2	1.15	1.47
3	2	11.13	11.05
4	2	7.66	7.01
5	2	9.13	9.37
6	2	6.72	6.54
7	2	6.93	6.83
8	2	3.74	3.42
0	3	10.00	9.04
1	3	4.65	4.45
2	3	14.58	14.74
3	3	15.69	15.42
4	3	7.04	7.13

Table 7 (*cont.*)

H	K	FO	FC
5	3	2.25	2.21
6	3	7.23	6.94
7	3	6.50	6.42
8	3	3.53	3.39
0	4	7.04	7.15
1	4	13.41	13.36
2	4	7.85	7.82
3	4	4.65	4.62
4	4	8.39	8.30
5	4	4.02	4.15
6	4	5.51	5.61
7	4	2.56	2.67
8	4	3.63	3.62
0	5	6.53	6.86
1	5	9.88	9.82
2	5	7.60	7.54
3	5	6.39	6.56
4	5	2.26	2.86
5	5	3.58	3.31
6	5	8.10	7.90
7	5	1.64	1.50

NOX 2 L · 3

H	K	FO	FC
0	0	1.72	1.93
1	0	1.24	0.63
2	0	2.22	2.28
3	0	0.97	1.11
4	0	9.66	9.51
5	0	0.46	0.40
6	0	4.57	4.51
7	0	5.75	5.63
0	1	10.69	10.97
1	1	7.10	7.08
2	1	7.54	7.67
3	1	7.83	7.88
4	1	2.04	2.17
5	1	5.33	5.28
6	1	4.05	4.24
7	1	6.51	6.36
0	2	7.10	6.83
1	2	11.23	11.26
2	2	1.86	1.66
3	2	6.21	6.42
4	2	5.51	5.54
5	2	7.95	7.63
6	2	3.63	3.68
7	2	3.37	3.21
0	3	4.08	3.86
1	3	5.00	5.21
2	3	5.52	5.48
3	3	7.14	7.46
4	3	3.19	3.16
5	3	4.05	3.90
6	3	1.94	1.84
0	4	8.26	8.57
1	4	3.78	4.18
2	4	4.03	4.13
3	4	3.23	3.15
4	4	4.71	4.81
5	4	1.28	1.48
6	4	4.15	4.09
0	5	7.63	7.47
1	5	3.77	3.86
2	5	4.17	4.16
3	5	7.33	7.12
4	5	6.10	6.25
5	5	2.40	2.23
6	5	6.11	6.26

NOX 2 L · 4

H	K	FO	FC
0	0	1.63	1.46
1	0	4.06	4.18
2	0	2.21	2.21
3	0	3.23	3.21
4	0	2.31	2.25
0	1	3.95	4.34
1	1	1.75	1.58
2	1	6.38	6.27
3	1	2.32	2.30
4	1	0.73	0.35
0	2	2.61	2.54
1	2	4.28	4.25
2	2	2.51	2.91
3	2	1.21	0.71
4	2	1.25	1.21
0	3	5.72	5.84
1	3	4.35	4.93
2	3	4.25	4.20
3	3	4.43	4.48
4	3	3.09	3.15
0	4	4.42	4.30
1	4	3.31	3.45
2	4	1.04	0.58
3	4	4.07	4.20
0	5	1.82	2.72
1	5	* 0.64	0.96
2	5	2.34	2.53

DOX 1 L · 0

H	K	FO	FC
2	0	6.47	6.60
4	0	22.93	22.72
6	0	4.58	4.85
8	0	7.28	7.32
2	1	9.96	10.18
3	1	36.36	36.59
4	1	5.78	5.67
5	1	6.87	6.90
6	1	5.22	5.03
7	1	10.73	10.87
8	1	1.81	1.51
9	1	3.86	3.69
1	2	9.29	9.32
2	2	* 0.42	0.98
3	2	31.32	31.65
4	2	19.93	19.41
5	2	6.29	6.53
6	2	4.83	4.80
7	2	12.30	12.31
8	2	5.12	5.45
9	2	3.62	3.73
1	3	7.74	7.45
2	3	22.37	22.55
3	3	10.57	10.66
4	3	19.14	18.91
6	3	10.46	10.47
7	3	11.75	11.65
8	3	1.12	0.99
9	3	1.46	1.64
0	4	20.92	21.27
1	4	27.41	27.64
2	4	12.35	12.41
3	4	4.04	4.22
4	4	6.56	6.43
5	4	3.16	2.93
6	4	* 0.56	0.39
8	4	* 0.52	0.57
9	4	2.23	2.17

H	K	FO	FC
1	5	13.65	13.72
2	5	26.19	25.94
3	5	1.99	1.69
4	5	10.54	10.41
5	5	4.73	4.92
6	5	2.49	2.79
7	5	4.76	4.99
8	5	3.32	3.65
0	6	6.20	6.14
1	6	10.07	9.86
2	6	14.41	14.39
3	6	8.35	8.18
4	6	* 0.56	1.05
5	6	2.77	2.87
6	6	1.49	1.57
7	6	4.44	4.19
8	6	4.51	4.45
1	7	8.22	7.96
2	7	5.25	5.34
3	7	7.92	8.11
4	7	9.15	8.97
5	7	21.81	21.94
6	7	5.92	5.84
7	7	3.25	3.20
8	7	2.64	2.37
0	8	7.80	7.44
1	8	1.65	1.73
2	8	12.51	12.53
3	8	6.34	6.09
4	8	7.63	7.80
5	8	0.77	0.43
6	8	0.85	1.14
7	8	2.82	2.66
1	9	8.92	9.43
2	9	11.35	11.40
3	9	4.53	4.40
4	9	3.12	2.89
5	9	3.64	3.54
6	9	4.03	3.94
0	10	7.87	7.91
1	10	10.76	10.98
2	10	5.81	5.76
3	10	8.36	8.35
4	10	5.05	5.16
5	10	2.41	2.69
1	11	3.94	3.97
2	11	4.05	3.85
3	11	* 0.65	0.80
4	11	1.36	1.54
0	12	2.43	2.49
1	12	* 0.45	0.06
2	12	2.45	2.13

DOX 1 L · 1

H	K	FO	FC
2	0	2.27	2.10
3	0	* 0.43	0.57
4	0	3.44	3.85
5	0	13.26	13.62
6	0	1.94	2.09
7	0	3.60	3.95
8	0	* 0.52	0.89
9	0	2.87	2.82
0	1	15.07	15.32
2	1	39.81	39.75
3	1	22.99	23.01
4	1	11.40	11.32
5	1	8.64	8.73
6	1	14.96	15.32
7	1	6.44	6.64
8	1	6.69	6.59
9	1	5.17	5.07
1	2	27.19	27.47
2	2	7.66	7.48
3	2	16.70	16.68
4	2	11.31	11.04
5	2	7.99	7.95
6	2	5.54	5.55
7	2	7.19	7.15
8	2	1.69	1.66
9	2	5.84	5.88
0	3	2.21	2.04
1	3	21.63	21.42
2	3	12.65	12.24
3	3	4.65	4.65
4	3	9.24	9.20
5	3	6.50	6.51
6	3	6.00	5.93
7	3	7.18	7.44
8	3	4.58	4.75
9	3	3.68	3.78
0	4	14.11	13.79
1	4	9.97	10.07
2	4	4.42	4.16
3	4	11.43	11.33
4	4	17.55	17.56
5	4	5.37	5.48
6	4	7.02	7.16
7	4	4.28	4.09
8	4	6.25	6.11
0	5	1.27	1.54
1	5	7.02	6.75
2	5	6.91	6.98
3	5	10.40	10.36
4	5	11.60	11.81
5	5	8.61	8.79
6	5	5.64	6.01
7	5	4.88	4.82
8	5	* 0.71	0.36
0	6	14.48	14.25
1	6	15.83	15.98
2	6	10.42	10.32
3	6	7.31	7.36
4	6	11.88	12.01
5	6	6.14	6.31
6	6	4.98	5.19
7	6	4.70	4.64
8	6	6.21	6.24
0	7	12.41	12.60
1	7	4.37	4.26
2	7	1.95	1.63
3	7	3.17	3.38
4	7	6.11	5.87
5	7	8.41	8.71
6	7	3.55	3.57
7	7	2.48	2.47
0	8	2.79	2.34
1	8	8.99	9.46
2	8	7.39	7.30
3	8	14.08	14.57
4	8	5.23	5.39
5	8	3.05	3.08
6	8	3.38	3.32
7	8	8.72	8.45
0	9	4.96	5.46
1	9	2.13	2.27
2	9	12.30	12.56
3	9	2.47	2.48
4	9	6.30	6.27
5	9	6.76	6.73
6	9	6.10	6.25

H	K	FO	FC
0	10	5.81	5.88
1	10	3.65	3.89
2	10	2.92	2.46
3	10	4.71	4.56
4	10	3.21	3.24
5	10	1.44	1.09
0	11	6.09	5.96
1	11	8.34	8.18
2	11	3.14	3.03
3	11	4.65	4.56
4	11	* 0.61	0.84
0	12	1.08	1.30
1	12	3.40	3.34

DOX 1 L · 2

H	K	FO	FC
2	0	2.41	2.46
3	0	8.71	8.81
4	0	22.19	21.75
6	0	9.06	8.98
7	0	4.50	4.42
8	0	4.80	4.66
1	1	16.67	16.71
2	1	11.78	11.80
3	1	2.28	2.11
4	1	2.75	2.90
5	1	5.57	5.93
6	1	4.68	4.88
7	1	3.35	3.19
8	1	5.10	5.22
0	2	9.00	9.18
1	2	13.33	13.32
2	2	1.12	1.46
3	2	10.91	10.72
4	2	7.34	7.65
5	2	8.94	9.10
6	2	6.52	6.56
7	2	6.88	6.83
8	2	3.62	3.27
0	3	9.72	9.79
1	3	4.14	4.25
2	3	14.39	14.32
3	3	15.54	15.35
4	3	6.73	6.91
5	3	2.82	2.36
6	3	6.77	6.76
7	3	6.19	6.32
8	3	3.38	3.34
0	4	6.90	6.92
1	4	13.26	13.13
2	4	7.67	7.51
3	4	4.62	4.51
4	4	8.29	8.13
5	4	4.02	4.12
6	4	5.42	5.43
7	4	2.46	2.42
8	4	3.47	3.55
0	5	6.68	6.62
1	5	9.63	9.77
2	5	7.42	7.61
3	5	5.99	6.13
4	5	2.23	2.59
5	5	3.54	3.22
6	5	7.92	7.70
7	5	1.57	1.23
0	6	3.00	2.20
1	6	1.63	1.45
2	6	4.59	4.53
3	6	6.47	6.81
4	6	8.46	8.66
5	6	3.09	3.11
6	6	7.51	7.43
7	6	1.64	1.50
0	7	* 0.55	1.37
1	7	10.96	11.21
2	7	5.61	5.82
3	7	8.24	8.24
4	7	8.05	7.97
5	7	5.24	5.23
6	7	5.09	5.24
0	8	5.66	5.81
1	8	4.66	4.53
2	8	2.38	2.19
3	8	5.52	5.41
4	8	6.71	6.76
5	8	4.73	4.62
6	8	2.40	2.44
0	9	9.92	9.82
1	9	2.19	2.05
2	9	7.27	7.17
3	9	2.84	2.57
4	9	2.27	1.76
5	9	4.14	4.04
0	10	4.14	4.09
1	10	4.67	4.78
2	10	6.98	6.83
3	10	6.55	6.47
0	11	* 0.53	0.41
1	11	3.85	3.63

