References

- ALEXANDER, L. E. & SMITH, G. S. (1964). Acta Cryst. 17, 1195.
- BERGHUIS, J., HAANAPPEL, IJ. M., POTTERS, M., LOOPSTRA, O., MACGILLAVRY, C. H. & VEENENDAAL, A. L. (1955). *Acta Cryst.* 8, 478.
- COCHRAN, W. (1951). Acta Cryst. 4, 81.
- DAWSON, B. (1960). Acta Cryst. 13, 403.
- Howells, E. R., Phillips, D. C. & Rogers, D. (1950). Acta Cryst. 3, 210.
- IBALL, J. & WILSON, H. R. (1965). Proc. Roy. Soc. A 288, (1414), 418.
- KRACHOW, M. H., LEE, C. M. & MAUTNER, H. G. (1965). J. Amer. Chem. Soc. 87, 892.

LEPAGE, G. A. (1960). Cancer Res. 20, 403.

- MACINTYRE, W. M. (1964). Biophys. J. 4, 495.
- MAUTNER, H. G. & SHEFTER, E. (1967). J. Amer. Chem. Soc. 89, 1249.

- PAULING, L. (1960). The Nature of the Chemical Bond, 3rd Ed. p. 260. Ithaca: Cornell Univ. Press.
- PIMENTEL, G. C. & MCCLELLAN, A. L. (1960). The Hydrogen Bond. p. 262. San Francisco & London: Freeman.
- PULLMAN, B. & PULLMAN, A. (1963). Quantum Biochemistry, p. 791. New York: Interscience.
- RINGERTZ, H. (1966). Acta Cryst. 20, 397.
- ROBINS, R. K. (1964). J. Med. Chem. 7, 186.
- SINGH, C. (1965). Acta Cryst. 19, 861.
- SKIPPER, H. E. (1954). Ann. N. Y. Sci. 60, 315.
- STEWART, J. M. et al. (1964). Crystal Structure Calculations System, Computer Sciences Center, University of Maryland.
- STEWART, R. F., DAVIDSON, E. R. & SIMPSON, W. T. (1965). J. Chem. Phys. 42, 3175.
- SUTOR, D. J. (1958). Acta Cryst. 11, 83.
- SUTOR, D. J. (1963). J. Chem. Soc. p. 1105.
- WATSON, D. G., SWEET, R. M. & MARSH, R. E. (1965). Acta Cryst. 19, 573.

Acta Cryst. (1969). B25, 1338

The Crystal and Molecular Structure of 6-Mercaptopurine Monohydrate. A Second, Independent X-ray Diffraction Determination*

BY GEORGE M. BROWN

Chemistry Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830, U.S.A.

(Received 30 October 1968)

The structure of 6-mercaptopurine monohydrate, $C_5H_4N_4S.H_2O$ was determined by the heavy-atom method. The space group is C2/c; $a=15\cdot3314(4)$, $b=7\cdot7255(1)$, $c=12\cdot3397(2)$ Å, $\beta=101\cdot526(2)^\circ$, Z=8. These cell parameters differ significantly from those of Sletten, Sletten and Jensen (Acta Cryst. (1969) B25, 1330) Counter data were recorded for 3792 independent reflections (Mo radiation, $\sin\theta/\lambda \le 0.86$) and corrected for absorption. The use of anisotropic thermal parameters for the hydrogen atoms results in no significant changes in other structure parameters. The most reliable parameters for the C, N and S atoms are taken to be those from refinement in which the low-angle data $(\sin\theta/\lambda \le 0.55 \text{ Å}^{-1})$ are excluded. The corresponding bond lengths (with standard errors from 0.0011 to 0.0016) show some small but significant differences from those of Sletten *et all*. On analysis by the Schomaker-Trueblood method the thermal parameters of the C, N and S atoms are remarkably close to those found by Sletten *et all*.

Introduction

The determination at Oak Ridge (OR) of the crystal structure of 6-mercaptopurine monohydrate was undertaken for essentially the same reasons that prompted the parallel study at the University of Washington (UW)-see the accompanying paper of Sletten, Sletten & Jensen (1969). The discovery of the duplication of effort was made when a summary of the preliminary OR results was published in abstract form (Brown, 1967) at just the time that the manuscript of Sletten *et al.* on the UW results was refereed for publication in this journal. On interchange of information it was found that the agreement between the two sets of cell parameters was rather poor, though each set had apparently been determined with moderately high precision. Furthermore, even when the same set of cell parameters was used in calculating bond lengths from the two sets of coordinates, there were maximum and mean absolute differences of 0.007 and 0.0034 Å for bonds not involving hydrogen atoms, showing a level of agreement somewhat disappointing in view of the apparent high precision of the two determinations. On the other hand, the agreement for the bonds C–H, N–H, and O–H (maximum and mean absolute differences of 0.030 and 0.015 Å) could hardly have been expected to be better.

The cell parameters have now been redetermined in both laboratories with nearly the same results as before,

^{*} Research sponsored by the U.S. Atomic Energy Commission under contract with the Union Carbide Corporation.

1339

the discrepancies remaining much as they were. Further extensive refinement calculations have been performed at Oak Ridge, including some with the data of Sletten et al., which show that the small differences in bond lengths are inherent in the two sets of data and are not consequences of the different weighting schemes and different choices of the observational variable $(F_o versus F_o^2)$ in the two original least-squares determinations. The work at Oak Ridge evolved, because of the stimulus of the slightly different results from the University of Washington, into a study of the sensitivity of the derived structure parameters to some parameters or conditions of the least-squares refinement, especially the sensitivity to the weighting of the data. The structure parameters considered to represent the actual structure most accurately are those from a least-squares calculation using only the reflections having sin θ/λ values greater than 0.55 Å⁻¹.

Experimental

Crystal specimen

The 6-mercaptopurine monohydrate used was Mann's analyzed grade* ('paper chromatography, homogeneous,' 10.4% H₂O and 37.07% N found, 10.59% H₂O and 36.82% N calculated). Larger, faintly yellow crystals were grown from the supplier's fine crystals by slow cooling in a Dewar flask of a boiling saturated aqueous solution, as described by Hoogsteen (1956). Most of the crystals grown were needles, but the specimen used in the cell determination and in the collection of intensity data was cut from one of a few

* Supplied by Mann Research Laboratories, 136 Liberty Street, New York, N.Y. 10006.

chunky crystals in the batch. It measured approximately 0.5, 0.5, and 0.3 mm in the **a**, **b**, and **c** directions and was mounted approximately along **b**.

Unit cell and space group

From the preliminary X-ray investigation of Hoogsteen (1956) approximate unit-cell dimensions and the possible space groups were known, and the number of formula units per cell was known to be 8. In preliminary work with film methods this information was readily verified; the systematic extinctions, hkl absent for h+k=2n+1, h0l absent for l=2n+1, confirmed the possible space groups as Cc and C2/c. The choice of the centrosymmetric group C2/c was later made by application of the zero-moment test (Howells, Phillips & Rogers, 1950), using the whole set of three-dimensional data; and this choice was confirmed by the success of the subsequent structure determination.

Brief descriptions and results are shown in Table 1 for four different least-squares determinations of cell parameters as follows: OR-1, the original determination on the OR crystal; OR-2, a redetermination on the same crystal; UW-2, a redetermination on the UW crystal by Sletten et al.; UW-3, a redetermination on the UW crystal at Oak Ridge. Determinations OR-2, UW-2, and UW-3 were made because of the rather poor agreement of parameters between OR-1 and UW-1, the original UW determination (see Sletten et al., 1969). The differences between OR-1 and UW-1 of about 1 part in 600 for a, 1 part in 1100 for b, 1 part in 300 for c, and 0.124° for β are much larger than the estimated standard errors and were therefore considered to be unacceptable without further investigation.

Table 1. Least-squares determinations of unit-cell parameters for 6-mercaptopurine monohydrate

OR-1 and OR-2 are determinations on the Oak Ridge crystal; UW-2 and UW-3 are redeterminations on the University of Washington crystal done at Seattle and Oak Ridge respectively. In each case the angle data were from measurements on a four-circle diffractometer. A standard error* from the least-squares calculation appears in parenthesis adjacent to each parameter value. Unit weights were used in all determinations.

	OR-1	OR-2	UW-2	UW-3
No. of reflections	12	12	40	12†
2θ range	50–55°	119–132°	13-40°	80-94
Doublets resolved?	See text	Yes	No	No
Observations	$2\theta, \chi, \varphi$	$2\theta, \chi, \varphi$	2 heta	2θ
Wavelength (Å)	0.70926	1.54051	0.71069	1.54178
	(Mo $K\alpha_1$)	(Cu $K\alpha_1$)	(Mo $K \alpha_1 - \alpha_2$ wtd. mean)	(Cu $K \alpha_1 - \alpha_2$ wtd. mean)
a (Å)	15.3208 (29)	15.3314 (4)	15.3013 (13)	15.2975 (9)
Ь	7.7252 (16)	7.7255 (1)	7.7348 (11)	7.7313 (3)
с	12.3409 (19)	12.3397 (2)	12.3789 (13)	12.3719 (5)
β (°)	101.514 (14)	101.526 (2)	101.623 (8)	101.638 (5)
σ_1	0.020	0.006	_	0.007

* The effects of absorption were not considered in the least-squares determinations. For determinations OR-1 and UW-2 with Mo K α radiation, for which the absorption coefficient μ is only 3.84 cm⁻¹, no significant error results from absorption. However, for determinations OR-2 and UW-3 with Cu K α radiation, for which μ is 34.6 cm⁻¹, the error from neglect of absorption must be considered. The maximum error in the cell translations from this source is estimated to be only 1 part in 10,000 for either OR-2 or UW-3. Although this error is larger than the least-squares standard errors given in the Table for OR-2 and UW-3, it is still quite small and of no significance in bond length and angle calculations – nor would it be if it were actually larger than the estimate by a factor of 2 or 3.

† In this determination for nearly every reflection hkl used 2θ observations were made for both hkl and $\bar{h}kl$, each on both sides of the 2θ zero. In the least-squares calculation 12 average values of 2θ were entered.

In the process of making a new set of very careful angle measurements for OR-2, it was discovered that the OR crystal gave very broad peaks in θ -2 θ scans for reflections having reciprocal-lattice vectors near a*. For such reflections, even those at the highest 2θ angles accessible with the diffractometer (~160°), the α doublet of Cu K radiation could not be resolved; consequently, it was clear that some of the reflections used in OR-1 could not possibly have been resolved as assumed. (Unfortunately and unwisely, the scan check for resolution had been made for only 2 or 3 of the reflections.) In the new determination each one of the reflections used was scanned and found to be resolved. Thus the small parameter differences appearing between OR-1 and OR-2, which approach chemical significance only in the case of a, have a reasonable explanation.

In redetermining the cell parameters of the UW crystal, Sletten *et al.* first used new measurements on the same 17 reflections used originally (2θ range 13–30°, Mo $K\alpha$ radiation) and obtained essentially the same results as before, except that the standard errors derived were about $\frac{3}{4}$ as large. When another 23 reflections in the 2θ range 17–40° were included (see UW-2 in Table 1), the most significant change was an increase a of 1 part in 2200.

For the redetermination (UW-3) of the UW crystal at OR it was intended to use exactly the same apparatus and technique as used in OR-2. It proved impossible, however, to use exactly the same procedure, because the crystal shows more generally the broad peaks observed for a few reflections of the OR crystal. Every reflection which was scanned as a possible candidate for use in the least-squares determination turned out to be not resolvable into α_1 and α_2 components. It was decided to use data for reflections near $90^{\circ}2\theta$, recorded with the tube take-off angle set to 4° to minimize the separation of α_1 and α_2 , and, correspondingly, to use the weighted mean wavelength 1.54178 Å in the calculations. The parameters derived are very close to those of the original determination, UW-1, except in the case of c, for which the difference amounts to about $6\sigma(c)$ of UW-1. This difference is, however, only about 1 part in 1500 - hardly enough to be concerned about in its effect on derived distances and angles.

Thus we find that for both the OR and UW crystal specimens the revised parameters are at most slightly different from the original ones and that the discrepancies between the parameters for the two crystals remain about as they were. One must conclude that the two crystal specimens are indeed to be described by slightly different sets of cell parameters. For this paper the parameters of determination OR-2 have been used in the calculation of bond lengths and angles. Sletten *et al.* (1969) have quite reasonably chosen to use their original cell parameters (UW-1), which, as already discussed, are not very much different from those of UW-2 and UW-3.

It now appears probable that the differences in cell parameters arise as effects of a difference of purity between the two samples of 6-mercaptopurine monohydrate used. An inquiry by Professor Jensen to the supplier of the 6-mercaptopurine used at UW has elicited the information that chromatographic analysis of the batch indicated the presence of about 2% of unknown impurity.

Another possibility is that the differences in cell parameters are related to the difference in extent through the reciprocal lattice of the peak-broadening effect, which was possibly caused by different conditions of growth for the two specimens. On the other hand, the different extent of broadening may itself be related to the difference of purity.

Intensity data

Molybdenum $K\alpha$ radiation was chosen for recording the reflection intensities because of the low value of the absorption coefficient of the 6-mercaptopurine monohydrate (calculated $\mu = 3.84$ cm⁻¹) for this radiation. The X-ray tube was operated at 50 kV constant potential and 14 mA current. Intensity data were recorded for 3792 independent reflections with the Oak Ridge computer-controlled X-ray diffractometer (Busing, Ellison, Levy, King & Roseberry, 1968), with the $\theta - 2\theta$ step-scan method to 75° in 2θ (sin $\theta/\lambda = 0.857$ Å⁻¹) and an Ω step-scan method as well for the reflections in the range 0° to 30°2 θ .

In the θ -2 θ technique the step width was 0.05° in 2 θ , and the total width of scan for each reflection was 2° plus the width of the α doublet separation. The counting time was set at 10 seconds for the background counts taken at the beginning and end and at 2 seconds for all other points in each scan. The take-off angle at the X-ray tube was 3°, giving an effective target 0.13° wide by 0.32° high; the receiving aperture at the counter was 1.6 by 1.6°. A 0.004" niobium filter was used in recording data below 60° 2 θ ; no filter was used above 60° 2 θ .

In the Ω step-scan technique (Levy, 1966) three scans were run for each reflection: (1) with the counter arm set at the computed 2θ value of the reflection; (2) and (3) with the arm offset first lower and then higher in 2θ just enough to determine background. The net count of the reflection was taken as the integrated count of scan (1) minus the average of the integrated counts of (2) and (3). This procedure, with proper choice of take-off angle, counter aperture, and 2θ offset, provides more reliable background determinations for the low-angle reflections than does the $\theta-2\theta$ technique, in which the effect of the absorption edge of the filter just below the peaks for α -radiation makes proper background measurement impossible on the low-angle sides of the peaks.

The width of each Ω scan was 1°, the step width was 0.025°, and the count time at each point was 2 seconds. The take-off angle at the X-ray tube was set at 1° (effective target 0.04 by 0.32°), and the counter

slit and the counter offset $\Delta(2\theta)$ were set so that the three scans for each reflection would not overlap and so that the slit width and $\Delta(2\theta)$ (both the same) were always less than the angular separation between the α peak and the 2θ position corresponding to the absorption edge but the slit width was always wide enough to admit the α doublet. It was not possible to satisfy these conditions for the 2θ range 0° to 30° with a single value for counter aperture and $\Delta(2\theta)$; accordingly, scans were run in the range 0 to 15.2°2 θ with the aperture and $\Delta(2\theta)$ at 0.46° and in the range 15.2 to 30°2 θ with the aperture and $\Delta(2\theta)$ at 0.67°.