DOX 1 L · 3

H	K	FO	FC
2	0	2.02	2.44
3	0	1.05	1.12
4	0	9.19	9.10
5	0	1.70	0.32
6	0	4.39	4.20
7	0	5.75	5.64
1	1	5.98	6.49
2	1	7.14	7.18
3	1	7.41	7.61
4	1	1.99	2.14
5	1	5.07	5.15
6	1	3.93	3.78
7	1	6.43	6.19
0	2	6.68	6.79
1	2	10.75	10.99
2	2	1.42	1.54
3	2	6.11	6.31
4	2	5.29	5.23
5	2	7.49	7.47
6	2	3.45	3.31
7	2	3.20	3.13
0	3	4.04	4.03
1	3	4.06	5.01
2	3	5.73	5.31
3	3	6.93	6.90
4	3	3.34	3.07
5	3	3.95	3.55
6	3	1.96	1.83
0	4	8.03	8.12
1	4	3.68	4.37
2	4	4.02	3.96
3	4	3.07	3.19
4	4	4.47	4.52
5	4	1.22	1.30
6	4	4.10	3.9[illegible]
0	5	6.84	7.00
1	5	3.47	3.48
2	5	4.08	4.07
3	5	6.91	6.72
4	5	5.89	5.86

H	K	FO	FC
5	5	2.18	2.33
6	5	5.79	5.71
0	6	3.60	3.64
1	6	3.71	3.64
2	6	3.62	3.32
3	6	4.77	4.70
4	6	4.64	4.42
5	6	7.02	6.88
0	7	2.95	2.74
1	7	6.09	5.71
2	7	2.95	2.92
3	7	3.36	3.41
4	7	1.99	1.88
0	8	4.28	4.19
1	8	3.69	3.66
2	8	8.09	7.95
3	8	3.03	2.87
0	9	2.90	2.72
1	9	4.19	4.28

DOX 2 H · 0

K	L	FO	FC
0	2	20.49	20.68
0	3	1.86	2.34
0	4	2.06	1.65
1	1	14.67	15.01
1	2	13.13	13.32
1	3	10.94	11.04
1	4	4.16	4.39
2	2	9.38	9.41
2	3	7.04	6.92
2	4	2.43	2.58
3	1	2.26	2.17
3	2	10.02	9.79
3	3	4.45	4.17
3	4	5.62	5.97
4	1	14.04	14.75
4	2	7.29	7.30
4	3	8.26	8.62
4	4	4.43	4.40
5	1	1.15	1.52
5	2	6.67	6.85
5	3	7.47	7.51
5	4	1.92	2.20

DOX 2 H · 1

K	L	FO	FC
0	1	15.55	15.59
0	2	23.35	23.73
0	3	0.97	0.61
0	4	4.01	4.14
1	1	23.65	24.25
1	2	17.[illegible]	16.98
1	3	7.15	7.06
1	4	1.78	1.63
2	2	13.76	13.91
2	3	11.30	11.21
2	4	4.23	4.22
3	1	22.35	22.28
3	2	4.90	4.55
3	3	5.01	5.18
3	4	4.94	4.97
4	1	10.05	10.01
4	2	13.57	13.41
4	3	3.78	4.33
4	4	3.29	3.44
5	1	7.03	7.04
5	2	9.77	9.06
5	3	3.66	3.92
5	4	* 0.28	1.01

DOX 2 H · 2

K	L	FO	FC
0	1	2.19	2.28
0	2	2.79	2.50
0	3	2.23	2.15
0	4	2.00	2.21
1	0	10.04	10.43
1	2	12.15	12.23
1	3	7.59	7.55
1	4	6.06	5.95
2	0	0.70	0.64
2	1	7.01	7.76
2	2	1.31	1.57
2	3	1.56	1.55
2	4	2.88	2.92
3	0	24.23	23.49
3	1	12.88	12.69
3	2	14.63	14.87
3	3	5.47	5.50
3	4	3.95	4.15
4	0	12.54	12.25
4	1	4.42	4.24
4	2	7.80	7.74
4	3	4.09	4.24
4	4	* 0.59	0.65
5	0	27.87	27.33
5	1	6.88	6.89
5	2	7.61	8.03
5	3	4.20	4.23
5	4	2.44	2.44

DOX 2 H · 3

K	L	FO	FC
0	1	* 0.28	0.31
0	2	8.88	8.77
0	3	1.11	1.09
0	4	3.25	3.28
1	1	23.15	23.38
1	2	2.20	2.08
1	3	7.79	7.89
1	4	2.39	2.38
2	1	17.00	16.87
2	2	11.28	11.08
2	3	6.36	6.46
2	4	* 0.46	0.68
3	0	10.74	10.70
3	1	4.70	4.65
3	2	15.79	15.48
3	3	7.29	7.36
3	4	4.30	4.43
4	0	4.11	4.12
4	1	11.68	11.67
4	2	4.79	4.65
4	3	3.15	3.06
4	4	4.15	4.31
5	0	2.36	2.11
5	1	10.54	10.60
5	2	6.05	6.50
5	3	7.08	7.17

DOX 2 H · 4

K	L	FO	FC
0	0	22.34	22.90
0	1	3.54	3.82
0	2	22.05	21.86
0	3	9.28	9.37
0	4	2.31	2.12
1	0	5.74	5.65
1	1	11.70	11.62
1	2	3.61	3.07
1	3	2.04	2.08

K	L	FO	FC
1	4	0.66	0.81
2	0	20.32	19.82
2	1	11.49	11.09
2	2	7.68	7.88
2	3	5.50	5.50
2	4	1.25	1.18
3	0	19.66	19.30
3	1	9.49	9.48
3	2	6.98	7.12
3	3	3.10	3.16
3	4	3.04	3.13
4	0	6.08	6.62
4	1	10.04	10.44
4	2	8.59	8.30
4	3	4.83	4.81
5	0	10.82	10.84
5	1	11.95	12.08
5	2	2.29	2.81
5	3	6.08	6.16

DOX 2 H · 5

K	L	FO	FC
0	1	13.34	13.64
0	2	1.63	1.06
0	3	0.59	0.49
1	0	6.94	7.11
1	1	8.74	8.84
1	2	5.09	5.97
1	3	5.34	5.26
2	0	6.38	6.54
2	1	8.27	8.18
2	2	9.04	9.37
2	3	7.76	7.64
3	0	2.10	1.10
3	1	6.79	6.63
3	2	2.32	2.33
3	3	3.97	3.90
4	0	3.25	3.17
4	1	5.53	5.42
4	2	3.91	4.07
4	3	1.38	1.28
5	0	4.73	4.85
5	1	8.49	8.80
5	2	3.63	3.61
5	3	2.39	2.39

DOX 2 H · 6

K	L	FO	FC
0	0	4.63	4.60
0	1	2.02	1.88
0	2	9.43	9.36
0	3	4.43	4.46
1	0	5.25	5.22
1	1	15.16	15.30
1	2	4.83	4.87
1	3	4.11	3.95
2	0	4.14	4.09
2	1	5.68	5.62
2	2	6.75	6.54
2	3	3.63	3.70
3	0	10.63	10.98
3	1	6.19	6.09
3	2	7.20	7.00
3	3	1.92	1.81
4	0	* 0.33	0.23
4	1	7.19	7.24
4	2	5.60	5.65
4	3	4.26	4.20
5	0	2.44	2.71
5	1	5.87	6.09
5	2	8.06	8.03
5	3	5.92	6.12

DOX 2 H · 7

K	L	FO	FC
0	1	3.61	3.78
0	2	4.46	4.46
0	3	5.70	5.78
1	0	10.57	10.83
1	1	6.49	6.64
1	2	3.25	3.15
1	3	6.54	6.41
2	0	12.33	12.37
2	1	7.37	7.41
2	2	6.96	6.92
2	3	3.34	3.24
3	0	11.64	11.70
3	1	7.46	7.53
3	2	6.56	6.56
4	0	0.89	0.69
4	1	4.47	4.43
4	2	2.65	2.58
5	0	4.85	4.85
5	1	5.01	5.14
5	2	1.51	1.52

DOX 2 H · 8

K	L	FO	FC
0	0	7.10	7.10
0	1	* 0.33	0.05
0	2	4.94	4.76
1	0	1.58	1.46
1	1	6.78	6.77
1	2	5.30	5.32
2	0	5.29	5.37
2	1	1.77	1.63
2	2	3.79	3.43
3	0	1.33	1.32
3	1	4.74	4.64
3	2	3.60	3.58
4	0	* 0.31	0.48
4	1	6.32	6.27
4	2	3.67	3.70
5	0	3.30	3.61
5	1	0.53	0.33

DOX 2 H · 9

K	L	FO	FC
0	1	3.06	2.85
1	0	3.86	3.69
1	1	5.24	5.07
2	0	3.64	3.69
2	1	6.08	5.84
3	0	1.56	1.72
3	1	3.92	3.83
4	0	2.23	2.40

HOX NEUTRON H · 0

K	L	FO	FC
0	2	4.36	4.31
0	3	1.66	1.73
0	4	2.00	1.98
0	5	0.94	1.09
1	1	2.02	2.02
1	2	2.89	2.89
1	3	4.30	4.34
1	4	1.75	1.69
2	0	4.05	4.18
2	1	3.36	3.37
2	2	4.29	4.39
2	3	1.01	0.97
2	4	0.40	0.18
3	1	1.08	1.11

K	L	FO	FC
3	2	2.82	2.95
3	3	0.99	0.97
3	4	3.90	3.76
4	0	2.20	2.14
4	1	3.69	3.75
4	2	1.36	1.35
4	3	3.40	3.40
4	4	2.50	2.39
5	1	0.37	0.45
5	2	0.14	0.19
5	3	3.21	3.22
5	4	0.84	0.88
6	0	1.18	1.15
6	1	3.91	4.08
6	2	1.53	1.50
6	3	1.12	1.09
6	4	1.47	1.38
7	1	1.51	1.51
7	2	0.15	0.27
7	3	0.12	0.07
7	4	2.92	2.81
7	5	0.20	3.58
8	0	2.45	2.45
8	1	0.36	0.47
8	2	1.39	1.41
8	3	1.59	1.52
8	4	0.36	0.56
9	1	0.66	0.58
9	2	2.89	2.83
9	3	1.23	1.27
10	0	2.73	2.67
10	1	1.66	1.69
10	2	0.83	0.81
10	3	1.11	1.11

HOX NEUTRON H · 1

K	L	FO	FC
0	1	1.71	1.74
0	2	5.77	5.69
0	3	3.45	3.40
0	4	3.39	3.23
1	0	3.77	4.04
1	1	3.59	3.84
1	2	4.79	4.87
1	3	4.15	4.05
1	4	2.14	2.09
1	5	1.15	1.27
2	0	0.71	0.65
2	1	2.16	2.23
2	2	2.59	2.64
2	3	5.10	4.99
2	4	2.00	1.93
2	5	1.30	1.29
3	0	1.50	1.51
3	1	1.74	1.77
3	2	1.39	1.36
3	3	2.89	2.84
3	4	2.42	2.36
4	0	2.29	2.32
4	1	2.02	1.99
4	2	2.97	2.91
4	3	1.78	1.83
4	4	1.99	1.98
5	0	2.17	2.12
5	1	3.31	3.39
5	2	2.23	2.27
5	3	1.71	1.67
5	4	0.36	0.37
6	0	0.67	0.66
6	1	3.01	3.03
6	2	2.28	2.27
6	3	1.51	1.52
6	4	1.17	1.06
7	0	5.70	5.73
7	1	1.44	1.42
7	2	4.12	4.15
7	3	2.06	2.11
7	4	1.35	1.38
7	5	0.79	0.93
8	0	0.48	0.35
8	1	4.32	4.26
8	2	0.97	0.92
8	3	1.63	1.63
9	0	3.98	4.03
9	1	2.68	2.58
9	2	0.63	0.66
9	3	1.30	1.34
10	0	4.78	4.67
10	1	0.91	0.91
10	2	2.04	2.04
10	3	0.42	0.52

HOX NEUTRON H · 2

K	L	FO	FC
0	0	0.12	0.05
0	1	0.12	0.13
0	2	0.12	0.03
0	3	0.63	0.78
0	4	0.12	0.07
1	0	1.15	1.13
1	1	2.84	2.95
1	2	1.72	1.74
1	3	2.25	2.27
1	4	2.22	2.20
2	1	0.63	0.63
2	2	1.00	0.99
2	3	0.66	0.60
2	4	1.81	1.81
3	0	2.42	2.41
3	1	3.44	3.46
3	2	3.62	3.69
3	3	0.97	0.96
3	4	1.83	1.84
4	0	1.58	1.53
4	1	0.65	0.60
4	2	1.10	1.07
4	3	2.45	2.35
4	4	0.67	0.74
5	0	5.16	5.25
5	1	1.97	1.96
5	2	3.07	3.07
5	3	0.81	0.75
5	4	1.19	1.21
6	0	2.73	2.71
6	1	1.07	1.07
6	2	2.23	2.26
6	3	0.78	0.78
6	4	2.94	2.72
7	0	2.65	2.66
7	1	1.78	1.81
7	2	1.98	2.05
7	4	0.37	0.96
7	5	0.15	0.12
8	0	0.03	0.04
8	1	2.25	2.24
8	2	0.59	0.66
8	3	3.56	3.49
9	0	2.06	1.99
9	1	3.76	3.72
9	2	2.12	2.19
9	3	0.95	0.91

K	L	FO	FC
10	0	3.10	3.18
10	1	0.12	0.18
10	2	3.54	3.54
10	3	0.99	1.12

HOX NEUTRON H · 3

K	L	FO	FC
0	1	1.37	1.40
0	2	2.86	2.92
0	3	0.67	0.90
0	4	1.59	1.64
0	5	0.79	0.74
1	0	3.66	3.93
1	1	4.34	4.45
1	2	2.56	2.65
1	3	3.48	3.48
1	4	1.65	1.86
2	0	4.01	4.25
2	1	3.60	3.56
2	2	2.01	1.94
2	3	2.65	2.63
2	4	3.76	0.83
3	0	2.29	2.33
3	1	1.23	1.20
3	2	3.43	3.53
3	3	2.22	2.12
3	4	2.28	2.24
4	0	0.28	0.34
4	1	3.85	3.87
4	2	1.57	1.53
4	3	1.24	1.21
4	4	1.03	1.06
5	0	0.23	0.18
5	1	2.31	2.32
5	2	2.50	2.58
5	3	2.38	2.33
5	4	1.34	1.28
6	0	6.11	6.09
6	1	1.32	1.25
6	2	2.26	2.27
6	3	0.90	1.01
6	4	1.06	1.22
7	0	2.04	1.98
7	1	1.53	1.54
7	2	3.15	3.11
7	3	1.02	0.98
7	4	1.50	1.63
7	5	1.42	1.68
8	0	0.12	0.04
8	1	6.97	6.67
8	2	1.81	1.78
8	3	1.32	1.29
9	0	1.19	1.19
9	1	0.91	1.03
9	2	1.37	1.33
9	3	2.12	2.17
10	0	5.09	5.02
10	1	1.28	1.26
10	2	2.68	2.79
10	3	1.19	1.30

HOX NEUTRON H · 4

K	L	FO	FC
0	0	4.54	4.76
0	1	0.36	0.45
0	2	2.74	2.82
0	3	2.18	2.19
0	4	1.22	1.19
1	0	0.27	0.24
1	1	2.10	2.16
1	2	1.40	1.40
1	3	1.57	1.53
1	4	0.69	0.81
2	0	4.92	5.13
2	1	1.71	1.67
2	2	3.00	3.00
2	3	1.01	1.00
2	4	1.12	1.19
3	0	3.79	3.84
3	1	1.00	1.01
3	2	2.48	2.45
3	3	1.06	1.07
3	4	1.92	1.79
4	0	1.03	0.98
4	1	5.43	5.43
4	2	0.89	0.80
4	3	1.93	1.91
4	4	1.15	1.06
5	0	1.15	1.12
5	1	3.53	3.42
5	2	1.94	1.94
5	3	2.24	2.26
5	4	0.69	0.73
6	0	1.71	1.71
6	1	3.06	2.99
6	2	3.17	3.09
6	3	1.95	1.91
6	4	0.69	0.73
7	0	1.25	1.31
7	1	1.60	1.52
7	2	2.84	2.88
7	3	0.61	0.97
8	0	2.77	2.69
8	1	2.03	2.04
8	2	2.29	2.25
8	3	1.58	1.58
9	0	2.72	2.71
9	1	1.56	1.56
9	2	0.55	0.62
9	3	1.93	1.92
10	0	2.00	2.07
10	1	0.80	0.79
10	2	1.51	1.45

HOX NEUTRON H 5

K	L	FO	FC
0	1	4.92	5.04
0	2	1.89	1.84
0	3	0.12	0.10
0	4	4.03	3.80
1	0	3.16	3.26
1	1	3.97	4.04
1	2	3.37	3.41
1	3	1.97	1.98
1	4	1.34	1.26
2	0	2.53	2.59
2	2	3.51	3.49
2	3	2.37	2.39
2	4	0.72	0.68
3	0	3.15	3.06
3	1	2.14	2.14
3	2	1.44	1.47
3	3	1.05	1.06
3	4	0.0	0.60
4	0	2.60	2.51
4	1	2.57	2.54
4	2	2.18	2.15
4	3	0.65	0.63
4	4	2.49	2.58
5	0	1.15	1.08
5	1	4.50	4.33
5	2	1.42	1.39

K	L	FO	FC
5	3	0.63	0.62
5	4	0.97	0.99
6	0	0.12	0.15
6	1	2.39	2.42
6	2	0.48	0.53
6	3	2.42	2.46
7	0	8.27	7.69
7	1	2.06	1.97
7	2	2.79	2.77
7	3	0.70	0.40
8	0	0.28	0.47
8	1	0.83	0.87
8	2	1.62	1.68
8	3	0.10	0.25
9	0	0.74	0.78
9	1	3.55	3.57
9	2	1.57	1.67
10	0	0.65	0.71
10	1	0.52	0.71
10	2	0.86	0.87

HOX NEUTRON H · 6

K	L	FO	FC
0	0	2.36	2.29
0	1	0.37	0.28
0	2	1.73	1.69
0	3	1.34	1.34
0	4	0.21	0.40
1	0	2.22	2.22
1	1	4.61	4.65
1	2	1.95	1.92
1	3	2.47	2.49
1	4	0.79	0.97
2	0	2.49	2.47
2	1	1.79	1.76
2	2	1.61	1.57
2	3	0.92	0.93
2	4	0.8	0.32
3	0	5.02	5.76
3	1	0.97	0.98
3	2	3.12	3.06
3	3	0.43	0.46
3	4	1.85	1.98
4	0	1.88	1.77
4	1	2.63	2.60
4	2	1.48	1.57
4	3	1.07	1.10
4	4	0.38	0.86
5	0	1.56	1.60
5	1	2.30	2.22
5	2	2.01	1.98
5	3	3.12	3.04
6	0	0.81	0.81
6	1	1.73	1.68
6	2	2.81	2.85
6	3	0.76	0.64
7	0	2.07	2.19
7	1	1.38	1.42
7	2	2.35	2.36
7	3	0.71	0.81
8	0	0.24	0.17
8	1	1.48	1.38
8	2	0.53	0.62
9	0	0.73	0.82
9	1	2.39	2.49
9	2	0.44	0.55
10	0	0.45	0.23
10	1	1.24	1.28