In the 2θ scans the intensity of a reference reflection, 008, was recorded after each group of 20 reflections as a check on the stability of the instrument and that of the crystal specimen. Similarly, for the Ω scans reflection 420 was used as a reference. There was no evidence from the reference intensities, or from visual inspection, of any deterioration of the crystal specimen. There were slow fluctuations (over many hours) of the standard intensities amounting to about 7.5%, probably resulting from imperfect stabilization of the line voltage to the X-ray generator and poor performance of the tube-current stabilizer. However, the short term fluctuations between the recording of reference intensities were no more than 1%.

As the data were recorded, corrections for coincidence loss were made on the counts of the individual points in the step scans before integration. The computer subroutine for this purpose used for the apparent dead time the value 1.8×10^{-6} seconds obtained in a calibration study by H. A. Levy & R. D. Ellison.

The raw data from the diffractometer were converted by calculations with the CDC 1604A computer to a set of structure-factor squares F_o^2 and statistical standard errors $\sigma_c(F_o^2)$. The variations of intensity of each reference reflection were assumed to represent slowly varying X-ray output; and the reference intensities were used to normalize the data to a single standard value of intensity for each reference reflection, a linear relation of X-ray intensity to time being assumed to hold between each pair of adjacent reference intensities. Absorption corrections calculated by the method of Busing & Levy (1957) were applied; the range of correction factors was only 1.12 to 1.16. The first three derivatives of the transmission factor with respect to μ were also computed, for subsequent use in correcting

					Hydrogen	No. of		Measures of	agreement ^a	
Cal. no.	Data set	Obs.	Weight scheme	sin θ/λ range	thermal parameters	observa- tions	$\overline{R(F)}$		Rw	σ_1
1	UW	F	UW	≤0.71	ISO.	1838 ^b 2186¢	0.038		0.037	2.01
2	UW	F	UW	≤0.71	ISO.	1860 ^d 1835 ^b	0.038 0.038	0·041 0·041	0.037	2·18 2·19
						2186°	0.024	0.043	0.042	2.25
3	UW	F^2	OR	≤0.71	ISO.	1859 ^a 2186°	0·038 0·054	0·044 0·046	0·087 0·093	1·64 1·62
4	OR	F	UW	≤0.71	ISO.	1877 ^d 1842 ^b	0.033	0.038	0.031	2·01 2·03
_						2182°	0.048	0.040	0.034	2.04
5	OR	F^2	OR	≤0.71	180.	2182 ^{c, a} 1967 ^e	0·040 0·034	0.039	0·076 0·076	1.57
6	OR	F^2	OR	≤0.86	ISO.	3792c, d	0.063	0.045	0.086	1.40
7	OR	F^2	OR	≤0.86	ANISO.	3139 ^e 3792 ^{c, d}	0.048 0.062	0·044 0·044	0·085 0·084	1.52
8	OR	F^2	OR	> 0.55	ISO.	3139 ^e 2792 ^c , d	0·048 0·101	0.048 0.078	0.083 0.093	1·50 1·06
9	OR	F^2	OR	≤0.55	ISO.	2182 ^e 1000 ^{c, d}	0·071 0·024	0·072 0·035 0·035	0·090 0·064 0·064	1·16 1·72

Table 2. Summary of various least-squares refinement calculations on 6-mercaptopurine monohydrate

^a Definitions:

$$R(F^m) \equiv \Sigma ||F_o|^m - |F_c|^m|/\Sigma |F_o|^m,$$

$$R_w \equiv [\Sigma w(|F_o|^m - |F_c|^m)^2/\Sigma w|F_o|^2m]^{1/2}$$

$$\sigma_1 \equiv [\Sigma w(|F_o|^m - |F_c|^m)^2/(n-p)]^{1/2}.$$

In these equations w is the weight of the observation F_o or F_o^2 , and p is the number of parameters fitted to the n observations; the weights and the structure factors are on the correct absolute scale established by the refinement. For each refinement calculation both R(F) and $R(F^2)$ are given; however, the values R_w and σ_1 are calculated either for m=1 or m=2 according as the observational variable in column 3 is F or F^2 .

- ^b Omits any observation included in refinement for which $F_c < 2\sigma(F_o)$.
- ^c Total number of observations in specified range of $\sin \theta / \lambda$.
- ^d Number of observations actually included in least-squares refinement.
- ^e Number of observations for which $F^2 > \sigma(F_0^2)$.

for extinction. The data for the Ω scans in the range 0° to 15°2 θ were scaled to the other data by use of the intensities of reflections 420 and 008, after correction for absorption. Different scale-factor identifiers were assigned to groups of data as follows: (1) Ω scans, 0 to 15.2°2 θ ; (2) Ω scans, 15.2 to 30°2 θ ; (3) 2 θ scans, 0 to 60°; 2 θ scans, 60 to 75° (unfiltered radiation).

The value $2 \cdot 0 \text{ Å}^2$ for the overall isotropic temperature factor and an approximate scale factor on the observations were obtained by the method of Wilson (1942).

Solution and refinement

The procedures by which the structure was found through application of the classic heavy-atom method and subsequently refined to the stage reported earlier (Brown, 1967) were nearly identical with those used by Sletten, Sletten & Jensen (1969). At this stage the usual reliability index calculated on F was 0.035 and the estimated errors of the lengths of bonds not involving hydrogen atoms were about 0.002 Å. No absorption corrections had been made, and only 1634 reflections ($\sin \theta/\lambda \le 0.65 \text{ Å}^{-1}$, equivalent to the Cu K α sphere) were included in the refinement. Data from Ω scans and $\theta-2\theta$ scans had simply been averaged.

Further refinement of the 6-mercaptopurine structure based on the OR data was made after the data had been corrected for absorption effects. The data from θ -2 θ scans below 30°2 θ were omitted in favor of the data from the Ω scans which had been run in the same range. The data set showed very slight effects of extinction, the maximum effect for the most intense reflection being a reduction of intensity of only about 15% below the calculated intensity. The least-squares program used includes a correction for extinction according to the second-order approximation of Zachariasen (1965). Correction factors for the values F_c^2 are calculated with Zachariasen's equation (14) to the third power in $X_{\rm H}$, the quantity g_2 being optimized with the other parameters. Only 20 reflections were affected by as much as 1%.

Least-squares refinement calculations based on the OR data were carried to convergence under a number of different conditions as regards weighting scheme, range in sin θ/λ of the data included, kind of thermal parameters used for hydrogen atoms, and so forth. Some additional refinement calculations were also performed with the UW data, which were kindly supplied by Sletten et al. for this purpose. In all of the calculations the scattering factors used were the same as those used by Sletten *et al.* The conditions of the various refinement calculations and the fits obtained are set forth in Table 2. In all of the calculations, except as noted below, the extinction parameter, the four scale factors for different groups of data, and the parameters of the hydrogen atoms were optimized, along with the usual nine parameters for each of the other atoms. Each calculation is assigned an arbitrary identifying number for purposes of discussion. It is not possible, of course, to show in detail how the structure parameters vary from one calculation to another; however, the results of the various calculations are placed in structural-chemical perspective by appropriate comparisons of the bond lengths derived from the coordinates (see Table 3).

The entries in Table 2 under calculation (1) describe the refinement of Sletten, Sletten & Jensen (1969) with

 Table 3. Comparison of bond lengths in the 6-mercaptopurine molecule as calculated from the parameters from the various refinement calculations of Table 2

Column 1-3, for example, contains the differences in bond lengths ($Å \times 10^3$) between calculations 1 and 3 (1 *minus* 3). The atoms are numbered as in Figs. 4 and 5. The last column gives the standard errors of the bond lengths from the parameters of calculation 8.

							••••		-						
Bond	1-3	1-4	1–5	1-7	1-8	3–5	4-5	5-6	5–7	5-9	6-7	6–8	7–8	8-9	σ
C(2) - N(3)	-1	4	2	1	-2	3	-2	0	-1	0	-1	-4	-3	4	1.6
N(1)-C(2)	0	-2	-2	-3	-8	-2	0	Ó	-1	6	-1	-6	-5	12	1.3
N(9)-C(4)	0	-1	-2	-2	-5	-2	-1	0	0	-1	0	-3	- 3	2	1.4
N(3)-C(4)	1	-4	-1	-1	3	-2	3	0	0	-3	0	4	4	-7	1.3
C(4) - C(5)	-1	7	4	4	3	5	-3	1	0	7	-1	-2	-1	8	1.1
C(5) - N(7)	0	-4	-2	-2	-3	-2	2	0	0	0	0	-1	-1	1	1.4
C(5) - C(6)	-2	-2	-6	-6	-8	4	-4	0	0	2	0	-2	-2	4	1.4
C(6) - N(1)	0	4	6	9	12	6	2	2	3	-2	1	4	3	8	1.3
C(6)–S	1	4	3	1	-3	2	-1	-1	-2	1	-1	- 5	-4	7	0.9
C(8) - N(9)	-2	1	-2	-2	-7	0	-3	-1	0	4	1	-4	- 5	9	1.4
N(7)-C(8)	0	-2	-2	-2	-6	-2	0	1	0	4	-1	-5	-4	8	1.5
Avg. ⊿	0.7	3.2	2.9	3.0	5.5	2.7	1.9	0.2	0.6	2.7	0.6	3.6	3.2	6.4	1.4
R.m.s. ⊿	1.0	3.6	3.3	3.8	6∙2	3.2	2.3	0.8	1.2	3.4	0.8	3.8	3.3	7.1	
C(2)-H(2)	0	-24	-15	-11	-11	-15	9	1	4	20	3	3	0	-16	12
C(8) - H(8)	7	-22	-7	-9	5		15	6	-2	-2	- 8	6	14	14	13
N(1)-H(1)	9	-40	-25	-15	-26	- 34	15	-6	10	7	16	5	-11	8	15
N(7)-H(7)	-13	-36	-16	-7	-12	- 3	20	-1	9	31	10	5	- 5	27	15
OH(10)	-19	-32	-28	- 37	-25	-9	4	2	-9	0	-11	1	12	-3	16
O—H(11)	12	-16	-8	-15	-12	-20	8	2	7	-13	-9	-3	6	-12	18
Avg. [⊿]	10	28	17	16	15	16	12	3	7	12	10	4	8	13	15
R.m.s. 1	12	30	18	19	17	19	13	4	7	16	10	· 4	9	15	

the UW data. Calculation (2) in Table 2 was in effect a replicate of (1) performed at OR, partly to check results from the OR least-squares computer program against those from the UW program and partly to check a subroutine used in calculation (4) for computing weights according to the UW scheme. The parameters from (2) are nearly identical with those from (1). There is an inconsistency in the fact that the standard errors of parameters from (2) are generally 5 to 10% higher than those from (1) and the σ_1 values for (2) are also higher by about 10%, but the weighted discrepancy indices R_w are equal for (1) and (2).

In calculation (3) with the UW data, the quantities F_o^2 were used as observations, and the weighting scheme was the one usually found appropriate in this laboratory:*

$$\sigma^{2}(F_{o}^{2}) = \sigma_{c}^{2}(F_{o}^{2}) + (0.03F_{o}^{2})^{2}$$
$$w(F_{o}^{2}) = 1/\sigma^{2}(F_{o}^{2}) .$$

The variance $\sigma_c^2(F_o^2)$ is, as defined earlier, the purely statistical variance. The quantity F_o^2 in the correction term is systematically somewhat smaller than the term $\sigma_c^2(F^2)$ used (in effect) by the UW group, but the important difference between the OR and UW weighting schemes is the use of the coefficient 0.03 in the former rather than the factor 0.01 in the latter. This term in the OR scheme has the effect of diminishing to a greater degree the importance of the larger F_o^2 values relative to that of the smaller ones.† It was intended to allow, at least in part, for deficiencies in the model, as well as for imperfect stability of the circuitry. It turns out that the parameters and standard errors from (3) are only trivially different from those of (2).

Calculation (4) on the OR data corresponds to (1) and (2) on the UW data. The fit is slightly better than for (2) and the parameter errors are generally about 10% smaller than in (1).

Calculation (5) on the OR data corresponds to (3) on the UW data; that is, the observational data were the quantities F_{σ}^2 and the OR weighting scheme was used. The fit in (5) is distinctly better than in (4), just as it was better for (3) than for (2). The standard errors of coordinates from (5) are about 10% smaller than those from (4).

Comparison 1-3 in Table 3 shows how little the derived bond lengths change when one changes from the UW to the OR refinement scheme. Comparisons 1-4 and 3-5 show that appreciable differences in bond lengths remain between the structures based on the UW and OR data sets for the same $\sin \theta/\lambda$ range, no matter which scheme is used in refinement. The difference of purity between the OR and UW specimens

may be, in part, the source of these differences as well as those between the two sets of cell parameters. See, however, the discussion at the end of this section regarding the comparison of the two sets of F_a^2 data.

From comparison 4-5 it appears that the bond lengths derived from the OR data are slightly more sensitive to the choice of refinement scheme than those from the UW data (comparison 1-3). Comparisons 5-6, 5-7, and 6-7 in Table 3 show that the effects on the molecular geometry of increasing the sin θ/λ cutoff value from 0.714 to 0.877 Å⁻¹ and of using anisotropic instead of isotropic thermal parameters for the hydrogen atoms are trivial.

The conditions of calculation (6) were the same as those of (5) except that the full set of OR data was used in (6). Number (7) differed from (6) only in that anisotropic thermal parameters were used for the hydrogen atoms. A calculation (7a), not represented in Table 2, differed from (7) in that the ~ 200 negative values of F_a^2 (resulting from statistical fluctuations and instrumental errors) were retained in the least-squares calculations, rather than replaced by zero values as in all the other refinements with the OR data. The standard errors from (6), (7), and (7a) are all about the same and generally somewhat smaller than those from (5). The values of σ_1 from (6), (7), and (7a) are about 10% lower than σ_1 for (5), indicating a better fit, on the average, to the high-angle data than to the lowangle data. The parameters (other than the thermal parameters of the hydrogen atoms) from (6) and (7) are only trivially different; the parameters from (7) and (7a) are nearly identical.

It is of some interest that the quadratic forms of the temperature-factor exponents of the hydrogen atoms all remained positive-definite in calculation (7), since attempts at refinement from X-ray data of anisotropic thermal parameters have rarely been successful. The *R* factor ratio test (Hamilton, 1965) indicates that the slight improvement in fit from (6) to (7) is significant (probability >99.5%). It is clear, however, that the thermal parameters of H(1) and H(7), on N(1) and N(7) respectively, cannot be interpreted in terms of thermal motion only, for the probability ellipsoids of H(1) and H(7) have odd shapes and orientations (see Fig. 1), which may be related to deficiencies in the model as it describes the electron density in the molecule.

Following calculation (7) a partial-difference Fourier synthesis (not shown) in the average plane of the nearly planar 6-mercaptopurine molecule was computed by use of magnitudes $|F_o| - |F_c|$ calculated without including the hydrogen contributions, and signs calculated including these contributions. There is some elongation in the peaks representing H(1) and H(7), though not so much as might be expected from Fig.1. A striking feature of the map, which is quite similar to the composite difference map shown by Sletten *et al.* and also similar to the final map (see Fig.2) subsequently prepared in this analysis, is that there is a low peak (0.15 to 0.40 e.Å⁻³, to be compared with peaks of 0.82

^{*} It was not possible to calculate exactly from the values F_o and $\sigma(F_o)$ supplied by Sletten *et al.* the weights according to the OR scheme; however, a satisfactory approximation was made by correcting each variance $\sigma^2(F_o^2)$ by the addition of the term 0.0008 F_o^4 .

[†] For the largest values F_{o^2} the OR weights are $\sim \frac{1}{2}$ those of the UW weights; for the weakest reflections, $\sim \frac{1}{3}$.

to $0.87 \text{ e.}\text{Å}^{-3}$ for the hydrogen atoms) centered near the middle of each ring bond.

The appearance in the difference map of the 'bonding' peaks suggests the desirability of modifying the model for structure-factor calculation so as to take account of bonding effects on electron distribution, as has been done in a few X-ray structure studies (see, for example, Cady & Larson, 1965, and Fritchie, 1966). As a practical matter, however, it is not possible for me to undertake the necessary computer programming at this time. Instead I have chosen to make further refinement calculations excluding the low-angle reflections, for which the calculated structure factors are most aberrant because of the use of scattering factors for spherical atoms. Such a procedure was originally suggested by Jeffrey & Cruickshank (1953) as a means of obtaining more reliable thermal parameters; but, as pointed out by Cruickshank (1956), it should in principle result in more reliable coordinates as well. The OR data set for 6-mercaptopurine is particularly suitable for this purpose, since it extends unusually far in sin θ/λ , at least for an organic crystal.

In least-squares calculation (8) the reflections included were limited to those having $\sin \theta / \lambda$ greater than 0.55 Å-1. The starting parameters were from calculation (6), and the scale factors, extinction parameter, and hydrogen-atom parameters were held fixed. It appears that the parameter changes from (6) to (8) are significant, though small, for the R factor ratio test shows that the parameters from (8) fit the limited data set much better than those from (6) (probability >99.5%). The standard error of fit is markedly smaller than for calculation (6), 1.06 instead of 1.38, showing that the model fits the high-angle data better than the low-angle data, if one assumes that the relative weights for high and low angle data have been assigned correctly. This point is emphasized by calculation (9), including only the reflections having $\sin \theta / \lambda \le 0.55 \text{ Å}^{-1}$, for which the value of the standard error of fit is 1.72. Although the fit is better for (8) than for (6), the standard errors derived for (8) are higher by about 20%. Additional least-squares calculations (not represented in Table 2) with lower limits in $\sin \theta/\lambda$ of 0.50 and 0.59 Å⁻¹ established that the structure parameters are insensitive to the precise choice of limit in this range.

A partial-difference Fourier synthesis in the average molecular plane was computed following refinement calculation (8), including the terms with $\sin \theta/\lambda \le 0.71$. The resulting map (see Fig.2) shows peaks for the hydrogen atoms from 0.87 to 0.90 e.Å⁻³ in height and bonding peaks of 0.21 to 0.46 e.Å⁻³. Curiously, the peaks H(1) and H(7) do not have shapes that are consistent with the corresponding ellipsoids of Fig.1. Another map computed only with terms having $\sin \theta/\lambda \le 0.55$ shows the same general features and has bonding peaks 0.82 as high, on the average, as those in Fig.2. It thus appears that most of the perturbing effects of the deficiencies of the structure model have been avoided by the omission of the low-angle data in cal-



Fig. 1. The 50% probability thermal ellipsoids from the thermal parameters of least-squares calculation (7). View is in the direction of the normal to the molecular plane for each molecule.



Fig. 2. Partial difference maps in planes of 6-mercaptopurine and the water molecule, after least-squares calculation (8). Terms were included for those reflections having $\sin \theta/\lambda \le$ 0.71. The lowest positive contour is at the 0.1 e.Å⁻³ level; the highest negative contour is at the -0.1 e.Å⁻³ level.



Fig. 3. Plot showing the agreement between the OR F_{o^2} data and the UW F_{o^2} data as a function of $(\sin \theta/\lambda)^2$. $\langle F_{o^2} \rangle_{OR}$ and $\langle F_{o^2} \rangle_{UW}$ denote averages over small ranges of $(\sin \theta/\lambda)^2$.

culation (8). It therefore seems reasonable to regard the parameters from (8), shown in Table 4, as representing most accurately the actual structure of 6-mercaptopurine monohydrate. Similarly, Furberg & Jensen (1968) have concluded that the best parameters for the crystal structure of thiocytosine result when only the higher-order data are used. In Table 4 the extra entries for the hydrogen coordinates are empirically corrected values (see text below under the heading *Molecular structure*).

Table 5 compares the values F_o with the values F_c computed from the parameters of calculation (8). The values of F_o are on the scale established in the refine-

ment and include the computed extinction corrections. The standard error $\sigma(F_o)$, computed as $\sigma(F_o^2)/2F_o$, is given for each reflection for which $F_o^2 > \sigma(F_o^2)$; the error $\sigma(F_o^2)$ is given for each reflection (marked W) for which $F_o^2 \le \sigma(F_o^2)$.

At the end of the refinement process, the sets of F_c^2 data from the OR and UW determinations were systematically examined over their common range of $\sin \theta / \lambda$ for their agreement as a function of diffraction angle. Reflections which were weak in either set $(F_o^2 \le \sigma(F_o^2)$ for OR data; $F_o^2 < 2\sigma(F_o^2)$ for UW data) were excluded in the comparison. The OR data were scaled as in Table 5, and the UW data as in Table 6 of Sletten et al. (1969). Fig.3 shows a plot against $(\sin \theta/\lambda)^2$ of $\langle F_{\theta}^2 \rangle_{OR}/\langle F_{\theta}^2 \rangle_{UW}$, where the brackets $\langle \rangle$ denote an average over a small range of $(\sin \theta / \lambda)^2$. Each point in the plot represents roughly the same number of reflections. It is clear from the Figure that systematic errors of measurement were made in the determination of one of the other, or both, of the sets of data. The value of the discrepancy index R = $\Sigma |F_{o,OR}^2 - F_{o,UW}^2| / (\frac{1}{2}) \Sigma (F_{o,OR}^2 + F_{o,UW}^2)$ is 0.092; the corresponding index computed on F_o instead of on F_o^2 is 0.070. These indices in comparison with the corresponding discrepancy indices given in Table 2, which are smaller by a factor of ~ 2 , demonstrate that the R values can give a misleading impression as to the quality of a crystal-structure determination. In one or the other of the two determinations, or perhaps in both, the parameters have been adjusted in such a way as

Table 4. Final parameters of the structure of 6-mercaptopurine monohydrate

The elements B_{ij} form the symmetric matrix **B** in the temperature factor exp $\{-\frac{1}{4}[h_ib_i]^T \mathbf{B}[h_ib_i]\}$, where each of the three elements of the one-column matrix $[h_ib_i]$ is the product of a reflection index and the corresponding reciprocal translation (Å⁻¹). The additional entries in italics for the coordinates of the hydrogen atoms are empirically adjusted values (see text under heading *Molecular structure*).

,	15	st digit = tenths o	ligit			1st digit=	= units digit		
	x	<i>y</i>	z	$\overline{B_{11}}$	B ₂₂	B ₃₃	B ₁₂	B ₁₃	B ₂₃
S	44648 (2)	66250 (5)	35602 (3)	269 (1)	330 (1)	230 (1)	-034(1)	127 (1)	013 (1)
C(2)	36701 (8)	66858 (18)	64218 (8)	244 (4)	261 (4)	151 (2)	-009(3)	031 (2)	-010(3)
C(4)	26259 (7)	51572 (14)	53004 (7)	186 (3)	205 (3)	145 (2)	004 (2)	055 (2)	010(2)
C(5)	30600 (7)	53754 (14)	44193 (7)	194 (3)	205 (3)	137 (2)	-005(2)	052 (2)	000(2)
C(6)	38605 (7)	63068 (14)	45370 (8)	194 (3)	202 (3)	165 (2)	004 (2)	057 (2)	022(2)
C(8)	18392 (8)	38889 (18)	39083 (10)	233 (4)	259 (4)	211(3)	-034(3)	037 (3)	-027(3)
N(1)	41145 (7)	69536 (14)	55884 (8)	206 (3)	239 (3)	179 (2)	-022(2)	030 (2)	-002(2)
N(3)	29289 (7)	58078 (16)	63318 (7)	240 (3)	271 (4)	139 (2)	-005(3)	055 (2)	003 (2)
N(7)	25409 (7)	45494 (15)	35347 (7)	240 (3)	259 (3)	150 (2)	-018(3)	044 (2)	-028(2)
N(9)	18610 (7)	42230 (16)	49729 (8)	212 (3)	266 (4)	206 (2)	-030(2)	068 (2)	-012(3)
0	44130 (9)	09908 (19)	36929 (10)	282 (4)	343 (4)	282 (4)	-082(3)	101 (3)	008 (3)
H(1)	4587 (10)	7623 (18)	5755 (12)	40 (3)					
	4684	7759	5789						
H(2)	3939 (8)	7255 (15)	7093 (10)	29 (3)					
	3974	7328	7180						
H(7)	2637 (10)	4510 (19)	2861 (12)	46 (3)					
	2655	4503	2733						
H(8)	1380 (9)	3231 (15)	3456 (10)	28 (3)					
	1319	3144	3396						
H(10)	4067 (11)	1023 (21)	4122 (12)	45 (4)					
	3988	1030	4220						
H(11)	4525 (11)	-0068(23)	3678 (13)	52 (4)					
	4543	-0236	3676						
				$g_2 = 1.02$	2×10 ⁻⁶				
				$\sigma(g_2) = 0.22$	7×10-6				

Table 5. Observed and calculated structure factors for 6-mercaptopurine monohydrate

For each reflection, identified by the indices K and L of a subheading and the running index H, the values of $F_0 \times 10$ and $F_c \times 10$ are given. The standard error $\times 10$ of F_0 (see text) is given under the heading SG, except that for each reflection marked W, for which $F_0^2 < \sigma(F_0^2)$, the standard error $\times 1$ of F_0^2 is given instead.