HOX NEUTRON H · 7

K	L	FO	FC
1	3	1.87	1.93
1	4	1.52	1.46
2	0	3.58	3.44
2	1	1.89	1.82
2	2	2.90	2.98
2	3	1.01	0.97
3	0	1.69	1.64
3	1	4.25	4.05
3	2	2.71	2.69
3	3	0.03	0.63
4	0	0.55	0.58
4	1	2.16	2.00
4	2	1.14	1.08
4	3	1.03	1.00
5	1	2.22	2.26
5	2	0.33	0.19
5	3	1.33	1.73
6	0	2.24	2.20
6	1	0.90	0.94
6	2	0.99	0.99
7	0	1.24	1.24
7	1	0.12	0.07
7	2	1.93	1.96
8	0	1.51	1.54
8	1	4.57	4.31
8	2	3.80	0.88
9	0	0.50	0.48
9	1	0.93	0.95
10	0	2.94	3.20

HOX NEUTRON H · 8

K	L	FO	FC
0	0	3.19	3.07
0	1	1.33	1.33
0	2	2.30	2.17
0	3	0.33	0.62
1	0	1.66	1.75
1	1	2.55	2.48
1	2	1.54	1.59
1	3	0.57	0.72
2	0	2.57	2.49
2	1	0.90	0.91
2	2	1.79	1.78
2	3	0.41	0.55
3	0	1.72	1.74
3	1	1.96	1.98
3	2	1.23	1.26
3	3	0.41	0.56
4	0	0.17	0.11
4	1	2.14	2.09
4	2	1.04	1.13
4	3	2.56	2.53
5	0	2.10	2.23
5	1	0.49	0.59
5	2	0.82	0.88
5	3	1.12	1.13
6	0	1.47	1.53
6	1	1.95	1.97
6	2	1.38	1.42
7	0	1.55	1.56
7	1	0.92	0.86
8	0	3.08	3.05
8	1	1.82	1.87

HOX NEUTRON H · 9

K	L	FO	FC
0	1	1.39	1.36
1	1	2.57	2.58
2	0	1.77	1.82
2	1	1.34	1.49
2	2	0.34	0.33

Table 7 (*cont.*)