и 895 CRLSG и 085 CRLSG и 885 CRLSG и 88	S CALSG H 885 CALSG H 885 CALSG I	1 845 CALSG H 885 CALSG H 885 LALSG	н өөз cause н өөз cause н өвз cause 'н өвз cause
жив н 0 0 жив -12 241-240 4 13 165 173 3 1 36 2 714 -685 11 -10 288 -287 5 15 85 -79 4 3 38 4 275 265 4 -8 390 -374 6 174 21 10 7 5 7 6 987 -991 15 - 6 129 -174 4 3 6 174 21 10 7 5 7	9 370 6 4 1037 1036 16 -16 208 -205 4 1 7 394 6 6 92 90 2 -14 416 -411 6 8 6 86 4 8 40 54 3 -12 257 -247 4 8	1 57 56 7 -7 439 431 7 -17 192 193 4 5 92 89 5 -5 351 347 6 -15 195 195 4 5 54 49 8 -3 436 413 7 -13 124 -128 4	-5 58 -50 7 6 274 -268 4 -8 90 95 5 17 87 83 5 -3 127 122 4 844 11 16 5 -64 12 -26 7 19 108 109 4 -1 65 -76 5 10 200 191 4 -4 39 29 9 21 35 -39 11 -1 65 -65 5 200 191 4 -4 39 29 9 21 35 -39 11
8 316 -924 14 -4 95 93 4 214 13 -22 7 9 12	0 -122 4 12 103 112 3 -8 348 345 5 12	1 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2	3 18 - 23 8 14 49 44 7 0 118 -113 4 - 234 0 - 23 8
10 294 -314 5 -2 95 95 4 234 0 -25 8 11 3	7 -46 10 14 157 -176 3 -6 520 515 8 14		5 29 - 37 14 16 48 - 44 9 2 88 -87 5 - 214 13 5 - 23 8
12 308 -298 5 0 455 44 3 7 +5 +5 13 10	5 -102 5 16 133 139 4 -4 392 402 6 884		лин н 3 20 лин 18 52 44 5 4 140 - 149 4 - 19 75 - 70 6
14 220 -213 4 2 133 134 4 -25 61 -63 7 15 3	N -32 10 18 12N -13N N -2 796 800 12 -20	57 -53 7 7 160 158 3 -3 36 9 3	-11 74 80 6 20 61 -58 7 6 131 133 4 -17 101 -111 5
16 480 503 7 44 0 1 7 -23 60 50 6 174 2	5 39 8 20N 23 26 8 0 183 -181 3 -18	9 145 -146 4 9 268 261 4 -1 307 301 5	-94 23 -19 8 *** H 4 7 *** 84 0 20 7 -15 59 -60 6
184 18 17 13 6 83 -71 5 -21 148 141 4 19 3	0 38 15 22 7N -6N 5 2 293 -31N 5 -18	3 35 23 9 11 32 -18 7 1 556 615 9	-7 76 22 6 -24 19 -40 8 19 85 77 4 -13 31 -29 10
20 404 133 5 6 35 -54 5 -13 105 4 4 -234 7 22 46 53 13 10 65 -67 5 -17 33 87 4 -234 7 24 52 -38 12 12 56 -51 6 -15 81 -85 4 -21 15	1 12 19 24 10 -25 8 4 114 -120 3 -1 5 7 8 200 H 2 1 200 6 657 -653 10 -1 1 -145 4 -24 86 89 5 8 196 -194 4 -1 1 0 10 -22 00 87 1 10 108 -104 4 -1	1 218 -220 4 13 82 81 4 3 97 92 4 5 52 -55 8 15 129 135 4 5 49 -54 9 -54 5 67 80 6 17 48 -32 8 7 43 -46 7	-5 159 156 4 -22 97 -104 4 12 33 28 14 -11 203 139 4 -34 15 23 10 -20 46 -40 7 14 53 -40 8 -9 364 373 6 -1 57 -41 7 -18 53 34 7 мая H 4 15 мая -7 154 152 3
-25 102 100 5 ++ 0 16 ++ -11 126 -116 3 -17 4 -24 51 -442 7 -204 0 10 10 -9 485 -495 8 -15 23 -22 213 221 4 -18 10 -94 5 -7 532 -531 8 -13 11	1 -4 10 -20 113 -109 4 12 310 305 5 -4 1 233 4 -18 200 -197 4 14 352 351 6 -4 5 113 4 -18 205 -221 4 154 20 -21 8 -2	204 205 205 5 19 32 24 7 9 132 -119 4 204 246 4 21 84 -75 5 11 121 -119 4 14 15 -4 7 23 38 3 10 134 13 -20 8 45 45 -4 7 23 38 3 10 134 13 -20 8	1 117-110 4 -18 173 172 4 -20 133 177 -3 73 -369 6 0 761 -770 12 -12 236 236 5 -16 160 162 4 -1 555 -558 9 2 513 -512 8 -10 279 -248 5 -14 110 -103 4 1 37 -39 5
-20 142 150 4 -16 191 -191 4 -5 227 -209 4 -11 13 -18 132 -133 4 -144 18 -27 6 -3 567 603 9 -9 33 -16 323 -314 5 -12 243 -240 5 -1 315 318 5 -7 10	2 131 4 -14 392 -405 6 18 30 -23 11 5 -341 5 -12 236 239 4 204 0 -19 7 9 124 3 -10 255 247 4 22 34 -13 11	א 13 – 17 - 7 – 25₩ 13 29 8 17₩ 0 19 8 2939 – 295 5 – 23 94 89 4 19 31 12 13 92 – 92 5 – 21 143 133 3 אמיי 1 11 אמיי 92 – 92 5 – 21 143 133 3 אמיי 1 11 אמיי	u 384, 373 6 -8 181 -162 3 -12 146 146 3 3 32 -33 7 6 91 -88 3 -6 139 -135 3 -10 130 -128 4 5 400 405 6 8 488 472 9 - 4 238 -227 4 -8 200 -128 4 7 114 -127 3
-19 002 -053 10 -10 101 32 4 1 510 513 0 -3 24 -12 351 -333 5 -6 352 -88 5 3 542 527 8 -3 24 -10 307 303 5 -6 352 584 6 5 134 -120 3 -1 22 -8 321 306 5 -4 158 170 4 7 303 -313 5 1 6	11 305 5 -6 1014 1014 15 -244 19 34 8 4 5 +226 4 -4 64 69 2 -22 59 -63 7 10 5 6 68 7 -2 64 54 10 -20 84 -63 7 10	5 102 -107 5 -19 372 18 11 -234 0 -2 8 9 154 154 4 -17 94 99 4 -214 27 28 7 5 3 45 7 -15 108 109 4 -19 59 68 5 30 3 15 7 -15 108 109 4 -19 59 68 5	10 84, 70 3 -24 0 -99 4 -6 59 -52 7 9 179 169 3 12 308 325 5 0 72 -66 4 -4 89 -64 5 11 44 31 7 144 19 -7 6 2 114 106 3 -24 0 5 7 13 94 95 4 16 160 -169 4 290 5 0 196 190 4 15 90 -67 5
-6 978 955 15 -24 0 -10 7 9 369 -392 6 3 10	2 104 4 0 123 -124 2 -18 59 -64 5 1	i 48 42 8 -11 552 552 9 -15 80 81 5	18 177 172 4 6 41 131 6 7 48 32 8 17 104 106 5
-4 1691 1630 25 0 76 75 10 114 3 26 5 5 15	0 151 4 2 3099-3196 47 -16 55 56 6 ma	H 2 16 ### -9 694 -676 11 -13 40 29 8	20 26 141 3 8 311 239 5 4 231 234 4 134 0 74
-2 551 -570 8 24 -0 -3 8 134 0 12 5 74	0 5 7 4 187 -206 3 -144 5 -7 6 -21	H 0 -1 8 -7 216 228 4 -11 329 -333 5	22 28 15 13 10 55 45 6 6 168 171 4 21 29 2 13
0 1402 1449 21 4 261 - 3 5 15 53 - 46 5 94	0 7 7 6 327 - 309 5 - 12 313 323 5 - 14	3 57 59 7 -5 147 164 3 -9 183 205 4	2414 25 23 8 12 171 - 174 4 8 55 - 446 7 345 H 5 545 7
2 917 - 650 14 5 56 - 63 6 174 0 17 7 11 10	1 - 95 4 8 629 626 10 - 10 101 93 3 - 14	5 73 77 5 -3 729 741 11 -7 249 -256 4	445 H 1 545 H 1 51 - 144 5 1 10 35 - 9 10 - 234 - 0 0 8
4 1868-1881 28 8 206 - 213 4 194 25 23 6 13 8	1 77 4 10 535 582 9 - 8 59 64 4 - 14	1 15 115 4 -1 431 427 7 -5 137 133 3	-24 51 - 47 8 16 38 - 29 12 124 16 - 25 8 - 21 111 - 111 4
5 707 - 708 11 10 256 252 4 21 80 71 5 13 8	8 - 92 5 12 121 123 3 - 6 59 64 10 - 14	1 15 115 4 -1 431 427 7 -5 137 133 3	-29 39 - 0 11 - 112 - 124 5
8 558 – 532 9 12 31 13 13 23 46 39 9 174	0 8 8 14 359 354 6 -4 97 -98 3 -11) 151 149 4 3 176 -165 3 -1 110 -111 3	-20 57 58 6 20 21 1 8 -18 0 16 8 -17 10 -10 4
10 515 511 9 жил H 0 18 жил ля H 1 6 ля ля H	1 13 xax 164 22 4 6 -2 187 -172 3 -1	3 183 -176 4 5 240 -257 4 1 254 252 4	-18 154 154 4 88 18 -18 45 37 8 -15 83 -88 5
12 596 591 9 – 16, 67 – 59 7 – 25 43 – 91 9 – 23 3	7 47 11 18 268 -273 5 0 190 181 4 -1	31 14 12 7 488 -486 7 3 61 -71 6	-16 131 131 4 -24 18 32 8 -14 76 75 5 -13 86 63 5
16 368 372 6 -144 12 -22 7 -51 74 70 5 -19 13 18 77 -92 5 -12 114 114 4 -19 107 107 4 -17 12 20 172 -173 4 -10 310 310 5 -17 132 127 4 -15 9	13 132 4 22 85 -73 4 4 153 130 3 -2 3 113 4 24 0 8 6 49 60 5 7 -92 5 ### H 2 2 ### BM 4 -1 5	1 87 -90 5 9 66 79 4 5 187 -173 4 2 38 -32 10 11 329 -335 5 7 42 46 8 24 0 10 8 13 73 79 4 9 94 -97 5 5 59 55 8 15 32 19 10 11 138 137 4	-14, 115 -125 4 -22 146 -147 4 -12 31 12 11 -11 35 18 8 -12 158 187 3 -20 88 -83 5 -10 149 146 3 -9 579 579 5 -10 268 -274 4 -18 35 -38 11 -8 145 -148 3 -7 344 338 5 -8 267 -272 4 -18 58 100 4 -5 123 -116 4 -5 116 -125 3
22 102 -113 4 -8 51 -24 7 -15 72 71 4 -13 90 24 38 -31 11 -6 112 112 4 -13 130 -132 3 -11 29 ann H O 4 ann -4 191 -197 4 -11 62 62 4 -9 15 -26 80 80 5 -2 115 -117 4 -0 730 -735 5 -7 10	S-303 S =24 80 =76 S 10 93 91 4 1 N =291 S =224 11 0 7 124 4 34 7 1 3 =153 4 =20 95 90 S 14 32 =35 13 1 7 149 3 =184 0 9 7 14 32 =35 13 1	M 0 -17 7 17 113 113 4 13 33 -16 10 5 207 210 4 19 300 110 4 15 31 -16 11 5 37 -136 4 21 70 66 5 174 13 -21 8	-5 165 195 3 -14 52 57 5 -4 126 -129 5 -3 61 -60 4 -4 336 -326 5 -12 205 209 4 -2 85 7 4 5 -1 405 -407 6 -2 223 -228 4 -12 205 209 5 5 5 5 3 3 -43 10 1 104 -102 3
-24, 134, -130, 4, 0, 127, -103, 7, -7, 146, 127, 3, -5, 17	1 175 4 -16 257 266 4 184 20 -10 7 1	2 65 -65 6 838 H 3 5 888 -23 110 103 4	2 94 - 57 2 - 5 247 - 258 4 6 157 152 4 7 455 5
-22, 73, -69, 5, 2, 41, 41, 9, -5, 439, 495, 8, -3, 36	0 359 6 -14 47 34 6 204 24 5 8 44	H 2 17 888 -25H 0 -8 8 -21 81 80 5	4 1276 1242 19 - 4 253 - 258 4 6 157 152 4 7 455 47 7
-20, 111, -115, 4, 4, 56, -55, 7, -3, 482, 473, 7, -1, 5	5 54 6 -12 54 58 5 44 H_2 9 44 -11	H 14 10 8 -23 69 60 5 -19 176 175 4	5 532 - 552 8 - 2 238 - 244 4 8 64 0 3 8 9 119 139 3
-18 195 -189 4 6 144 120 4 -3 129 144 3 1 3 -18 74 -87 4 8 121 -118 4 1 23 29 4 34 1 -14 330 -349 5 Ame H 0.20 Ame 3 28 -14 4 5 7 -12 777 771 12 -124 0 -10 9 5 274 247 5 7 11	16 -13 9 -104 0 -22 2 -24 67 81 6 -11 5 -9 6 -8 234 -229 4 -22 36 -44 10 -11 0 -67 5 -6 129 -120 3 -20 176 -180 4 -1 2 -117 4 -4 219 217 4 -18 239 -237 4 -11	5 46 38 8 -214 24 14 7 -17 234 -232 4 1 120 123 4 -194 0 -10 8 -15 154 -156 4 2 151 157 4 -174 0 -9 6 -13 377 -367 6 1 71 67 6 -13 177 -367 6	8 133 -128 3 0 404 -411 6 10 39 -30 10 11 137 141 4 10 145 -140 3 2 150 175 3 max H 4 17 max 13 103 101 4 12 254 -269 4 4 103 184 4 -16 55 -80 7 15 75 -77 6
-10,1017,1015,15,-10, 65, 65, 7, 7, 188,-175, 3, 9, 7 -8,676,652,10, -8,106,-104,5, 9,1199,208,4,11, 3 -6,48,-72,2, 8, 4, 6, -77, 9, 11,523,-549,8, 124	9 -80 5 -2 296 -272 5 -16 109 -111 4 - 3 -12 9 0 512 500 8 -14 58 56 5 - 0 23 7 2 723 -730 11 -12 248 236 4 -4	5 36 -37 11 -11 204 -211 4 -7 368 374 6 234 -229 4 -9 43 27 5 -5 53 68 4	16 46 53 8 8 295 294 5 13 80 -153 5 19 56 -56 6 18 29 32 13 104 25 0 7 -104 10 -4 7 21 81 -71 2 38 20 14 7 7 14 15 6 7 15 15 15 15 15 15 15 15 15 15 15 15 15
-4,1195-1135 18 -44,212 -212 4,13 257 259 4, 154 -2 756 -744 11 -24 23 -18 8 15 598 -103 4, 17 5 0 1847-1834 28 0 145 148 6 17 36 47 11 лял H 2 522 -559 8 2 38 35 10 1954 0 0.7 -21 6	0 -4 7 4 191 192 3 -10 244 225 4 - 1 51 8 6 526 508 8 -8 571 584 9 1 1 14 same 8 77 -63 3 -6 446 448 7 1 1 47 same 8 77 -63 1 -6 446 448 7 1	2 98 -104 5 -7 265 277 4 -3 335 325 5	22 54 -65 7 14 49 -55 9 -6 129 123 4 -23 38 -36 10 24 58 59 7 15 177 -174 4 -4 27 36 13 -21 67 -67 6 maar H 4 2 maar 18 65 59 6 -2 75 81 5 -19 30 -26 11
- Ψ Ψ03 Ψ06 6 ΑΛΒ Η ⁻¹ Ο ΛΑΔΒ 21Ψ 27 -32 7 -19 5 6 818 825 12 1 534 518 8 234 15 38 8 -17 5 8 4Ψ9 Ψ77 7 3 316 -307 5 ΑΛΒ Η ₋₁ 7 ΑΛΒ -17 5 8 4Ψ9 Ψ77 7 3 316 -307 5 ΑΛΒ Η ₋₁ 7 ΑΛΒ -15 15	8 103 4 12 186 -181 3 -2 745 -745 11 0 -60 7 14 145 -151 3 0 231 -231 4 1 160 4 16 30 39 11 2 492 -492 8 1	5 30 -4 12 1 337 -324 5 5 244 -254 4 9 130 123 4 3 19 -32 6 7 129 -135 4 9 98 103 5 5 171 -155 3 9 39 19 9	-22 47 -20 8 A A H 4 9 A A 2 126 -123 4 -15 81 77 5 -20 73 -60 6 -24 78 72 5 4 90 86 4 -13 87 92 4 -164 25 -3 7 -22 29 -18 13 6 62 -67 7 114 15 -39 6
12 52 51 4 7 330 - 5 50 - 13 1	U-14/4 10 51 -33 / 4 230 306 5 ми	* H 2 18 AAAA 7 305 347 5 114 0 7 8	-15 1866 187 4 -20 105 112 4 8 85 -90 5 -9 75 -59 4
12 52 51 4 7 330 - 356 6 -23 115 1 - 11 1	4 -176 4 20 88 94 5 64 0 -14 5 -11	5 63 51 6 8 155 +177 3 13 67 70 5	-14 173 150 4 -18 110 102 4 ламе 1 4 18 лам -7 104 101 3
14 23 30 6 9 517 509 8 -21 132 132 4 -9 14	3 -137 4 224 23 25 7 8 226 226 4 -1	4 36 -18 10 11 62 94 4 154 24 33 8	-12 295 307 5 -16 40 45 9 -144 0 23 9 -5 418 411 6
16 283 -285 5 11 229 -419 7 -19 61 -46 6 - 7 5	4 -61 6 24 37 -39 11 10 263 265 5 -11	6 133 -133 4 13 40 40 8 17 94 86 5	-10 147 -153 3 -14 71 -77 5 -12 80 -87 6 -3 194 -195 3
10 86 -81 5 13 43 16 6 -17 183 -184 4 -5 1 20 183 -190 4 15 27 -7 11 -15 242 -235 4 -3 5 22 51 28 7 174 13 12 6 -13 407 404 6 -1 15 22 51 28 7 174 13 12 6 -13 407 404 6 -1 15	13 143 3 AAN H 2 3 AAN 124 0 11 7 -11 77 57 6 -24 85 -82 5 14 55 53 7 -1 13 117 4 -22 28 5 11 16 35 -40 10 -1	9 34817 9 15 5119 7 жла н 3 13 жла 9 133135 4 17 35 37 1121 40 20 9 5 157 157 4 19 5961 619 3729 10	-86 162 -151 3 -12 74 -63 4 -104 13 5 7 -1 197 -181 4 -6 280 -290 5 -10 266 -263 4 -8 117 -122 4 1 326 315 5 -4 590 -603 9 - 0 289 -262 4 -6 389 29 9 2 270 227 4
A A A A A A A A A A A A A A A A A A A	10 -123 4 -10 107 -100 4 10 106 -101 4 -	2 81 82 6 23 29 17 14 -15 86 -83 5	-2 249 -245 4 -5 133 -134 3 -44 5 -6 9 5 3483 376 5
	5 -336 5 -16 122 -117 4 204 23 4 8	2 81 82 6 23 29 17 14 -15 9 -24 8	0 86 84 2 -4 401 406 5 -2 124 128 4 7 100 -112 4
	5 -336 7 -16 46 -52 7 али н 2 10 али	3 30 311 μ 3 6 μμ -13 90 -33 5	2 140 154 3 -2 209 204 4 04 19 13 7 9 91 -92 4
	10 -63 7 -14 430 427 7 -24н 11 4 8	2 32 30 11 -25 112 108 4 -11 31 41 12	4 1057 1055 15 0 417 415 6 244 20 34 8 11 177 -175 4
-72 122 -126 4 AAAB H 1 1 AAAB -3 170 173 3 9 2 -20 132 126 4 -25 73 79 6 -1 101 -99 3 11 3 -16 122 124 5 -234 0 20 7 1 58 -65 3 134 -16 672 605 9 -214 27 -15 9 1 100 -189 3 154	89 -43 9 -12 738 740 11 -22 44 40 8 11 16 10 -10 339 523 5 -20 42 30 8 0 -4 7 -8 189 -186 3 -18 106 97 4 0 13 8 -6 183 -186 3 -18 106 97 4	4 101 -100 4 -23 85 90 5 -9 29 37 11 5 91 89 5 -214 0 -25 7 -7 193 199 4 9 67 -69 6 -19 179 -161 4 -5 119 -130 4	6 31 39 4 2 266 - 269 4 4 36 - 26 12 13 59 - 66 7 8 29 21 8 4 147 154 3 64 0 3 8 15 42 22 9 10 147 - 149 3 6 247 - 243 4 mar H 1 19 mar 174 0 25 7
-14 63 -49 4 -19 131 -136 4 5 135 -127 3 *** H	1 15 mm -4 775 -763 12 -14 55 41 6 -1	44 0 -30 8 -15 273 -272 5 -1 115 -119 4	14 35 -13 8 104 0 12 7 -15 35 35 11 21 58 56 7
-12 442 458 7 -17 287 -282 5 7 254 -244 4 -21 6	2 -63 7 -2 1330-1333 20 -12 65 -65 5 -1	2 72 63 5 -134 17 26 5 1 102 101 4	15 183 -178 4 124 0 -4 7 -8 132 134 4 246 15 7 16
-10 327 -216 5 -154 17 -17 6 9 224 -233 4 -21 19 15	7 -161 4 0 104 -107 2 -10 52 -68 5 -1	2 106 -100 4 -11 150 145 3 3 61 -59 7	18 63 -59 7 14 62 -62 7 -6 107 106 4 -23 129 129 1
-6 352 -546 5 -13 31 -41 8 11 139 148 4 -17 4	78 -78 5 2 1356 -1494 33 -8 886 -694 4 -	5 145 -142 4 -94 13 06 5 83 -79 5	20 1068 110 y 16 y5 y7 8 y 118 122 y 21 79 77 5
-6 529 -524 13 -11 137 196 3 13 342 345 6 -15 4	10 -31 9 4 720 713 11 -6 1922 -197 4 -	5 89 -90 5 -7 719 730 11 7 31 -72 12	22 72 70 5 184 15 6 8 24 20 8 8 19 89 87 y
-4 1105-1096 17 -9 455 450 7 154 25 27 8 -13 14	14 -137 4 6 715 744 11 -4 177 -173 3 -4	4 52 -55 7 -5 176 188 3 9 57 55 7	Anar H y 3 Anar and H y 10 Anar 0 115 110 y 17 55 y9 7
-2 200 -150 3 -7 153 -134 3 12 12 -11 8	88 94 5 8 400 411 6 -2 192 192 4	2 154 156 4 -3 469 465 6 11 48 41 7	21 y 22 y 8 22 y7 26 c 5 2 97 112 15 127 34 c y
0 835 835 13 -5 190 194, 3 19 76 -81 5 -9 13 2 72 -35 2 -3 675 658 10 214 0 12 8 -7 28 4 1999 1983 23 -1 287 232 4 лин H 1 8 лин -5 12 6 956 972 4 1 657 252 4 лин - 5 16 - 3 1	19 193 4 10 227 204 4 0 100 97 3 A 263 5 12 218 -213 4 2 426 429 7 13 93 4 14 281 -217 5 4 254 -263 4	0 70 58 5 -1 967 -973 6 19w 20 27 7 2 62 54 7 1 251 -277 4 15 55 -51 7 44 0 5 8 3 366 -381 6 лял H 3 14 лял	22 59 72 5 -20 30 38 10 mm H 420 mm -13 216 211 4 -20 56 -51 8 -18 155 155 3 -84 19 -5 8 -11 85 85 4 -18 237 224 9 -18 155 129 9 -6 78 73 5 -9 5
8 239 42 5 3 1382 -1390 21 -23 5 43 5 -3 6 10 586 -587 9 5 475 -494 7 -21 52 52 6 1 1 1 12 516 -519 8 7 429 401 7 -19 120 118 4 3 23	12 -15 8 18 222 -221 4 8 223 -225 4 mm 12 -15 8 18 222 -221 4 8 223 -225 4 mm 14 -184 4 20 33 29 10 104 7 -14 7 -1 19 -221 4 22 69 73 6 124 18 -22 7 -1	ын 0 6 8 5 339 -543 5 5 -21 112 -118 4 в H 2 20 аллл 7 3668 -3397 6 -1944 0 -3 7 344 21 29 8 9 554 559 8 -17 185 -183 4 24 26 31 8 11 159 154 3 -15 72 -72 5	-16 90 -94 5 -14 103 90 4 -4 35 29 13 -7 226 -219 4 -14 262 -268 4 -12 107 -97 4 -2 46 26 9 -5 45-45 49 -12 250 -240 4 -10 197 -202 4 лар н 5 0 лар -3 121 -117 3 -10 197 -207 4 -84 13 -114 6 1 606 610 9 -1 302 -295 5
14 111 105 4 9 355 355 6 -17 123 -127 4 364 1	12 -5 8 24 105 109 4 14 53 52 8 -	8 4/5 -53 9 13 226 226 4 -134 0 23 8	-8 231 237 4 -6 225 -241 4 3 83 -85 3 1 360 349 6
16 36 -28 10 11 36 37 5 -15 215 -216 4 7 1	17 83 5 али н 2 4 али 15 38 -38 9 -	5 129 130 4 15 112 -107 4 -11 269 264 5	-6 225 224 4 -4 287 -233 5 5 37 -27 1 3 1 79 189 3
18 56 49 7 13 316 325 5 -13 87 106 4 9 11	19 116 4 -24 35 -26 10 184 30 39 9 -	14 24 12 8 174 0 -4 8 -9 316 316 5	-4 343 346 5 -2 264 267 4 7 353 -349 6 5 107 97 3
200 23 15 7 15 40 13 7 -11 419 -433 6 114 1	19 1-6 4 -24 35 -26 10 184 30 39 9 -	14 20 12 8 174 0 -4 8 -9 316 316 5	-3 316 318 4 7 10 7 9 10 3
22 81 69 5 17 58 -60 6 -9 181 189 3 1314	Ч. –1.8. –204 23. –3.7. –24.104. –388 4	3 44 –26 9 214 8 –25 8 –5 47 –46 7	0 4 10 12 2 2 2 2 303 306 5 11 47 41 6 9 151 157 4
man ri 0 8 man 19 70 -74 5 -7 121 110 3 man ri	1.16 мми –18. 53. –46.6. –22. 39. –41.10	2 83 –75 5 яля н 3 7 яля –3 70 –77 6	2 467 467 461 7 4 31 8 10 134 21 23 6 11 189 189 4
-244 16 28 10 21 71 -74 5 -5 131 122 3 -194	0.16.8. –16.211.206.4. –20. 90. –96.4. мм	н 3 0 яля –25 47 49 9 –1 322 –320 5	4 251 235 4 6 32 22 8 15 55 34 13 13 40 389 4
-26 239 239 1 25 13 - 25 10 -1 21 -216 1 -154 1 -18 125 115 1 225 H 2 230 1 1 25 -152 3 -13 -16 31 32 11 -25 51 40 6 34 9 24 4 -11	2 12 7 -12 13 13 -13 3 -16 10 10 10 10 10 10 10 10 10 10 10 10 10	1 1053-1053 10 -23 43 -34 8 1 151 -136 4 3 378 -402 6 -214 0 -2 7 3 39 -16 11 5 638 818 13 -19 121 -111 4 5 80 71 5 7 74 66 3 -17 127 -125 4 7 98 90 5	В 181 - 1/6 3 8 26 23 12 17 142 140 4 15 229 - 227 5 В 227 - 227 4 10 65 - 64 6 19 140 - 116 4 17 100 110 4 10 143 - 152 3 12 212 - 204 4 21 156 151 4 154 19 - 38 8 12 71 70 4 14 47 49 7 234 5 - 98 8 лия 4 5 8 лия
-14, 349 -351 6 -239 13 -21 7 5 206 -207 4 -9 11	73 169 4 -6 362 363 6 -10 124 -126 3	9 315 315 5 -15 169 179 4 9 30 1811	14, 1346 1340 3, 16, 67, -67, 7, жил н, 5, 1 жил -2344 15, -14, 8
-12 370 -543 6 -21 53 53 8 7 47 42 5 -74 2	77 25 7 -4 30 39 3 -6 226 -276 5 1	1 266 263 4 -134 0 -10 6 11 68 55 5	16, 156, 155, 4, 16, 109, 5, -23, 28, -33, 14, -21, 70, 72, 6
-10 738 -747 11 -19 51 -59 5 9 50 33 5 -5 22	25 206 4 -2 504 433 8 -6 136 -121 3 1	3 162 149 3 -11 170 171 3 134 0 -11 8	18, 352, -13, 12, жил н, 4, 11, жил -21, 33, 33, 11, -19, 42, 9, 8
-8 277 -277 5 -17 35 54 9 11 944 -272 4 -3 5	19 -26 9 0 223 216 4 -5 136 -121 3 1	5 80 -94 5 -9 50 -15 4 134 0 -11 8	24, 74, 77, 5, -22, 74, 74, 6, -10, 87, 66, 5, -17, -14, 42, 9, 8
6 447 460 7 -15 419 -421 7 13 117 125 4 -1 1	77 - 106 4 2 145 - 147 5 - 2 450 - 458 7 1	7 67 -64 5 -7W 0 19 4 -19 73 -66 5	222 1039 - 112 4 - 20 111 - 100 4 - 17 176 173 4 - 154 5 19 - 20 7
-4 935 919 14 -13 247 249 4 15 77 74 6 1	13 32 10 4 288 - 298 5 0 172 169 5 1	9 185 -190 4 -5 66 78 3 -17W 10 -6 7	### # 4 4 ### - 18 54 - 64 8 - 15 164 164 4 - 13 204 - 205 4
-2 248 258 4 -1 12 248 -204 4 174 23 -23 7 3	50 - 48 9 6 90 - 98 3 2 372 967 6 2	W 0 6 7 -3 58 53 3 -15 57 60 6	- 24 39 22 10 - 16 189 - 167 4 - 134 0 3 6 - 11 157 - 153 5
0 - 139 - 133 11 - 9 432 407 / 19 79 72 - 3 5 5	87 - 102 4 8 53 - 41 4 4 533 373 6 2	3 118 −111 4 −1 416 −423 7 −13 85 −91 4	22 92 86 94 -19 95 -94 4 -11 89 -78 4 -9 363-367 6
2 329 - 316 5 - 7 65 69 2 21 52 - 67 - 5 74 1	10 13 7 10 78 79 3 .6.194 197 4 2	5 46 50 8 1 21 24 6 −11 187 184 4	-20 96 95 9-12 53 -59 7 -9 564 -579 6 -7 259 262 4
4 104 - 122 3 - 5 169 - 159 3 лал H 1 9 лал 54 1	16 - 33 8 12 185 - 187 4 8 136 - 202 4 84	6 H 3 1 xaan 3 49 −57 5 −95H 0 16 8	-18 64 96 -65 10 170 176 4 -7 268 -278 5 -5 88 89 4
6 245 - 237 4 - 3 258 - 320 5 - 254 23 26 8 11 1	8 141 4 14 83 88 4 10 84 85 5 - 2	5H 0 −2 8 5 158 129 3 −7 28 40 14	-16 167 164 4 -8 192 192 3 -5 119 -116 3 -3 9 179 3
6 430 - 443 7 -1 377 394 6 -23 72 -72 5 min in 10 145 -146 3 1 277 -289 4 -21 73 -70 5 -19 (12 151 - 149 4 3 323 -325 -19 173 -174 4 -19 (12 151 - 149 4 3 323 -327 5 -19 173 -174 4 -19 (12 151 - 149 4 3 323 -327 5 -19 173 -174 4 -19 (12 151 - 149 4 3 5 -19 (12 151 - 149 4 5 -19 (12 151 - 149 5 -19 (13 151 - 149 5 -19 (13 151 - 149 5 -19 (15 151 -	1 17 maar 164 0 -16 7 12 163 -162 4 -2 i9 -53 7 18 111 113 4 14 162 -163 4 -2 18 24 8 204 0 -14 7 16 52 42 7 -1 19 192 4 8 204 0 -14 7 16 52 42 7 -1	3 99 -100 4 7 160 172 3 -5 65 -56 6 1 45 36 7 94 21 -32 6 -3 180 -177 4 9 67 69 6 11 71 -68 5 -1 33 36 12	-14 100 -94 4 -6 110 97 4 -3 234 20 4 -1 152 153 3 -12 174 -153 4 -4 129 122 3 -1 128 165 3 1 138 142 3 -19 490 -455 4 -2 91 72 4 1 420 178 7 3 5 -46 5