K	L	FO	FC
3	0	0.18	0.27
3	1	1.10	1.09
3	2	1.38	1.45
4	0	0.84	0.91
4	1	0.51	0.46
4	2	1.90	2.02
5	0	0.67	0.66
5	1	1.26	1.18
6	0	0.23	0.07
NOX NEUTRON H - 10			
0	0	5.71	5.20
0	1	0.99	1.00
1	0	0.31	0.22
1	1	2.45	2.32
2	0	1.31	1.27
2	1	2.96	2.80
3	0	0.42	0.46
3	1	0.78	0.89
4	0	1.93	2.03
NOX NEUTRON H - 0			
0	2	4.31	4.13
0	3	0.44	0.42
0	5	1.13	1.18
1	1	1.82	1.88
1	2	2.28	2.32
1	3	3.01	3.01
1	4	1.47	1.46
1	5	0.83	0.77
2	0	1.03	1.08
2	1	6.99	6.42
2	2	1.53	1.53
2	3	1.02	1.05
2	4	0.47	0.46
2	5	1.60	1.56
3	2	2.55	2.65
3	3	0.98	0.98
3	4	2.30	2.37
3	5	0.37	0.31
4	0	4.38	4.39
4	1	1.94	1.96
4	2	0.29	0.18
4	3	1.76	1.81
4	4	1.47	1.53
4	5	0.26	0.22
5	1	0.61	0.65
5	2	1.40	1.45
5	3	2.98	3.02
5	4	0.23	0.36
5	5	1.65	1.67
6	0	0.54	0.51
6	1	3.68	3.68
6	2	0.51	0.43
6	3	1.09	1.17
6	4	1.20	1.12
6	5	0.37	0.26
7	1	2.52	2.60
7	3	1.49	1.44
7	4	2.79	2.88
7	5	0.09	0.13
8	0	2.30	2.29
8	1	0.30	0.33
8	2	1.85	1.88
8	3	1.23	1.22
8	4	0.65	0.87
8	5	0.74	0.80
9	1	0.71	0.68
9	2	3.39	3.29
9	3	1.89	1.87
10	0	1.77	1.87
10	1	1.19	1.20
10	2	1.10	1.07
10	3	0.33	0.31
NOX NEUTRON H - 1			
0	1	1.17	1.16
0	2	4.68	4.65
0	3	0.28	0.19
0	4	2.17	2.11
0	5	0.31	0.33
1	0	2.24	2.18
1	1	3.27	3.27
1	2	3.33	3.08
1	3	2.10	2.07
1	4	1.06	0.99
1	5	1.30	1.28
2	0	1.15	1.23
2	1	3.45	3.48
2	2	2.00	2.02
2	3	3.43	3.49
2	4	1.27	1.28
2	5	1.49	1.38
3	0	0.84	0.84
3	1	2.30	2.39
3	2	1.19	1.19
3	3	1.58	1.61
3	4	1.97	2.00
3	5	0.93	0.84
4	0	2.91	3.14
4	1	0.63	0.59
4	2	3.40	3.47
4	3	1.11	1.13
4	4	1.77	1.79
4	5	0.57	0.58
5	0	1.67	1.71
5	1	1.76	1.83
5	2	1.98	2.02
5	3	1.66	1.69
5	4	0.09	0.09
5	5	1.71	1.72
6	0	0.13	0.11
6	1	3.04	3.02
6	2	0.73	0.69
6	3	1.74	1.80
6	4	0.61	0.51
6	5	1.08	0.99
7	0	2.69	2.73
7	1	0.68	0.67
7	2	3.27	3.30
7	3	1.56	1.57
7	4	1.60	1.63
7	5	0.59	0.58
8	0	0.35	0.02
8	1	3.01	2.97
8	2	1.26	1.25
8	3	1.22	1.21
8	4	0.64	0.67
8	5	0.62	0.66
9	0	2.71	2.69
9	1	1.46	1.44
9	2	0.74	0.73
10	0	3.72	3.77
10	1	0.63	0.69
10	2	1.84	1.87
10	3	0.13	0.12
NOX NEUTRON H - 2			
0	0	1.16	1.16
0	1	0.89	0.86
0	2	0.73	0.73
0	3	0.49	0.47
0	4	0.57	0.51
0	5	0.24	0.34
1	0	2.21	2.23
1	1	5.35	5.22
1	2	1.31	1.29
1	3	1.83	1.82
1	4	2.00	2.04
1	5	1.20	1.19
2	1	1.18	1.19
2	2	0.41	0.51
2	3	0.44	0.51
2	4	1.13	1.08
3	0	4.14	4.13
3	1	1.94	1.94
3	3	1.21	1.15
3	4	1.23	1.29
3	5	1.18	1.15
4	0	1.79	1.90
4	1	0.82	0.81
4	2	1.51	1.51
4	3	1.92	1.87
4	4	0.20	0.27
4	5	1.12	1.16
5	0	3.85	3.89
5	1	0.82	0.85
5	2	1.60	1.65
5	3	1.33	1.29
5	4	0.69	0.69
5	5	1.64	1.59
6	0	2.22	2.29
6	1	2.02	2.06
6	2	2.20	2.27
6	3	1.12	1.10
6	4	2.61	2.55
6	5	0.37	0.08
7	0	2.26	2.50
7	1	0.38	0.30
7	2	2.04	2.07
7	3	0.72	0.67
7	4	0.98	0.96
7	5	0.09	0.12
8	0	2.61	2.72
8	1	2.23	2.20
8	2	0.80	0.77
8	3	3.67	3.49
8	4	0.49	0.49
9	0	3.05	2.95
9	1	3.94	3.80
9	2	2.10	2.04
10	0	1.29	1.38
10	1	0.69	0.71
10	2	3.03	3.01
10	3	1.01	1.10
NOX NEUTRON H - 3			
0	1	0.43	0.29
0	2	2.08	2.13
0	3	0.41	0.40
0	4	1.50	1.46
0	5	0.67	0.66
1	0	3.99	3.70
1	1	3.27	3.18
1	2	1.20	1.23
1	3	2.69	2.66
1	4	1.35	1.34
1	5	1.02	0.96
2	0	4.43	4.16
2	1	2.08	2.06
2	2	2.28	2.25
2	3	2.18	2.12
2	4	0.41	0.45
2	5	0.69	0.67
3	0	1.71	1.72
3	1	0.49	0.48
3	2	3.55	3.63
3	3	1.86	1.88
3	4	2.85	2.80
3	5	0.62	0.69
4	0	3.45	3.36
4	1	2.01	2.00
4	2	1.26	1.25
4	3	0.86	0.83
4	4	1.55	1.59
4	5	1.16	1.08
5	0	0.64	0.66
5	1	2.49	2.53
5	2	1.21	1.21
5	3	2.80	2.79
5	4	0.61	0.49
5	5	0.96	0.83
6	0	3.10	3.13
6	1	1.13	1.19
6	2	1.89	1.94
6	3	1.56	1.57
6	4	1.39	1.40
6	5	0.72	0.76
7	0	2.14	2.22
7	1	0.71	0.72
7	2	2.96	2.93
7	3	0.96	0.92
7	4	1.80	1.78
7	5	0.84	0.90
8	0	1.03	1.14
8	1	4.89	4.64
8	2	1.07	1.06
8	3	1.24	1.22
8	4	1.11	1.08
9	0	1.65	1.64
9	1	1.25	1.23
9	2	1.15	1.12
9	3	1.55	1.53
10	0	3.69	3.74
10	1	1.14	1.11
10	2	2.62	2.69
10	3	0.56	0.66
NOX NEUTRON H - 4			
0	0	5.29	5.06
0	1	0.11	0.17
0	2	5.13	5.10
0	3	2.17	2.22
0	4	0.0	0.28
0	5	0.82	0.80
1	0	0.37	0.32
1	1	2.11	2.19
1	2	0.94	0.87
1	3	1.15	1.07
1	4	0.21	0.29
1	5	0.80	0.79
2	0	3.29	3.24
2	1	3.10	3.19
2	2	1.60	1.64
2	3	1.56	1.56
2	4	0.68	0.64
2	5	1.69	1.70
3	0	3.30	3.32
3	1	1.33	1.34
3	2	2.31	2.29
3	3	0.78	0.80
3	4	1.42	1.38
3	5	0.36	0.11
4	0	0.89	0.94
4	1	3.49	3.48
4	2	2.12	2.15
4	3	1.62	1.57
4	4	1.33	1.28
4	5	0.39	0.29
5	0	1.99	1.97
5	1	2.99	3.00
5	2	0.0	2.21
5	3	2.41	2.38
5	4	1.06	1.06
5	5	1.11	1.10
6	0	0.26	0.17
6	1	3.34	3.46
6	2	2.50	2.46
6	3	1.48	1.46
6	4	0.98	0.96
6	5	0.76	0.83
7	0	2.58	2.60
7	1	1.49	1.49
7	2	2.98	2.97
7	3	1.05	1.07
7	4	1.45	1.31
7	5	0.67	0.61
8	0	2.32	2.44
8	1	1.50	1.41
8	2	2.20	2.16
8	3	1.92	1.83
9	0	1.13	1.18
9	1	2.26	2.35
9	2	1.06	1.08
9	3	1.64	1.66
10	0	1.30	1.34
10	1	0.74	0.71
10	2	1.06	1.01
NOX NEUTRON H - 5			
0	1	2.36	2.36
0	2	1.45	1.42
0	3	0.62	0.60
0	4	3.70	3.81
0	5	0.74	0.72
1	0	1.94	1.90
1	1	2.56	2.58
1	2	1.74	1.80
1	3	1.66	1.66
1	4	1.24	1.19
1	5	0.37	0.30
2	0	1.02	1.04
2	1	0.76	0.72
2	2	2.17	2.22
2	3	3.49	3.44
2	4	0.72	0.65
2	5	1.94	2.00
3	0	1.77	1.74
3	1	0.96	0.96
3	2	0.60	0.60
3	3	1.31	1.32
3	4	0.92	0.86
3	5	0.48	0.52
4	0	0.97	0.95
4	1	1.16	1.13
4	2	2.17	2.16
4	3	0.25	0.20
4	4	2.76	2.80
4	5	0.16	0.24
5	0	0.85	0.82
5	1	3.12	3.14
5	2	1.12	1.08
5	3	0.65	0.64
5	4	0.66	0.67
5	5	0.66	0.88
6	0	0.37	0.42
6	1	2.04	2.08
6	2	0.52	0.54
6	3	2.92	2.84
6	4	0.58	0.54
6	5	0.94	0.94
7	0	6.65	6.35
7	1	1.71	1.66
7	2	2.19	2.14
7	3	0.88	0.83
7	4	0.43	0.42
8	0	0.71	0.74
8	1	0.92	0.90
8	2	1.78	1.72
8	3	0.67	0.66
9	0	0.18	0.23
9	1	2.79	2.82
9	2	1.58	1.63
10	2	0.63	0.66
NOX NEUTRON H - 6			
0	0	2.49	2.45
0	1	0.68	0.70
0	2	2.86	2.98
0	3	1.29	1.34
0	4	0.09	0.07
0	5	0.27	0.35
1	0	0.29	0.21
1	1	4.23	4.16
1	2	1.65	1.66
1	3	1.62	1.60
1	4	1.14	1.20
1	5	0.92	0.97
2	0	1.25	0.37
2	1	2.12	2.14
2	2	1.88	1.85
2	3	1.16	1.20
2	4	0.28	0.35
2	5	1.17	1.21
3	0	5.01	4.80
3	1	1.03	1.03
3	2	2.65	2.59
3	3	0.81	0.81
3	4	1.07	1.08
3	5	0.73	0.77
4	0	1.17	1.19
4	1	1.81	1.87
4	2	1.67	1.67
4	3	1.70	1.70
4	4	1.01	0.97
4	5	0.58	0.55
5	0	1.26	1.30
5	1	2.19	2.21
5	2	2.44	2.43
5	3	1.98	2.05
5	4	0.86	0.84
6	0	0.70	0.67
6	1	1.78	1.75
6	2	2.74	2.67
6	3	1.09	1.01
6	4	1.00	0.95
7	0	2.25	2.26
7	1	0.89	0.87
7	2	2.15	2.11
8	0	0.68	0.70
8	1	1.38	1.36
8	2	1.13	1.16
9	0	1.10	1.08
9	1	2.19	2.26
9	2	1.00	1.02
10	0	0.51	0.42
10	1	1.07	1.07
NOX NEUTRON H - 7			
0	1	1.25	1.22
0	2	1.21	1.21
0	3	2.19	2.22
0	4	0.70	0.67
0	5	0.40	0.59
1	0	2.91	2.91
1	1	2.24	2.22
1	2	1.23	1.29
1	3	2.60	2.58
1	4	1.51	1.47
1	5	0.51	0.82
2	0	3.32	3.22
2	1	1.62	1.54
2	2	2.26	2.29
2	3	1.06	1.01
2	4	1.28	1.28
3	0	3.30	3.17
3	1	2.25	2.21
3	2	2.64	2.59
3	3	0.99	0.92
3	4	1.49	1.43
4	0	0.33	0.29
4	1	1.91	1.76
4	2	0.89	0.85
4	3	1.22	1.21
4	4	0.56	0.62
5	0	1.60	1.65
5	1	1.83	1.80
5	2	0.43	0.45
5	3	2.00	1.97
5	4	0.59	0.52
6	0	2.44	2.32
6	1	1.61	1.60
6	2	0.45	0.45
6	3	0.98	0.98
7	0	0.80	0.77
7	2	1.95	1.95
8	0	0.22	0.33
8	1	3.49	3.49
8	2	1.23	1.29
9	0	0.45	0.02
9	1	0.92	0.93
10	0	2.93	2.90
NOX NEUTRON H - 8			
0	0	2.68	2.66
0	1	0.77	0.76
0	2	1.79	1.77
0	3	0.81	0.86
0	4	0.72	0.69
1	0	0.83	0.81
1	1	2.16	2.09
1	2	1.63	1.69
1	3	0.63	0.64
1	4	0.57	0.55
2	0	2.33	2.29
2	1	0.99	1.04
2	2	1.39	1.44
2	3	0.15	0.14
2	4	0.98	0.93
3	1	1.53	1.49
3	2	1.30	1.31
3	3	0.88	0.81
3	4	0.83	0.86