16 277 277 5 7 152 -159 3 -15 176 185 4 -13 15 18 53 53 7 9 177 172 3 -13 121 116 3 -11 20 41 -42 10 11 43 41 5 -11 152 132 3 -9	1 154 3 24 60 -60 7 лая H 2 12 лан -1 15 83 4 лая H 2 5 лая -22 35 13 11 -1 17 26 8 -24 75 -81 5 -20 140 145 4 -1	5 165 162 4 15 216 -215 4 34 16 -3 9 3 62 65 4 17 126 126 4 5 58 44 8 70 86 3 19 62 126 4 5 58 44 8	-0 210 -202 4 0 30 9/ 3 3 30/ 36/ 9 5 50 42 5 -6 180 175 3 2 303 -310 5 5 225 220 4 7 151 -1\8 4 -4 100 111 3 4 274 -270 5 7 346 -313 5 9 42 -50 8 -2 116 129 3 6 80 91 4 9 27 -2 9 11 56 -51 7
anan H 0 10 anan 13 74 -81 4 -9 535 536 8 -7 13	-152 4 -22 110 111 4 -18 84 -87 5 -	9 14/5 14/3 3 21 4/2 35 9 9 69 76 6	0 667 674 10 8 399 -86 4 11 266 -285 5 13 34 -23 11
-24 442 32 9 15 225 230 4 -7 130 121 3 -5 -	18 -76 6 -20 88 88 5 -16 50 -40 8 -	7 4/73 4/68 7 лаан H 3 8 лал 11 4/3 -31 9	2 179 187 3 10 178 179 4 13 119 -12 1 3 15 6 1 56 5
-22 54 57 7 17 49 -29 7 -5 43 51 5 -3 10	86 -183 4 -18 263 267 5 -14 176 -178 4 -4	5 4/7 -57 2 -25 19 15 8 1314 0 22 8	4 122 117 3 12 46 46 7 15 196 -194 4 17 126 127 4
-20 124 -120 4 19 32 22 12 -3 630 -635 10 -14	0 -14 9 -16 54 48 5 -12 86 -122 4 -4	3 135 -136 9 -72 8 1 -77 5 лаан H 3 16 лаа	6 390 -940 6 104 0 40 7 15 36 6 6 100 10 - 12 4
-16 216 -213 4 21 38 -29 8 -1 604 -594 9 1	Ni 8Ni 5 -10, 188 19Ni V -10Ni 0 -2Ni 6 -	1 174 - 192 3 -21 186 - 188 4 -19 103 99 4	8 442 - 445 7 164 0 19 8 19 136 140 4 5 9 8 8
-16 323 -315 5 23 84 -80 5 1 55 66 4 34	Ni -23 7 -12, 112, 129 3 -8, 122 131 3	351 350 5 -19 156 -157 4 -17 116 114 4	10 27 - 9 10 8 4 4 12 888 214 28 49 8 - 23 42 - 55 10
-14 141 -137 4 25 77 75 6 3 22 25 9	17, 14 13, -10, 532 -537 8 -6, 139, 128 3	3325 334 5 -17 104 105 4 -15 184 187 4	12 115 - 112 4 - 22 39 - 25 10 23 36 7 1121 123 121 4
-12 81 101 4 and H 1 3 and 5 144 142 3 7 8	SJ 89 5 -8 /9/-/SS 12 -4 /5 // 4	5 257 244 4 -15 54 55 5 -1394 23 20 7	14, 157 155 4, −20 77 73 6 anar H 5 2 mar −19 189 196 4,
-10 241 248 5 -25 52 -48 6 7 202 135 4 8 8	56 78 5 -6 742 -732 11 -24 18 -8 6	7 42 -35 3 -13 365 360 6 -11 76 75 5	164 16 734 8 −18 43 42 8 −234 0 −10 8 −17 89 83 5
-8 248 229 4 -23 52 -40 6 94 23 28 6 114 1	16 -8 8 -4 15 27 5 0 48 -45 6 1	8 114 -110 3 -11 398 395 6 -9 51 -43 8	18 76 75 6 −15 97 −86 5 −21 217 222 4, −15 60 −57 6
-6 524 520 9 -21 55 -49 7 11 55 48 6 and H	18 mm -2 81 806 12 2 177 181 4 1	1 400 -416 5 -9 218 213 4 -7 203 -203 4	20 57 37 7 −14 133 −172 4, −19 73 −37 6 −13 127 −128 4
	Avi -51 12 0 122 1348 3 4 219 -233 vi 1 10 85 6 2 1316 1315 20 64 22 -12 7 1 12 31 8 4 365 356 6 8 109 -104 4 1 1 125 8 8 101 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1	3 247 242 4 -7 98 -103 3 -5 186 -186 4 5 55 -34 5 -5 645 -538 10 -3 71 -87 7 7 82 79 5 -3 361 -358 6 -1 77 73 6 82 79 5 -3 361 -358 6 -1 77 73 6	224 22 26 8 -12 129 -125 4 -17 78 -68 5 -11 286 -281 5 ### H 4 5 ### -10 110 -115 4 -15 91 -99 4 -9 52 -10 6 -24 50 -11 13 -6 53 98 4 -13 251 -246 4 -7 444 -449 7
4 546 -551 8 -11 122 121 3 214 27 -32 8 -9	17 9 9 8 590 -577 9 12 59 52 5 2	91 - 67 4 1 53 - 55 4 3 73 56 6	-20 243 -244 4 -0 103 105 4 -10 0 -12 5 -3 100 297 5
6 417 442 7 -9 108 91 3 ama H 1 10 ama -7	12 83 4 10 435 -447 7 14 38 40 9 2	41 52 10 3 200 132 4 5 158 159 4	-18 59 42 6 -2 133 194 4 -7 67 71 4 -1 259 278 4
8 125 -135 3 -7 650 -652 10 -254 0 -1 8 -5 18	15 -186 4 124 16 -22 6 164 19 11 8 мм	H 3 2 mm 5 188 187 3 7 104 110 4	-16 115 -118 4 0 101 102 4 -5 622 602 9 11 216 213 4
10 285 252 5 -5 477 475 7 -21 54 65 7 -34 1	73 3 7 14 29 -276 11 ### H 2 13 ### -2	5 31 513 7 95 97 3 9 106-104 4	-14, 46, 48, 7, 24, 12, -16, 7, -3, 127, -119, 3,3, 25, -25, 12,
12 185 185 4 -3 236 -232 4 -21 181 -183 4 -1 18	15 -118 4 164 18 1 7 -22 80 75 5 -2	3 125-126 4 9 202 202 4 11 47 -37 6	-12, 35, 34, 6, 4, 232, -240, 4, -1, 110, 108, 3, 5, 25, -41, 13,
14 42 -19 9 -1 197 -171 3 -19 51 37 8 14	0 20 8 18 95 104 5 -20 62 62 6 -2	14 0 -5 6 11 95 96 4 лая H 3 17 лая	-104, 19, -26, 5, 6, 78, -78, 5, 1, 62, 62, 3, 7, 106, -108, 4,
164 0 -16 7 1 258 270 4 -17 203 -196 4 3 f	33 59 6 20 62 69 6 -18 150 151 3 -1	9 133-129 4 134 0 -5 8 -17 46 35 9	-8, 478, 481, 7, 8, 78, -5, 3, 261, -271, 4, 6, 451, -165,
16 34 33 11 3 231 31 5 -15 121 -131 4 54 1	3 0 7 22 81 72 5 -16 177 169 4 -1	7 302 303 5 15 179 -177 4 +15 51 52 8	-6 3300 322 5 10 99 96 5 5 5 223 213 1 1 61 68 7
201 121 -119 6 5 76 -67 2 -13 336 350 5 74 1	15 5 8 алл H 2 6 алл -14 257 -255 5 -1	5 339 337 5 17 123 -128 4 -13 45 -53 8	-4 286 296 5 12 70 71 6 7 414 400 6 134 0 7 8
444 10 12 44 7 375 371 5 -14 12 -15 5 44 1	1 19 алл -24 69 71 6 -12 259 -257 5 -1	3 270 269 4 194 15 27 7 -114 0 -17 7	-2 97 93 9 14 42 42 9 9 52 51 5 154 20 -12 7
-22 182 -185 4 114 0 27 4 -9 350 54 3 -154	7 9 8 -22 39 1 8 -10 275 -277 5 -1	1 101105 3 21H 0 13 89 86 86 4	0 5615-5634,10 15 440 45 9 11 215 209 4, 17 858 82 5
-22 182 -185 4 114 0 27 4 -7 298 311 5 -134	0 -4 8 -20 39 27 10 -8 153 -150 4 -	9 173 -172 3 888 H 3 9 888 -7 143 -149 3	2 72 -770 3 mar H 4,13 mar 13 275 258 5 mar H 5 10 mm
-20 73 -62 5 13 139 -132 3 -5 313 -334 5 -11	72 -69 5 -18 230 -233 4 -6 127 126 3 -	7 686 -691 10 -23 100 -101 4 -5 64 49 5	4 272 -270 4 -224 18 1 9 154 0 -7 7 -234 0 -14 8
-16 207 -139 4 15 139 -126 3 -3 446 477 7 -9 9	18 -102 4 -16 60 50 5 -4 178 179 4 -	5 101 -108 2 -21H 0 10 83 107 -105 5	6 290 293 5 -20 54 -52 5 17 350 25 13 -21 92 14 12
-16 303 309 5 17 173 -177 4 -1 584 -506 9 -7 10	i9 -189 4 -14 292 -291 5 -2 290 282 5 -	3 424 - 424 7 - 194 10 - 10 8 - 1 69 57 5	8 315 316 5 -18 127 -129 4 19 114 -117 4 -194 23 -22 7
-14 239 254 5 19 47 -49 9 1 216 -205 4 -5	19 -47 10 -12 362 381 6 0 147 151 3 -	1 81 75 2 - 17 73 - 84 6 1 71 75 5	10 55 36 5 -16 59 52 6 214 0 -30 8 -15 57 60 5
-12 332 340 5 214 11 14 7 3 167 145 3 -5 1	12 164 4 -10 151 -166 3 2 115 114 4	1 143 - 121 3 - 15 297 301 5 3 32 - 9 14	124 23 -18 6 -19 50 52 8 234 8 4 -154 25 15 8
-8 164 -161 3 454 84 17 18 3 166 5 1 1	10 4 13 -4 261 256 4 8 21 -278 6	5 811 816 12 -11 162 -167 3 74 23 -8 8	in
-6 343 -353 6 -25 48 -47 8 9 26 22 12 3	10 4 13 -4 261 256 4 8 219 -224 4	7 373 376 5 -9 157 171 3 94 0-25 8	
-9 198 -176 9 -23 70 -71 5 11 9 5 7 5	10 58 7 -2 49 -52 3 10 47 -40 8	113 -103 3 -7 518 -528 8 888 1 3 18 888	
	1 20 400 0 202 18 4 4 12 36 17 9 1	1 284 +265 5 -5 247 263 4 -15 64 57 6	2224 9 -2 8 -4 325 -321 5 -17 101 88 5 -5 252 256 5
	16 -53 11 2 532 -522 8 14 4 0 - 20 7 1	3 207 -207 4 -3 59 -61 4 -13 183 -183 4	-24 81 76 -25 -25 124 -125 4 -15 256 -256 5 -34 15 25 5
	15 18 8 4 139 110 3 16 52 40 8 1	5 180 -190 4 -1 125 -124 3 -11 92 -87 4	-24 81 76 6 04 16 7 -15 256 -256 5 -34 15 25 -35 3
	15 18 8 4 139 110 3 16 52 10 1 - 2	113 -112 4 1 248 256 - 266 - 266 - 266	-24 81 76 6 04 16 7 -15 256 25 4 -1 125 -143 3
6 487 506 7 -13 86 83 3 487 11 887 -7 6	3 50 6 8 200 202 4 -22 117 -109 4 1	9 61 59 7 3 43 47 9 -74 0 34 7	-204 10 -10 7 44 0 -20 8 -9 208 261 4 3 284 28 28 -9 11 4 3 284 28 28 3
8 75 76 5 -11 433 492 8 -23 112 -116 4 -5 1	9 -117 4 10 213 213 4 -20 67 53 6 2	1 130 139 4 54 0 1 5 -5 101 96 4	-18 136 -132 4 6 88 85 5 -7 52 -50 4 5 52 47 7
10 72 -74 5 -9 288 5 -23 51 2 -26 6 -3 5	19 -49 10 12 40 44 8 -18 127 -121 4 2	34 6 18 8 7 31 35 9 -3 99 100 4	-16 12 -23 7 8 35 2911 -5 502 495 8 74 0 9 7
лет зо чілі -/∞чыс ччя 7 -19 134 130 ч -1 1	י	л з заяж у ус. – 533 5 – 1 118 117 ч.	-14 c347 4 10 94 104 4 -3 271 271 5 9 123 118 4
14 115 -118 ч -5 1055-1054 16 -17 159 161 ч 1		5 ч.ч. – 18 9 11 348 27 9 1 316 39 10	-124 0 -9 5 12 54 -43 7 -1 397 398 5 11 65 63 7
2 16 149 -150 ч -3 376 377 6 -15 270 272 5 3 6		3 52 – 19 7 13 50 39 7 3 85 – 82 5	-10 148 -159 3 144 22 -10 8 1 46 -39 3 134 0 -18 7
18 ч8 ч3 9 -1 52 -55 2 -13 173 165 ч ша м		1 37 18 8 15 107 – 110 ч. 5ч 0 я я	-8 515 521 8 -36 H 1 48 and 3 27 -31 9 15 5
Amar H 0 14 Amar 1 183 - 152 3 - 17 265 - 278 4 - 7 10 - 274 27 - 15 6 3 178 155 3 - 9 113 - 105 3 - 5 4 - 270 180 183 4 5 267 266 3 - 9 113 - 105 3 - 5 4 - 20 180 183 4 5 267 266 4 - 7 273 - 264 4 - 3 11 - 10 187 - 10 19 4 5 267 266 4 - 7 273 - 264 4 - 3 11 - 10 187 - 10 19 4 5 267 266 4 - 7 273 - 264 4 - 3 11 - 10 187 - 10 19 4 5 267 266 4 - 7 273 - 264 4 - 3 11 - 10 187 - 10 19 4 5 267 266 4 - 7 273 - 264 4 - 3 11 - 10 187 - 10 19 4 5 267 266 4 - 7 273 - 264 5 - 7 273 - 264 4 - 3 11 - 10 187 - 10 19 4 5 267 266 4 - 10 19 19 19 19 19 19 19 19 19 19 19 19 19	NU SI 9 22W 0 8 8 -8 17 162 8 -1 17 -38 9 AAA H 2 7 AAA -6 17 162 8 -1 11 128 1 -21 115 117 1 -1W 18 33 7 -1 2 0 AAA -22 110 110 11 -3 32 -3 -3 -	9 115 120 4 17 40 44 10 74 22 -7 8 7 31 33 11 19 36 28 11 явя H 3 19 явя 5 73 -79 5 явя H 3 10 явя -134 24 40 8 182 183 3 -25 18 18 8 8 8 8 8 8 8 8 8 8 8 8 8 8 8 8	-6 299 298 5 -204 0 -5 8 5 521 -572 8 17 7 58 6 -4 459 454 7 -18 45 -55 8 7 150 -137 3 ### h 51 ### -2 916 -313 5 -16 80 -50 5 7 150 -137 3 ### h 51 ###
-16 269 277 5 9 246 285 5 -3 155 -157 3 0 73 -14 176 170 4 11 226 -234 4 -1 256 -266 5 2 165	11 709 11 -20 92 86 5 0 71 -71 5 -1 10-1690 25 -184 25 -36 7 2 47 -43 9 -	i 491 -489 8 -21 33 2010 -9 57 49 7 9 275 268 5 -19 191 195 4 -74 16 18 8	v 107 - 130 3 - 138 (1 - 103 - 11 - 104 105 4 - 119 65 - 566 4 2 126 - 130 3 - 12 - 29 100 5 13 110 107 4 - 17 155 - 159 4 4 292 - 284 5 -10 191 194 4 15 92 85 4 -15 174 -176 4

Table 5 (cont.)

H 085 CAL 3G H 085 CAL 9C H 083 CAL 9G	H 685 CALSG H 685 CALSG H 68	S CAL SG H 683 CAL SG H 685	CAL 5G H 685 CAL 5G H 685 CAL	56 H 085 CAL 56 H 085 CAL 56 H 085 CAL 56
-13 139 -138 4 6 305 -293 5 4 70 63 4 -13 139 -138 4 6 305 -293 5 4 70 63 4 -11 49 -224 9 6 117 112 4 6 195 188 4	-6 64 51 7 -7 76 -70 4 134 (-4 55 25 6 -5 141 -146 3 154 20 -24 0 -2 8 -3 550 -539 8 888 4	0 -6 7 -6 34 3 9 -18 63 0 22 8 -4 75 68 4 -164 14 7 10 aaa -2 36 -38 8 -144 23	-66 7 17 42 -38 10 -11 182 187 -22 8 ### H 9 1 ### -9 65 58	4 6M 24 31 8 6H 0 -19 8 -3 45 37 8 6 8H 0 -15 7 mm / 10 12 mm -14 0 2 8
-74 0 56 10 333 -343 5 8 41 -35 8 -74 0 -5 6 12 73 -61 6 10 224 -228 4 -5 724 276 5 14 27 26 13 12 117 -107 4	0 69 -92 5 -1 25 7 10 -19 7 2 118 -119 5 1 97 -90 3 -17 14 4 53 71 7 3 254 254 5 -15 33	4 -75 6 0 130 -132 3 -12 190 9 -150 4 2 130 -134 4 -10 38 2 -25 10 4 117 -112 4 -8 57	191 4 -17 42 32 9 -5 124 -121 44 11 -15 156 -159 4 -34 28 -7 -44 11 -15 156 -159 4 -34 28 -7	V 12 62 65 7 84 0 3 8 1 44 45 9 8 144 0 -13 8 -6 70 73 6 54 0 2 8 14 12 62 65 7 64 0 3 8 3 44 45 9 8 144 0 -13 8 -6 70 73 6 54 0 2 8 14 16 0 -13 8 -6 70 73 6 54 0 2 8
-3 130 134 4 16 47 39 8 14 47 22 8 -1M 0 10 6 18 59 58 7 16M 19 -11 7 1 124 -119 4 20 45 36 9 18M 26 22 8	6 113 116 4 5 54 58 6 13 5 BM 20 -30 7 7 278 282 5 11 24 IOM 23 37 8 9 151 142 4	9 -48 7 6 56 -47 6 -6 99 - 5 250 5 8 176 165 4 -4 67	101 4 -11 69 53 6 1W 14 -21 -54 6 -94 0 28 9 3 37 -33	8 mm H 10 3 mm -2 43 42 9 mm H 10 10 mm 11 −154 <u>8</u> -22 8 04 0 -20 8 -94 20 -6 8
3 352 -363 6 22н 27 38 8 20н 0 8 8 5 60 -68 7 яяя н 6 1 яяя зяя н 6 7 яяа 7 96 86 ч -22 0 0 -3 8 -22 30 _20 1	12 78 -85 6 114 17 24 7 -7 100 ana H 6 14 ana 13 132 -134 4 -5 111	0 91 4 12 159 158 4 0 75 1 113 4 14 49 58 9 2 28	-73 5 -54 0 -26 7 7 76 67 -31 13 -34 10 0 7 94 0 27	Ч -14, 70 -55 5 24 0 -31 9 -7 50 38 8 5 -12 51 -46 7 4 57 -49 7 -54 0 -18 8 8 -104 0 -5 7 жля H1013 ляя -3 39 1110
9 76 -84 6 -20 47 -57 9 -20 35 37 12 11 116 110 4 -184 11 -12 6 -18 47 50 7	-164 27 32 8 17 39 40 9 -1 12 -144 39 26 9 194 10 8 8 1 6	D -36 9 16 47 44 8 4 40 D -118 4 18 64 -51 6 6 50 Z -90 5 20 36 -22 11 84 25	349-13361011420-18 4481211-20441334026 -431034343418==+99•	8 -8 37 -43 12 -8 45 39 9 -1 49 -27 8 8 -64 0 17 9 -6 44 -12 9 14 14 20 8 8 -64 137 138 4 -44 13 -11 8 1 13 21 8
15 00 87 5 -14 63 66 6 -14 91 -87 5 174 23 -7 9 -12 43 53 7 -12 66 57 5	-10 213 -219 4 -21 41 -31 10 5 8 -20 25 -46 9 -19 66 -20 5 7 11	1 -117 4 ### H 8 2 ### 10H 19 N 80 5 -20 45 21 9 12H 0 1 -104 4 -18H 8 12 7 14 57	33 7 5H 25 -6 7 -17H 0 28 -8 8 7 29 0 13 -15 35 -8 54 7 5H 0 -14 8 -13 124 130	9 -2 120 119 4 -2 140 -143 4 5 31 15 13 12 0 42 -37 9 0 65 73 7 mm H 11 11 mm 4 2 55 -74 5 24 25 1 8 -7 65
-21 117 -113 4 -84 0 2 5 -8 135 141 4 -194 0 -9 7 -6 173 -175 3 -6 227 -239 4	-6 43 -43 10 -174 0 -7 7 9 54 -44 23 25 8 -15 58 15 8 11 51 -2 198 195 4 -13 88 90 5 13 86	4 57 6 -16 93 91 4 16 62 1 45 7 -144 0 -3 8 pas H 8 5 91 5 -124 27 -11 7 -18 123 -	-55 7 11W 9 -27 9 -11 46 -34 9 and 13 57 54 6 -9W 6 -3 115 4 15 105 98 4 -74 0 8	7 44 0 11 8 ### H 11 0 ### -5 41 34 10 10 6 39 -42 11 14 0 -15 8 -3 37 15 11 8 8 9 -42 11 14 0 -15 8 -3 37 15 11
-17 78 -79 5 -4 56 -53 4 -4 177 -128 3 -15 154 159 3 -2 28 30 8 -2 166 -167 3 -13 30 -8 12 0 292 239 4 0 178 190 3	0 бч 59 7 -11 2ч1 237 ч 15м 11 2 149 149 ч -9 161 161 ч лжил 4 149 -150 ч -7 180 -180 ч -194 (1 -3 8 -10 4) -18 8 -16 37 7 11 ### -8 67 70 5 -14 107 0 -13 8 -6 201 -204 4 -12 8)	31 10 17W 0 6 8 -5W 23 2W 112 W ### 1 9 2 ### -3 28 10 R2 5 -19W 0 2 # 1 70 -55	8 10 164 -154 4 54 25 -37 7 14 0 -32 8 14 12 107 102 4 7 39 -12 9 Pro 112 0 Pro
-11 144 144 4 2 50 62 5 2 53 51 6 -9 122 123 4 4 181 170 3 44 0 18 6 -7 136 -132 4 6 323 -309 5 6 55 50 6	64 6 21 8 -5 50 41 6 -17 47 8 65 -54 6 -3 169 -169 3 -15 68 10 44 -27 9 -1 107 -169 3 -13 68	7 65 8 -4 57 55 5 -10 44 68 6 -2 37 35 8 -8 167 5 87 4 0 37 39 8 -6 161 -	45 10 -17 81 79 5 1 39 -40 10 4 -15 28 43 14 3 33 -20 60 4 -13 75 77 5 5 1 33 -20	11 APA H10 4 APA 114 0 40 9 2 40 41 10 12 -164 6 -11 8 13 74 72 6 44 0 22 8
-5 107 -54 4 8 220 206 4 8 47 31 7 -3 38 33 8 10 82 -64 4 10 31 -41 12 -1 186 -184 4 124 0 15 5 124 19 36 8	nas H 6 15 nas 1 114 - 122 4 - 11 95 - 16 101 99 4 3 332 - 329 5 - 9 63 - 14 66 - 75 5 5 272 284 5 - 7 196	5 94 5 2 95 -94 4 -4 76 3 -60 6 44 8 -1 6 -2 258 - 5 -177 4 6 64 61 5 0 49	79 5 -11 73 79 6 7 36 15 862 5 -9 30 -13 12 9 43 22 7 8 -1 19 -13 12 9 43 22	9 -12 57 -55 7 -13 95 92 5 8 55 -50 7 11 -10 75 -72 6 -11# 0 -18 8 10 57 -63 8
3 193 -192 4 16 62 -64 5 16 15 -177 4 3 193 -192 4 16 62 -64 5 16 35 34 10 5 147 151 4 18 57 40 6 18 37 -410	-12 110 117 4 7 101 97 4 -54 6 -10 104 -111 4 9 203 203 4 -3 224 -8 74 -79 5 11 71 53 4 -10	0 11 7 8 204 197 4 24 0 -223 4 10 128 -119 4 4 85 -253 4 10 128 -119 4 85	2 8 -5 320 -326 5 134 12 0 78 5 -3 78 -77 5 == H 9 10 =	8 -6 49 -46 8 -7 115 -119 4 -104 0 -14 9 -4 98 98 5 -5 59 -57 6 -84 22 -34 8
7 144 138 4 20 114 110 4 AAAB H 6 8 AAA 9 44 33 7 22 46 -33 9 -22 56 46 8 114 25 6 7 AAAB H 6 2 AAAB -21 121 119 4	-6 33 27 12 13 69 -51 6 1 63 -4 73 69 5 154 0 -35 7 3 115 -2 41 -30 8 17 11 -15 1 3	67 6 14 38 -36 8 8 139 110 4 16 30 -29 12 10 58	129 4 1 101 109 5 -13 31 9 60 7 3 69 74 6 -114 23 -22	5 - 2 11 - 53 5 - 34 24 31 7 - 54 27 28 8 12 0 67 51 5 - 1 79 82 5 - 44 25 - 14 7 7 2 80 70 5 14 0 11 7 - 2 48 27 8
13 70 -71 6 -2214 0 6 8 -1614 6 -12 8 15 58 -51 7 -20 76 76 5 -16 52 -47 8 #### # 513 ### 15 86 -84 4 -14 153 -152	0 55 59 6 19 49 -54 8 7 31 2 160 -151 4 200 H 7 5 20 3 9 52 4 162 -151 4 200 H 7 5 20 1 5 2	1911 ### H 8 3 ### 14 52 - -51 7 -20 29 -2614 ### H 8	91 5 7W 13 32 8 -7 57 -57 0 xxx 9 40 32 10 -5 111 -117	4 4 0 21 8 3 136 134 3 0 37 23 10 6 6 79 - 62 5 5 30 - 25 11 2 43 - 19 8 9 9 0 - 13 7 7 0 20 7 4 22 - 1 7
-21 34 -23 13 -16 156 -154 4 -12 113 -115 4 +19 100 -39 4 -14 172 -173 4 -10 341 -342 6 -17 115 -115 4 -12	6 35 23 10 -19 100 104 4 13 117 BW 22 -9 8 -17 86 87 4 AND 104 5	7 -104 4 -164 0 14 7 -16 33 7 12 mag -14 130 125 4 -144 0	58 13 13 35 -34 10 -1 80 81 -1 7 15 90 -62 5 1 106 104	7 10 90 -92 5 9 27 -35 13 64 16 -19 8 4 12 42 49 10 11 60 -44 7 8 51 29 7 4 14H 0 -3 9 13 61 -49 7 104 13 14 8
-15 156 157 3 -10 62 -83 5 -6 159 154 3 -13 109 109 5 -8 238 243 4 -4 181 164 4 -11 277 277 5 -6 238 243 4 -4 181 164 4	-16 105 -106 5 -11 93 -103 4 -15 110 -18 105 -106 5 -11 93 -103 4 -15 110	7 -9 8 -12 112 112 4 -12 65 7 8 -10 63 -59 6 -104 0 - 107 4 -8 29 31 11 -8 58 -	59 Б 174 24 19 В 3 31 34. 27 10 даа 1 9 3 алл 5 90 92 54 7 -19 39 -30 10 7 58 -53	11 2888 H 10 5 2888 289 H 11 2 888 2888 H 12 2 288 4 -16 58 -60 7 -13 76 59 6 -10 79 -77 5 6 -14 56 72 5 -11 51 -49 8 -8 45 45 45 4
-9 41 44 9 -4 126 132 3 0 103 112 4 -7M 25 5 7 -2 75 65 3 2 134 126 3 -5 143 15 1 6 7 -2 75 65 3 2 134 126 3	-12 26 -7 13 -7 353 -154 6 -11 95 -10 34 -1 13 -5 159 -177 4 -94 22	39 4 -4 171 -170 4 -6 71 - 39 4 -4 171 -170 4 -44 18 -2 8 -2 84 -67 4 -24 0	-70 8 -17 105 99 4 94 0 -19 -6 7 -15 82 -76 5 11 112 -100 -8 8 -13 112 109 4 and 1 9 11 a	8 −12 46 −49 8 −99 0 −16 7 −6 48 52 8 4 −10 71 70 5 −7 36 35 10 −4 134 133 4 ∞ −8 61 73 4 −5 50 −46 7 −2 31 10 12
-3 111 -115 4 2 179 -176 3 6 201 -202 4 -1 252 -253 4 44 0 -17 5 8 67 62 5	-6 36 372 4 -3 70 70 4 7 110 -6 168 172 4 -1 194 193 4 -5 47 -44 23 23 7 1 328 315 5 -3 128	7 -105 4 0 139 -143 4 0 88 7 -42 6 2 94 93 4 2 35 - 7 -135 4 4 186 187 4 4 63	92 5 -1114 0 12 8 -15 44 48 12 11 -9 55 -53 8 -134 0 18 65 7 -74 21 29 8 -11 37 11	9 ~6 36 2211 -3 83 78 5 0 43 -40 9 8 ~4 30 91 5 ~1 133 -138 4 24 0 -16 8 10 ~2 106 -100 5 1 84 81 4 39 -44 10
3 119 - 117 4 8 143 130 3 124 14 - 36 8 5 152 147 4 10 66 82 4 14 107 109 4		56 6 8 288 277 5 8 54 5 27 14 10 136 133 4 10 36	1910 -5421 -137 -967 -59 547 -3104 -1024 -734 -37: 3010 -144 448 -5419 -6	6 0 90 -85 5 3 32 30 11 6 30 -30 13 0 2 83 -75 5 54 0 7 7 8 29 30 14 7 4 32 -34 13 7 47 -45 8 10 1 19 3
9 59 58 6 14 171 163 4 18 57 55 5 11 27 -22 13 154 0 4 8 8 88 4 6 9 88 5	4 104 - 94 4 9 158 - 151 4 5 178 6 71 - 81 7 11 96 - 94 4 74 84 21 29 8 13 29 31 12 9 57	3 176 4 124 0 12 8 124 3 0 -10 8 14 57 -54 6 14 66 1 -51 6 16 53 -10 6 ANN H 8 1	8 8 1 305 - 298 5 - 3 73 - 77 65 7 3 229 224 4 - 1 66 63 1 mm 5 52 50 7 1 53 28	5 6 49 43 7 9 35 2 10 88 H 12 3 88 5 5 8 85 89 5 114 24 2 8 -10 29 20 14 7 10 65 -61 9 13 54 54 8 -10 29 20 14
ana H 5 14 ana 20 72 -68 5 -20 127 119 4 -194 16 14 8 22 38 -46 12 -194 0 8 7	-14 42 -5 9 17 60 65 6 13 31 -12 50 54 8 19 73 74 6 64	5 41 9 18 37 -9 10 -18H 0 -14 13 AAA H 8 4 AAA -16 77 7 13 AAA -20 64 56 7 -14 112 :	-1 8 7 121 114 4 3 32 17 1 79 5 9 33 -515 5м 22 10 13 4 11 96 -94 4 7 53 -47	1 12 59 55 5 ann H11 3 ann -54 11 3 8 7 11 40 25 10 -134 20 19 8 -44 17 2 8 7 88 H10 5 ann -11 40 -47 10 -24 0 -18