4	0	0.86	0.86
4	1	2.44	2.36
4	2	3.36	2.87
4	3	1.49	1.51
5	0	0.84	0.86
5	1	0.67	0.64
5	2	0.94	0.88
6	0	0.63	0.69
6	1	2.22	2.30
6	2	0.94	0.97
7	0	1.38	1.38
7	1	0.98	0.93
8	0	2.94	2.93
8	1	1.38	1.36
NOX NEUTRON H - 9			
0	1	0.44	0.47
0	2	2.96	2.98
0	3	0.70	0.70
1	0	1.25	1.18
1	1	1.81	1.88
1	2	1.33	1.38
2	0	0.78	0.82
2	1	2.24	2.20
2	2	0.22	0.10
3	0	0.09	0.09
3	1	1.21	1.23
3	2	0.54	0.54
4	0	0.45	0.44
4	1	0.39	0.22
4	2	1.99	2.07
5	0	1.68	1.64
5	1	1.25	1.22
6	0	0.71	0.67
6	1	1.28	1.34
NOX NEUTRON H - 10			
0	0	5.24	5.02
0	1	1.86	1.87
1	0	0.73	0.66
1	1	1.95	2.01
2	0	1.73	1.73
2	1	2.72	2.74
3	0	1.28	1.27
3	1	0.82	0.88
4	0	2.26	2.25
DOX NEUTRON H - 0			
0	1	0.76	0.74
0	2	3.96	4.12
0	3	1.57	1.58
0	4	0.84	0.79
0	5	1.65	1.54
1	1	1.73	1.73
1	2	1.46	1.55
1	3	1.51	1.52
1	4	1.32	1.31
2	0	3.30	3.24
2	1	10.65	10.95
2	2	2.61	2.55
2	3	1.14	1.13
2	4	0.83	0.78
3	1	1.87	1.80
3	2	2.34	2.39
3	3	0.84	0.82
3	4	0.94	0.95
4	0	7.33	7.75
4	1	0.72	0.60
4	2	1.51	1.44
4	3	0.43	0.35
4	4	0.53	0.60
5	1	2.29	2.28
5	2	4.04	4.04
5	3	3.22	3.20
5	4	0.53	0.57
6	0	3.18	3.16
6	1	3.45	3.45
6	2	3.16	3.08
6	3	1.44	1.39
6	4	1.14	1.06
7	1	4.55	4.52
7	2	2.94	2.89
7	3	4.04	3.99
7	4	3.80	3.59
8	0	2.54	2.52
8	1	1.64	1.66
8	2	2.99	3.03
8	3	1.05	1.02
8	4	1.66	1.67
9	1	0.96	0.97
9	2	4.80	4.75
9	3	3.17	3.06
10	0	0.90	0.91
10	1	0.80	0.77
10	2	1.82	1.82
10	3	0.78	0.78
DOX NEUTRON H - 1			
0	1	0.49	0.42
0	2	3.50	3.58
0	3	4.52	4.47
0	4	1.09	1.11
1	0	0.70	0.52
1	1	2.79	2.84
1	2	0.65	0.66
1	3	1.81	1.81
1	4	0.73	0.73
2	0	2.13	2.02
2	1	5.48	5.45
2	2	2.35	2.39
2	3	1.79	1.82
2	4	1.98	1.98
3	0	0.39	0.32
3	1	4.55	4.55
3	2	1.01	0.99
3	3	1.81	1.80
3	4	1.89	1.83
4	0	4.43	4.38
4	1	1.50	1.58
4	2	4.88	4.86
4	3	0.95	0.95
4	4	2.91	2.82
5	0	1.33	1.31
5	1	0.66	0.63
5	2	2.20	2.24
5	3	2.05	2.10
5	4	0.59	0.56
6	0	1.45	1.47
6	1	4.28	4.27
6	2	1.69	1.70
6	3	2.43	2.52
6	4	0.91	0.92
7	0	1.73	1.71
7	1	1.24	1.25
7	2	2.58	2.63
7	3	1.42	1.44
7	4	2.61	2.60
8	0	0.24	0.26
8	1	1.27	1.28
8	2	1.96	2.00
8	3	0.84	0.93
8	4	0.73	0.71
9	0	1.22	1.28
9	1	0.28	0.33
9	2	1.59	1.56
9	3	3.15	3.15
10	0	2.83	2.76
10	1	0.68	0.72
10	2	1.95	2.03
10	3	0.72	0.76
DOX NEUTRON H - 2			
0	0	2.92	2.91
0	1	0.35	0.84
0	2	1.98	1.89
0	3	0.20	0.12
0	4	1.20	1.25
1	0	6.44	6.92
1	1	0.28	0.57
1	2	0.72	0.73
1	3	1.41	1.43
1	4	2.14	2.13
2	0	0.66	0.63
2	1	2.88	2.88
2	2	1.01	1.00
2	3	0.96	0.93
2	4	0.47	0.44
3	0	6.63	6.71
3	1	2.88	2.87
3	2	3.47	3.57
3	3	2.04	2.04
3	4	0.83	0.82
4	0	2.53	2.48
4	1	2.85	2.84
4	2	3.23	3.25
4	3	1.78	1.84
4	4	0.76	0.77
5	0	1.92	1.92
5	1	3.33	3.34
5	2	0.91	0.98
5	3	2.34	2.31
5	4	0.32	0.13
6	0	1.70	1.75
6	1	3.48	3.43
6	2	4.47	4.47
6	3	2.89	2.86
6	4	3.27	3.17
7	0	2.49	2.46
7	1	2.88	2.81
7	2	2.49	2.56
7	3	1.39	1.43
8	0	6.19	6.03
8	1	2.53	2.53
8	2	2.93	2.93
8	3	4.62	4.47
9	0	5.31	5.03
9	1	5.06	5.02
9	2	2.38	2.41
9	3	3.30	3.20
10	0	1.14	1.17
10	1	2.15	2.24
10	2	2.84	2.77
10	3	1.69	1.67
DOX NEUTRON H - 3			
0	1	1.38	1.33
0	2	1.05	1.09
0	3	0.48	0.47
0	4	1.49	1.53
1	0	3.58	3.50
1	1	1.54	1.57
1	2	0.91	0.85
1	3	2.07	2.07
1	4	1.05	1.09
2	0	4.00	3.98
2	1	0.76	0.69
2	2	4.01	3.98
2	3	2.08	2.06
2	4	0.20	0.27
3	0	0.99	0.96
3	1	0.78	0.74
3	2	5.11	5.10
3	3	1.66	1.68
3	4	2.71	2.65
4	0	0.95	0.94
4	1	0.0	2.18
4	2	3.50	3.54
4	3	0.61	0.59
4	4	1.58	1.59
5	0	1.32	1.33
5	1	3.87	3.97
5	2	0.61	0.60
5	3	3.87	3.88
5	4	1.75	1.81
6	0	1.46	1.42
6	1	2.45	2.52
6	2	1.45	1.47
6	3	2.95	3.08
6	4	2.32	2.32
7	0	2.81	2.81
7	1	0.44	0.39
7	2	3.84	3.94
7	3	1.53	1.57
7	4	3.10	3.08
8	0	2.78	2.77
8	1	1.87	1.96
8	2	0.59	0.56
8	3	2.49	2.52
9	0	2.44	2.36
9	1	2.32	2.35
9	2	1.48	1.46
9	3	1.09	1.10
10	0	2.47	2.46
10	1	1.87	1.98
10	2	3.19	3.25
10	3	1.94	2.04
DOX NEUTRON H - 4			
0	0	5.59	5.62
0	1	0.48	0.32
0	2	0.93	0.94
0	3	2.56	2.58
0	4	1.27	1.28
1	0	0.46	0.45
1	1	2.46	2.52
1	2	0.97	0.97
1	3	1.20	1.24
1	4	0.54	0.54
2	0	0.45	0.53
2	1	5.95	6.06
2	2	1.61	1.59
2	3	2.59	2.65
2	4	2.56	2.54
3	0	2.76	2.81
3	1	2.79	2.82
3	2	2.23	2.20
3	3	1.70	1.76
3	4	1.17	1.16
4	0	0.89	0.87
4	1	2.15	2.21
4	2	4.85	4.84
4	3	1.41	1.40
4	4	2.05	2.02
5	0	3.54	3.49
5	1	2.73	2.77
5	2	2.47	2.52
5	3	3.09	3.05
5	4	1.98	1.98
6	0	1.94	1.91
6	1	4.97	4.91
6	2	1.94	2.00
6	3	2.11	2.10
6	4	1.50	1.56
7	0	5.15	5.09
7	1	2.06	2.09
7	2	3.63	3.65
7	3	2.16	2.13
8	0	2.36	2.36
8	1	1.92	1.97
8	2	2.56	2.58
8	3	2.79	2.77
9	0	0.90	0.97
9	1	4.76	4.69
9	2	2.88	2.84
9	3	1.70	1.66
10	0	0.63	0.62
10	1	1.02	1.05
10	2	0.58	0.46
DOX NEUTRON H - 5			
0	1	1.42	1.35
0	2	1.05	1.06
0	3	1.16	1.19
0	4	5.24	4.99
1	0	0.28	0.13
1	1	2.42	2.42
1	2	0.55	0.54
1	3	1.83	1.87
1	4	1.39	1.41
2	0	1.34	1.26
2	1	3.61	3.58
2	2	0.75	0.76
2	3	6.03	5.78
2	4	0.95	0.99
3	0	0.50	0.37
3	1	3.55	3.57
3	2	0.96	0.92
3	3	2.37	2.39
3	4	1.62	1.62
4	0	1.37	1.39
4	1	2.01	1.95
4	2	2.81	2.82
4	3	0.79	0.88
4	4	4.52	4.18
5	0	0.48	0.43
5	1	2.12	2.13
5	2	2.62	2.63
5	3	0.89	0.98
6	0	0.80	0.74
6	1	1.88	1.91
6	2	0.55	0.47
6	3	4.39	4.16
7	0	4.72	4.68
7	1	1.33	1.37
7	2	1.55	1.52
7	3	1.54	1.53
8	0	1.09	1.07
8	1	1.22	1.24
8	2	2.07	2.06
8	3	1.66	1.72
9	0	0.61	0.60
9	1	2.50	2.46
9	2	2.60	2.62
10	0	1.33	1.34
10	1	0.83	0.77
10	2	0.73	0.69
DOX NEUTRON H - 6			
0	0	3.45	3.44
0	1	2.18	2.21
0	2	5.22	5.16
0	3	1.49	1.55
0	4	0.15	0.47
1	0	2.71	2.69
1	1	3.90	3.89
1	2	2.94	2.92
1	3	0.93	0.88
1	4	2.86	2.79
2	0	2.66	2.59
2	1	3.12	3.11
2	2	2.71	2.74
2	3	2.67	2.70
2	4	1.08	1.12
3	0	4.07	3.99
3	1	1.33	1.33
3	2	4.16	4.07
3	3	1.64	1.67
4	0	0.57	0.57
4	1	3.10	3.08
4	2	2.53	2.52
4	3	3.09	2.99
4	4	1.72	1.69
5	0	0.93	0.94
5	1	3.00	2.97
5	2	3.42	3.37
5	3	1.14	1.13
6	0	2.86	2.81
6	1	2.17	2.22
6	2	3.14	3.08
6	3	1.91	1.97
7	0	2.98	2.92
7	1	0.28	0.23
7	2	2.05	2.03
7	3	0.40	0.28
8	0	1.79	1.85
8	1	1.66	1.63
8	2	2.68	2.69
9	0	2.03	2.01
9	1	2.53	2.48
10	0	1.20	1.11
10	1	2.35	2.32
DOX NEUTRON H - 7			
0	1	0.28	0.29
1	3	4.31	4.19
2	0	3.25	3.15
2	1	1.91	1.94
2	2	1.63	1.71
2	3	1.37	1.42
3	0	6.22	5.86
3	1	1.14	1.15
3	2	3.06	3.05
3	3	1.62	1.62
4	0	1.46	1.47
4	1	1.57	1.53
4	2	1.21	1.24
4	3	1.10	1.08
5	0	1.32	1.38
5	1	1.69	1.67
5	2	1.13	1.14
5	3	3.18	3.13
6	0	2.89	2.83
6	1	3.04	3.02
6	2	0.48	0.49
7	0	0.48	0.32
7	2	3.23	3.24
7	1	1.30	1.26
7	3	0.74	0.83
8	0	1.73	1.75
8	1	3.49	3.36
8	2	2.24	2.27
9	0	0.69	0.76
9	1	1.13	1.17
DOX NEUTRON H - 8			
0	0	2.15	2.16
0	1	0.18	0.13
0	2	1.33	1.30
0	3	1.55	1.61
1	0	0.25	0.18
1	1	1.91	1.96
1	2	2.30	2.34
1	3	0.80	0.79
2	0	2.38	2.37
2	1	1.56	1.59
2	2	1.16	1.13
2	3	1.13	1.23
3	0	0.52	0.48
3	1	1.67	1.71
3	2	1.82	1.88
3	3	1.85	1.91
4	0	2.39	2.41
4	1	3.46	3.38
4	2	0.61	0.63
5	0	0.97	1.00
5	1	1.08	1.12
5	2	1.29	1.27
6	0	0.50	0.43
6	1	3.68	3.60
6	2	0.49	0.42
7	0	1.45	1.50
7	1	1.43	1.47
8	0	3.60	3.45
8	1	2.30	2.31
DOX NEUTRON H - 9			
0	1	0.82	0.84
0	2	4.94	4.77
1	0	0.36	0.31
1	1	1.00	1.03
1	2	1.19	1.23
2	0	0.43	0.43
2	1	4.15	4.04
2	2	0.68	0.67
3	0	0.23	0.13
3	1	1.77	1.81
3	2	1.08	1.17
4	0	2.76	2.80
4	1	0.41	0.37
4	2	2.69	2.68
5	0	[illegible]	[illegible]
5	1	1.68	1.68
6	0	1.36	1.36
6	1	1.07	1.08
DOX NEUTRON H - 10			
0	0	6.54	6.26
0	1	3.95	3.91
1	1	1.85	1.88
2	0	3.24	3.22
2	1	3.71	3.64
3	0	2.88	2.76
3	1	1.41	1.45

Table 8. *Refinements of DOX(N) data with and without an isotropic extinction correction*, $g'=2·0\ (1)$

Δ_1 = Absolute value of difference between parameters before and after extinction corrections.
Δ_2 = Difference between X-ray and neutron temperature factors, with extinction corrections in each case,

(*a*) R values and χ

	Before extinction correction	After extinction correction
R_W	0·104	0·032
R_U	0·108	0·027
χ	7·2	2·5

(*b*) Coordinates

	$10^4\Delta_1$ for x	$10^4\Delta_1$ for y	$10^4\Delta_1$ for z
C	1 (5)	5 (5)	18 (13)
O(1)	13 (7)	16 (7)	16 (19)
O(2)	13 (7)	5 (5)	48 (19)
N	7 (5)	3 (4)	9 (10)
O(*W*)	—	-	52 (28)
H(*W*)	25 (7)	3 (6)	33 (19)
H(1)	14 (8)	1 (6)	5 (18)
H(2)	12 (9)	9 (6)	19 (19)
H(3)	11 (7)	8 (7)	5 (17)
H(4)	11 (8)	8 (7)	77 (17)

(*c*) Temperature factors β_{ii}

	(i) For β_{11}		(ii) For β_{22}		(iii) For β_{33}	
	$10^4\Delta_1$	$10^4\Delta_2$	$10^4\Delta_1$	$10^4\Delta_2$	$10^4\Delta_1$	$10^4\Delta_2$
C	25 (6)	17 (3)	11 (5)	8 (4)	143 (28)	−27 (12)
O(1)	−14 (8)	11 (2)	19 (6)	−2 (2)	336 (40)	0 (13)
O(2)	16 (7)	9 (2)	32 (5)	−8 (2)	101 (46)	50 (14)
N	19 (5)	4 (1)	9 (4)	−2 (2)	140 (25)	49 (12)
O(*W*)	5 (13)	6 (4)	11 (8)	12 (4)	89 (65)	44 (16)
H(*W*)	−2 (9)		22 (7)		−19 (53)	
H(1)	14 (9)		14 (7)		14 (40)	
H(2)	−9 (11)		13 (6)		66 (44)	
H(3)	25 (8)		13 (6)		31 (42)	
H(4)	30 (9)		1 (7)		143 (36)	

Table 9. *Measured scattering lengths* b(H, D) *at the* H, D *sites in NOX(N)*

Site	b(H, D) in 10^{-12} cm
H(*W*)	0·021 (3)
H(1)	0·038 (3)
H(2)	0·042 (3)
H(3)	0·034 (3)
H(4)	0·034 (3)

tetramine of 0·018 Å towards the lone pairs (the difference between X-ray and neutron-diffraction locations). The displacement was four times the standard deviation. Hydrogen-atom electron-density centroids were displaced by about 0·1 Å towards the bonded atom (Coulson, 1970).

In this analysis, the averaged parameters from the DOX1(XR) and DOX2(XR) refinements were compared with the DOX(N) results (see Table 4). Within the experimental errors the C, O(1), O(2) and N atoms were not significantly shifted in the X-ray refinements by asphericity effects. The lone-pair effects in the oxygen atoms O(1) and O(2) were probably balanced by σ and π bonding effects; no shifts were expected for the more symmetrically bound carbon and nitrogen atoms. However, there was a displacement of 0·013 ± 0·003 Å of the water oxygen towards its lone-pair orbitals. This shift was significant and was of the same magnitude as observed for the N in hexamethylenetetramine (Duckworth *et al.* 1970).

The bond lengths O–D and N–D measured for the DOX1(XR), DOX2(XR) and DOX refinements are

Table 10. *Bond lengths* O–D *and* N–D *measured in DOX*1(*XR*), *DOX*2(*XR*)*and DOX*(*N*) *refinements, uncorrected for thermal motion*

	[DOX1(XR)+DOX2(XR)]/2 length (Å)	DOX(N) length (Å)	Δ(X–N) Å
O(*W*)–D	0·825 (40)	0·969 (3)	−0·144
N–D(1)	0·930 (50)	1·031 (2)	−0·101
N–D(2)	0·949 (60)	1·032 (2)	−0·083
N–D(3)	1·006 (50)	1·035 (2)	−0·029
N–D(4)	0·958 (60)	1·032 (2)	−0·074

$\overline{\Delta(\text{X–N})} = -0·086 \pm 0·037$ Å.

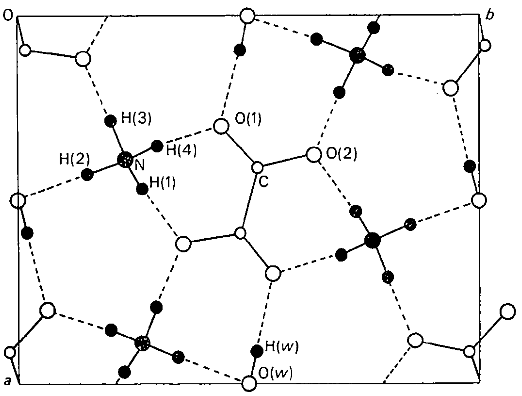
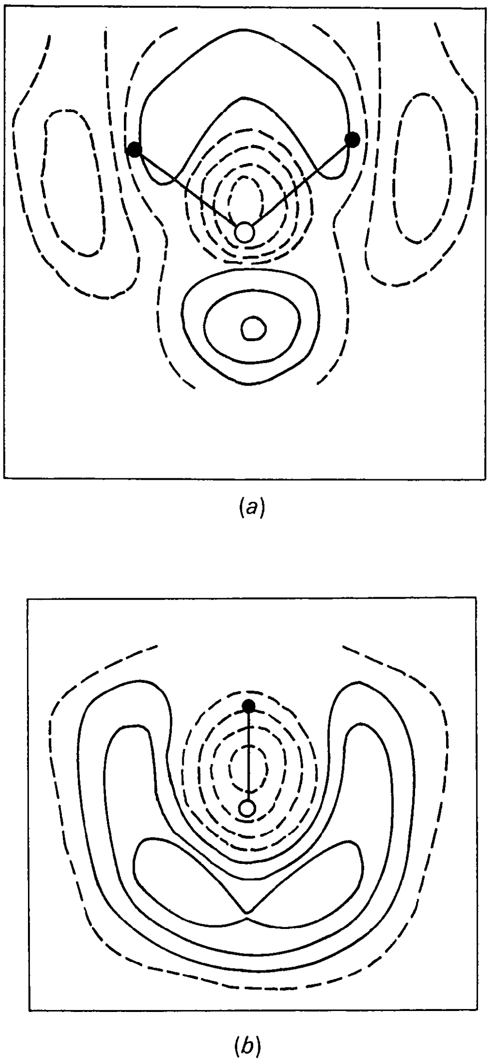


Fig. 1. Crystal structure of ammonium oxalate monohydrate.


(*a*)

(*b*)

Fig. 2. (*a*) X–N map in the plane of the water molecule in DOX. Contours 0·05 e.Å^{-3}, zero and negative contours dashed. (*b*) X–N map in plane perpendicular to water molecule plane in DOX and bisecting the D–O–D angle. Contour levels as in (*a*).

given in Table 10 (uncorrected for thermal motion). The mean asphericity shift for D was −0·086 ± 0·037 Å (standard deviation), about the value suggested by Coulson (1970).

(*d*) *Hydrogen bonding*

The hydrogen bonding in ammonium oxalate monohydrate forces the oxalate ion out of its planar configuration and the carboxyl planes are twisted about 28° relative to each other. The structure and hydrogen bond scheme are shown in Fig. 1. Water···O(1) hydrogen bonds link the oxalate ions in the direction of **a**. The ammonium ion H or D atoms form N···O hydrogen bonds. The water molecules and ammonium ions are both in tetrahedral hydrogen bonding environments. The ammonium ion is nearly regular with H–N–H angles between 107·5 (3) and 112·0 (3)° (Table 6); this is in contrast to the results of the two-dimensional neutron study of Padmanabhan *et al.* (1965) on this compound which gave angles between 104 and 117°. The present results with the three-dimensional neutron data are more accurate.

The hydrogen-bond distances and angles are given in Table 6. The hydrogen bonds are non-linear with O–H···O and N–H···O angles lying between 166·9 (4) and 177·8 (4)°. The water molecule angles 109·7 (4)° in DOX(N) and 108·9 (7)° in HOX(N) are identical in both isotopic forms, in agreement with the work of Coppens & Sabine (1969) on the protonated and deuterated forms of α-oxalic acid dihydrate. We have averaged the bond lengths and angles over the isotopic forms in Table 6, as very little difference occurred between the three forms.

In Table 11, the hydrogen bond lengths in HOX and DOX are compared. The bond-length changes on deuteration are not significant when compared with their standard deviations. In α-oxalic acid dihydrate (Delaplane & Ibers, 1969) all three hydrogen bonds expand significantly on deuteration by about 0·02 Å.

Table 11. *Changes in hydrogen bond lengths r_{DOX}–r_{HOX} in ammonium oxalate monohydrate on deuteration*

Bond	r_{DOX}–r_{HOX}
O(*W*)–H(*W*)···O(1)	−0·010 (6) Å
N——H(2)····O(2)	+0·003 (6)
N——H(3)····O(*W*)	+0·009 (5)
N——H(4)····O(2)	+0·005 (5)
N——H(5)····O(1)	+0·003 (6)

(*e*) *Thermal parameters*

The effects of the lack of an extinction correction on thermal parameters derived from least-squares refinements were discussed above in § (*a*). When extinction corrections are applied, X-ray thermal parameters are were larger than neutron values because of bonding effects. This is shown in Table 8, where the Δ_2 values are mainly positive.

In Table 12 are listed the r.m.s. displacements along the principal axes *R*1, *R*2 and *R*3 of the vibration ellipsoids calculated with the program *ORFFE* (Busing *et al.* 1962*b*) for the DOX(N) data. The vibrations of the oxalate atoms were very anisotropic. The angles which the principal axis *R*3 makes with **c** were small for the oxalate ion and the water molecules, in agreement with the anisotropic thermal expansion of the crystal, which, in going from 130°K to room temperature, expands most in the direction of **c** (~1·6%) (Robertson, 1965). The ammonium ion vibrations were largely isotropic.

(*f*) *Bonding electron effects in X-ray data*

X–N maps were calculated for the (HOX1+HOX2)/2, (NOX1+NOX2)/2 and (DOX1+DOX2)/2 X-ray data sets with corresponding neutron parameters. The X-ray scale factor was refined after including the neutron parameters. The (NOX1+NOX2)/2 map showed no bonding features. This was probably due to the fact that extinction in the NOX1(XR) crystal was severe. The protonated and deuterated crystals, however, showed features which could be attributed to bonding effects. The results for the deuterated crystals, which gave the best maps, are shown in Figs. 2–5. In all these maps, the contour interval is 0·05 e.Å^{-3} with zero and negative contours dashed.

(i) *Water molecule*

There was a positive peak at the back of the water oxygen atom, O(*W*), of 0·15 e.Å^{-3} [Fig. 2(*a*)]. This was similar to the peak found at the back of the water molecule in α-oxalic acid dihydrate by CSDI (1969). A higher negative peak at the oxygen position occurred in the latter case as the X-ray scale factor was not refined after inclusion of neutron parameters. The positive peak was also extended in a plane perpendicular to the water plane bisecting the water angle, Fig. 2(*b*), suggesting some tetrahedral disposition of the lone-pairs, and some *s*-character in the oxygen lone-pair orbitals. The peak behind the water molecule was consistent with the O(*W*) asphericity shift of 0·013±0·003 Å in the direction of the lone-pairs [§ (*c*)] and the tetrahedral extension was compatible with the O(*W*) tetrahedral environment.

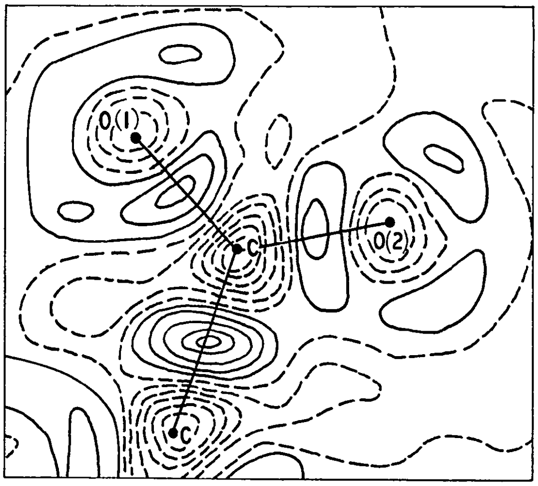


Fig. 3. X–N map in carboxyl plane in DOX. Contours levels as in Fig. 2(*a*).

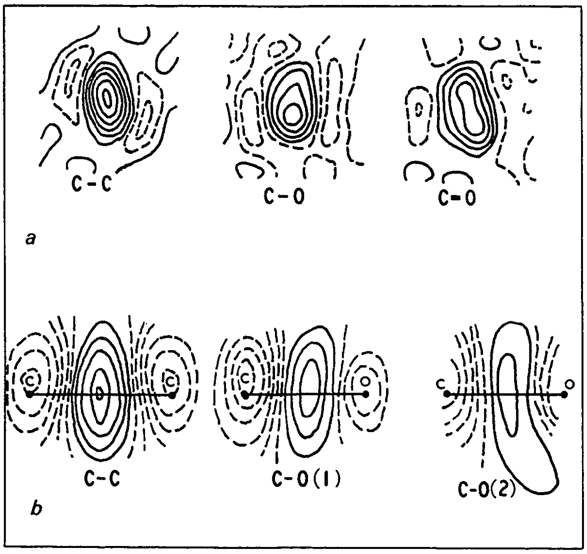


Fig. 4. (*a*) Extension of mid-bond peaks in (X–N) map for α-oxalic acid dihydrate (CSDI, 1969) in a direction perpendicular to the acid molecule plane. (*b*) Extension of mid-bond peaks in (X–N) map for DOX in a direction perpendicular to the carboxyl plane.

Table 12. *R.m.s. thermal displacements in* Å *along the principal axes* R_1, R_2 *and* R_3 *of the vibration ellipsoids and the angles between the principal axes and the unit-cell edges*

	R.m.s. displacement along			Angle between *R*3 and		
	*R*1 (Å × 10^3)	*R*2 (Å × 10^3)	*R*3 (Å × 10^3)	*a*	*b*	*c*
C	113 (3)	126 (3)	151 (3)	104 (3)°	67 (5)°	27 (5)°
O(1)	122 (3)	141 (4)	224 (3)	90 (1)	68 (2)	22 (2)
O(2)	114 (4)	143 (3)	219 (3)	106 (2)	74 (2)	22 (2)
N	140 (3)	159 (2)	167 (2)	138 (7)	52 (4)	105 (11)
O(*W*)	139 (5)	159 (4)	187 (4)	90	90	0
H(*W*)	139 (4)	173 (4)	206 (3)	77 (2)	96 (5)	14 (3)
H(1)	145 (3)	189 (3)	204 (3)	125 (3)	44 (7)	113 (8)
H(2)	149 (4)	193 (3)	205 (3)	33 (11)	96 (6)	123 (10)
H(3)	139 (4)	193 (3)	205 (3)	126 (2)	45 (9)	67 (12)
H(4)	150 (4)	187 (3)	206 (3)	41 (7)	109 (5)	56 (5)

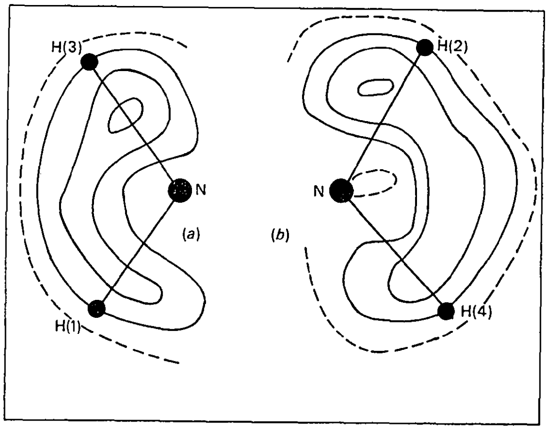


Fig. 5. (X–N) map of DOX in two H–N–H planes of the ammonium ion.

(ii) *Carboxyl plane*

The (X–N) synthesis was evaluated in the carboxyl plane for (DOX1 + DOX2)/2. The map is shown in Fig. 3. A simple hybridization scheme for the oxalate ion oxygen is:

$$(1s)^2\,(2sp^2)^1\,(2sp^2)^2\,(2sp^2)^2\,(2p\pi)^1$$
$$\leftrightarrow (1s)^2\,(2sp^2)^1\,(2sp^2)^2\,(2sp^2)^2\,(2p\pi)^2$$

with both trigonal sp^2 and π-bonding orbitals. The oxalate ion (X–N) map was similar to the X–N map in the plane of the oxalic acid molecule in CSDI (1969). Both maps had bond overlap peaks of 0·25 e.Å^{-3} at the centre of the C–C bond, overlap peaks of 0·1–0·15 e.Å^{-3} between the C and O atoms and peaks of 0·1–0·15 e.Å^{-3} in trigonal positions around the oxygen atoms (predicted lone-pair density). Corresponding negative peaks occurred at the atomic centres.

The peaks in the midpoints of the bonds were also extended in the present analysis in a direction perpendicular to the carboxyl plane (see Fig. 4). The extensions appeared to be greater than in α-oxalic acid dihydrate (CSDI, 1969) where the extension was considered to be due to π-bonding overlap. [The π-overlap should appear as an extension of the mid-bond peak in the perpendicular plane rather than peaks directly over the bonded atoms, as the σ-bonding leaves a large negative area above the atoms which is partly filled by the π contribution (O'Connell *et al.*, 1966)].

(iii) *Ammonium ion*

The (X–N) synthesis was evaluated in the NH(1)H(3) and NH(2)H(4) planes of the ammonium ion (Fig. 5). Overlap density between the N and H nuclei appeared to be present, which would be in accord with the observed deuterium asphericity shift, (Table 10). The features in each plane are similar.

The r.m.s. standard deviation of the X–N maps for (DOX1 + DOX2)/2 was about 0·05 e.Å^{-3}, according to the formula

$$\sigma(\Delta\varrho) = \langle \Delta\varrho^2 \rangle^{1/2}\,.$$

The above features in the (X–N) maps would appear to have some qualitative significance; they are also in accord with the above mentioned α-oxalic acid dihydrate results and hybridization theory.

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References

Busing, W. R., Martin, K. O. & Levy, H. A. (1962*a*). *ORFLS*. Report ORNL-TM-305, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

Busing, W. R., Martin, K. O. & Levy, H. A. (1962*b*). *ORFFE*. Report ORNL-TM-306, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

Coppens, P. (1967). *Science*, **158**, 1577.

Coppens, P. (1970). In *Thermal Neutron Diffraction*. Edited by B. T. M. Willis. Chapter 6. Oxford Univ. Press.

Coppens, P. & Hamilton, W. C. (1970). *Acta Cryst.* A**26**, 71.

Coppens, P. & Sabine, T. M. (1969). *Acta Cryst.* B**25**, 2442.

Coppens, P., Sabine, T. M., Delaplane, R. G. & Ibers, J. A. (1969). *Acta Cryst.* B**25**, 2451.

Coulson, C. A. (1970). In *Thermal Neutron Diffraction*. Edited by B. T. M. Willis. Chapter 5. Oxford Univ. Press.

Dawson, B. (1964). *Acta Cryst.* **17**, 990.

Delaplane, R. G. & Ibers, J. A. (1969). *Acta Cryst.* B**25**, 2423.

Duckworth, J. A. K., Willis, B. T. M. & Pawley, G. S. (1970). *Acta Cryst.* A**26**, 263.

Freeman, H. C., Guss, J. M., Nockolds, C. E., Page, R. & Webster, A. (1970). *Acta Cryst.* A**26**, 149.

Hamilton, W. C. (1964). *Statistics in Physical Science*. New York: Ronald Press.

International Tables for X-ray Crystallography (1962). Vol. III, pp. 202-203, Birmingham: Kynoch Press.

Mueller, M. H., Heaton, L. & Miller, K. T. (1960). *Acta Cryst.* **13**, 828.

Neutron Diffraction Commission (1969). Coherent Neutron-Scattering Amplitudes. *Acta Cryst.* A**25**, 391.

O'Connell, A. M., Rae, A. I. M. & Maslen, E. N. (1966). *Acta Cryst.* **21**, 208.

Padmanabhan, V. M., Srikantha, S. & Mehdi Ali, S. (1965). *Acta Cryst.* **18**, 567.

Pryor, A. W., Ellis, P. J. & Dullow, R. J. (1968). *J. Appl. Cryst.* **1**, 272.

Robertson, J. H. (1965). *Acta Cryst.* **18**, 410.

Zachariasen, W. H. (1967). *Acta Cryst.* **23**, 558.