-15 108 103 4 -22 35 31 11 -164 0 -7 8 -15 108 103 4 -22 35 31 11 -14 93 -86 5 -13 54 39 6 -20 72 -86 6 -12 53 -61 7	-10 33 -49 12 AAA H 7 5 AAA -17 38 -8 84 -82 5 -21 51 43 8 -15 55 -6 74 73 6 -194 17 12 8 -13 55	9 32:10 -164 7 10 7 -12 36 - 5 82 8 -16 36 -15 9 -10 52 - 5 -171 4 -14 60 47 7 -6 152 -	26 10 13 41 36 9 9 40 43 1 134 6 154 0 10 8 44 19 12 44 56 4 17 43 -21 8 -15 49 -46 1	2 -164/25 -30 8 -9 37 -36 10 0 46 44 8 = -144/18 -2 8 -7 28 6 12 24 0 4 7 0 -12 31 -24 12 -5 36 14 0 4 7
-11 22 -13 5 -10 37 63 5 -10 30 36 7 -9 93 -94 5 -16 58 -63 5 -6 107 105 4 -7 115 -114 4 -144 0 1 7 -6 236 -249 4	-44 25 34 7 -17 97 98 4 -114 0 -24 0 24 7 -15 63 63 6 -9 149 04 0 6 6 -13 79 79 5 -7 99	0 -4 7 -12 36 -24 10 -6 113 -1 1 -151 3 -10 123 -122 4 -44 0 1 -102 4 -6 68 68 5 -2 47 -	06 Ч. жжа н 9 Ч. ала -13, 39, -48 1 9 8 -19, 37, 23,11, -11, 52, -52 48 9 -17, 81, 62, 6, -9, 95,-101	0 -10 58 53 7 -3 115 115 4 6 44 -36 9 8 -8 74 69 5 -1 162 168 4 8 34 23 12 5 -6 107 101 4 1 116 -117 4 -8 112
-3 174 174 1 -174 0 -164 -177 3 -2 166 -165 4 -1 89 79 5 -8 161 166 3 0 229 244 4	4 42 21 9 -9 132 -125 4 -3 156 4 42 21 9 -9 132 -125 4 -3 156 man H 6 18 man -7 318 -320 5 -14 0	i 116 y -6 224 -224 y 0 64 i 156 y -4 197 199 y 2 139 1 i 25 9 -2 78 75 y y 31	67 7 -15 35 -12 10 -7 69 77. 42 4 -13 63 -50 5 -5 138 137 36 11 -11 139 -141 4 -34 0 17	64W 05 8 3 34 31 1010W 16 14 8 42 8580 6 5 114114 48W 24 26 8 9 10 2174 6 7 7269 56 1315 8
1 143 143 4 -0 31 -30 7 3 4 1 1 1 5 3 50 -52 9 -4 90 -77 3 44 0 18 6 5 61 -52 7 -2 281 276 5 6 36 16 10	-10 129 135 4 -5 190 -188 4 1 63 -8 36 -8 11 -3 70 70 5 3 120 -64 27 -19 8 -1 97 97 4 5 138	8335 0 117 119 4 694 22 - 1 116 4 2 949 -105 4 694 0 - 1 140 4 4 91 -91 4 104 20	13 7 -9 109 -112 4 -1 68 74 11 8 -7 86 -90 5 1W 20 -15 -6 7 -5 89 82 5 3 41 -22	6 2M 14 -3 8 9M 13 -2 7 -4 43 2M 9 7 4 26 1912 11M 14 -11 9 -2 80 -86 6 9 6M 20 -1 7 13 78 69 6 0 47 -6 8
9 50 -39 7 24 8 26 4 10 176 4 11 28 2 14 4 51 -58 5 12 52 56 7	-4 87 -58 5 1 241 239 4 7 125 -2 32 -27 13 3 114 -110 4 94 15 0 84 -60 5 5 197 187 4 11 37	-121 4 6 55 -53 8 12 29 - -48 8 8 178 179 5 ### 1 8 1 -40 11 10 73 -73 6 -16 63 -	10 14 -3 52 60 7 5 55 -58 2 mm -1 228 233 4 7 83 -77 58 7 1 214 213 4 9M 0 0	7 8 59 -54 6 444 11 4 444 2 30 -34 12 5 10 42 41 10 -134 25 31 8 4 44 31 10 9 124 25 -9 8 -11 10 -134 13 5 5 8 4 44 31 10
ARA H 5 15 ARA 8 193 106 4 16 112 109 4 -19 62 60 6 104 15 7 6 ARA H 6 10 ARA	Anne H 7 O Anne 9 90 -92 5 -17 53 1 256 251 4 11 46 -32 8 -15 48	714,000 12 75 -68 6 -14 35 1 46 8 144 0 8 7 -12 59 - 1 54 8 154 26 17 7 -10 49 -	1711 3 101 -105 4 mm r 9 13 m 51 6 5 53 -51 7 -13 63 -68 43 7 7 118 -112 4 -114 0 22	и мер H 10 7 жие -9 26 33 13 94 0 16 8 7 -164 0 -10 8 -74 14, 17 7 жие H 12 5 жие 8 -14, 150 155 4 -55 4 -55 15 1 7 -10 ис 72 9
-15 121 121 4 144 26 -22 7 -16 127 -124 4 -13 26 11 13 154 0 -22 9 -16 117 -126 4	5 35 -33 6 15 32 -18 11 -11 83 7 31 -1 13 17 25 -18 11 -11 13	-54 5 18 41 31 10 -5H 28 -74 5 mm 1 8 5 mm -6 49 -139 4 -20 44 37 8 -4 53	26 8 9 105 -39 5 -99 22 -21 50 7 11 51 -47 7 -7 31 171 53 7 13 45 -14 8 -54 10 4	8 -12 33 27 11 -3 27 9 13 -8 31 25 13 2 -10 58 -61 8 -1 81 -78 5 -6 76 -68 6 7 -8 33 22 10 18 0 21 7 -19 23
-11 202 -235 -1 107 13 0 -13 23 -21 12 -9 238 -244 4 20 66 72 6 -12 152 147 4 -7 187 -189 4 ада H 6 4 ада -10 35 90 4	3 133 -137 4 19 30 -37 11 -7 50 11 170 -172 4 88 H 7 7 88 -54 0 13 122 -114 4 -21 37 15 12 -3 40	45 8 -18 115 114 4 -2 57 4 7 -16 53 -42 6 04 22 - 33 8 -14 60 65 7 2 61	ЧБ 6 15 70 63 6 -334 0 -46 ЧО 8 17 39 92 5 -1 51 51 82 5 мм≉ Н 9 5 мма 1 100 104	8 –6 126 –128 ù 3 91 85 ù -2 55 –45 7 7 –4 101 100 4 54 17 –13 7 0 58 40 7 5 –72 167 –171 4 74 0 –6 7 24 0 26 4
-3 43 36 9 -20 40 -29 9 -6 143 4 -14 12 19 8 -154 0 -16 7 -4 55 54 6	17 102 185 4 -174 18 -33 7 1 123 19 38 8 10 -15 158 -173 4 34 0	-24 7 -12 111 -106 4 44 0 126 4 -10 110 -112 4 6 46 - 34 6 -8 187 -187 4 8 57 -	6 7 -19 46 18 8 3 63 -62 30 8 -17 53 48 8 5 55 38 59 7 -15⊭ 11 -6 7 7 123 -127	6 0H 17 -26 7 9 67 -62 6 4 97 -710 7 2 107 10H 4 11H 26 21 10 6 59 -52 7 4 4 36 40 10 50 H 11 5 505 50 -52 7
3 92 100 V -144 24 12 7 0 346 -355 6 5 103 105 5 -12 231 232 V 2 151 -145 V	and H 7 1 Anno -11 166 -186 4 7 41 -214 23 -17 8 -9 155 155 4 9 96	-25 9 -4 116 120 4 ### H 8 1 -100 5 -2 125 123 4 -164 0 -	3 8 -13 47 51 8 AAA H 9 14 AA 3 AAA -11 116 -114 5 -11 117 115 10 8 -9 98 -103 5 -94 15 -3	≠ 64/26 24/7 -13 61 51 7 -19 74 -76 6 4 8 40 3310 -11 34 2311 -64/0 -71 9 8 10 44 -44 9 -9 63 71 6 -4 48 -46 9
94 12 -3 8 -8 201 206 4 6 66 -57 6 11 70 -72 6 -6 234 -236 4 8 115 115 4 15 11 5 115 11 5 11 5 11 5 11	-17 34 24 10 -5 35 -27 0 -15 30 -15 151 -152 4 -3 382 360 6 -13 71	-27 13 2 102 108 4 -12 107 -1 -53 6 4 109 106 4 -10 0	8/5 -/ 53 -43 / -/ 81 /9 12 4 -5 104 99 4 -5 30 29 1 2 7 -7 35 21 10 -34 0 -17	5 12 42 35 10 -7 70 73 5 -24 22 -32 8 3 AAA 11 10 8 AAA -5 330 15 12 0 87 50 5 8 -144 10 8 6 8 -3 84 -67 5 24 0 32 9
-17 43 -53 10 -2 355 -354 6 124 26 -25 7 -154 16 -26 8 0 149 -156 3 14 65 64 8 -13 Ma -26 5 217 -217 -156 3 14 65 64 8	-11 27 27 12 1 65 51 5 -9 38 -9 36 38 9 3 208 -203 4 -7 105 -7 107 11 8 5 18 -19 105	22 10 8 81 -80 5 -6 28 105 4 10 96 -97 5 -4 131 1	25 12 1 118 -120 4 5 -1 36 -31 25 12 1 118 -120 4 1 59 -51 27 4 3 108 108 4 54 0 -22	5 -124 i7 2 7 -1 30 27 12 494 25 -1 8 7 -104 0 23 8 1 110 -112 4 6 51 45 8 8 -9 36 -35 10 3 30 -36 12 ameriti2 7 ameri 8 -9 36 -35 10 3 30 -36 12 ameriti2 7 ameri
-11 105 96 4 4 308 305 5 44 H 6 11 44 -9 129 -135 4 6 91 103 4 -20 56 58 7 -7 36 31 9 8 80 78 4 -188 20 22 7	-5 120 122 3 7 138 -132 4 -3 85 -3 23 -5 11 94 0 -6 7 -1 39 -1 105 95 1 114 21 -26 1 19	62 5 14W 0 -13 7 0 94 -40 9 16W 0 -11 8 2 50 -	23 10 5 82 30 5 844 1 1 1 1 5 84 89 5 74 0 -12 9 -7 74 63 54 8 94 26 -10 8 -54 27 -40	# ~6 270 1213 544 0 ~1 7 ~844 11 34 9 6 ~4 52 ~42 6 74 20 ~18 7 ~6 77 ~74 6 9 ~2 95 ~92 4 9 83 84 5 ~4 35 911
5 \$5 36 8 10 124 119 4 -16 81 -75 5 -3 50 52 7 124 0 5 7 -144 0 2 8 -1 112 120 4 14 31 30 12 -12 85 -83 5	1 420 -417 7 13 139 200 4 3 139 3 356 -345 6 154 0 21 7 5 30 5 152 -142 3 174 15 70 70 70 20	-138 4 Ann H 8 6 Ann 6 41 39 11 -204 27 16 8 8 103 -1 -68 -184 7 16 8 8 103 -1	37 9 13 40 8 9 -14 25 2 01 5 154 0 7 8 1 63 54	v U cri 5 11 114 256 21 9 -2 30 -17 13 0 24 19 -22 7 novi 6 11 6 novi 0 51 34 8 7 ¥ 53 50 7 -134 25 -32 9 2 36 19 11
1 40 -17 8 16 118 -114 3 -10 81 86 5 3 51 -46 7 18 45 -30 8 -8 35 24 10 5 76 -77 5 204 22 -14 8 -8 32 0 11	74 0 -6 6 ann 11 7 8 ann ann 11 9 329 323 5 -21 127 128 4 -13 115 11 150 153 4 -184 6 -3 8 -11 36	7 16 Avra - 16 134 - 139 4 - 144 19 -118 4 - 14 34 13 9 - 124 0 -29 10 - 12 95 102 5 - 10 31 -	15 8 -17 75 -79 8 0 40 -34 1 5 8 -15 86 -86 5 24 0 -20 7 12 -13 71 -76 5	8 36 101 4 -11 35 -400 7 444 28 14 8 0 8 59 -65 7 -9 10 47 9 area 11 2 8 area 8 10 66 56 7 -7 28 18 13 -6 57 94 7
74 21 - 33 8 ARA H 6 5 ARA - 4 108 121 4 SH 19 10 8 - 22 30 33 13 - 24 7 - 10 7 ARA H 5 17 ARA - 20 103 - 100 4 0 51 - 58 7	13 95 100 5 -17 49 -55 7 -9 70 154 0 0 8 -15 68 -68 5 -7 153 17 97 -93 4 -13 137 -140 4 -5 85	-60 6 -10 34 -35 11 -8 77 155 4 -8 49 33 7 -6 73 89 5 -8 99 39 4 -8 45	76 5 -11 27 27 12 64 0 -6 58 5 -9 86 78 5 8 41 -26 1	0 -114 - 20 11 8 -5 53 -53 -53 -54 -28 -30 35 9 12 32 -24 12 -14 24 -15 7 0 116 -30 11 4
-15 93 -97 5 -18 118 120 4, 2 48 -48 8 -13 97 -95 4 -16 154 -150 4, 4 66 -58 6 -11 79 -87 5 -14 42 -26 8 8 80 81 5	19 83 82 8 11 60 51 8 34 18 21 34 21 12 -9 209 212 4 1 82 86 7 56 57 58	5 7 -4 112 114 4 -24 0 -59 6 -2 139 140 4 0 179 -1	9 7 -5 254 255 5 12 65 68 51 4 -3 71 69 6 14 38 281	6 -154 123 -12 7 34 9 25 7 mm h 12 9 mm 1 -6 70 -79 6 54 25 -10 7 -24 5 -13 8
-944 0 -6 7 -12 40 42 8 8 32 -37 13 -7 31 -1 11 -10 108 112 4 10 46 -36 7 -5 156 153 4 -6 170 174 3 124 25 -26 7	-21W 29 ¥7 8 -5 52 53 6 3 ¥2 -19 56 -56 7 -3 2¥1 2¥8 ¥ 5 3¥ -17W 0 -11 8 -1 108 106 ¥ ana #	-2 9 2 41 -41 8 4 51 -24 12 4 107 -98 4 64 27 7 17 44 6 4 15 -11 7 8 4 -	22 7 1 53 -53 6 maa n 10 1 ma 7 8 3 197 -192 4 -16 34 -12 1	24 137 141 4 74 23 -7 8 4446 113 0 448 24 20 -5 7 544 0 -29 8 14 0 4 8 2 0 32 17 10 446 111 7 448 3 14 0 4 8
-3 92 95 4 -6 32 26 8 14 58 47 7 -1 75 80 5 -4 314 -301 5 ### 6 12 ### 1 95 -91 4 -2 185 180 3 -20 48 -44 8	-15 103 -97 4 1 131 130 4 -11 38 -13 179 -179 4 3 149 -147 4 -9 55 -11 60 71 5 5 51 -67 7 -7 65	46 11 8 38 48 10 ann 1 8 1 -58 8 10 7 12 30 -58 8 -12 57 -50 7 12 30 -58 12 10	ANN 7H 0 6 8 -12 52 53 16 7 9H 0 -2 7 -10H 25 -30	2 € 04 03 0 -13 00 08 6 54 27 8 8 6 4 45 19 8 -11H 24 7 8 mm H13 1 mm 7 5 53 -48 7 -9⊌ 21 15 7 -5 41 26 10
34 0 7 6 0 58 -81 5 -18 65 67 7 54 22 -12 8 2 57 44 5 -18 71 70 5 7 60 -58 7 4 172 -172 3 -14 165 187 4	-9 34 -20 8 7 114 -103 4 -5W 0 -7 166 179 4 9 63 -65 7 -3 68 -5 263 274 5 114 22 -21 7 -1 68	-37 8 14H 18 28 7 -8 143 1 -70 6 15H 10 -17 8 -8 30 -5 12 8 H 8 7 8	15 4 13 45 29 8 -6 94 -95 4 12 154 22 9 8 -40 0 13	0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0
	-3 68 67 4 13 52 55 7 1 79 -1 123 123 3 15W 26 6 7 888 H 6 1 117 -110 3 17 57 59 6 0 166	-74 5 -204 22 -10 8 -2 100 -1 8 0 mm -18 46 -37 8 0 44 -153 4 -16 90 -90 5 2 80 -	25 4 -17 37 -20 10 0H 27 13 3 9 -15 32 -29 12 2 74 78 40 6 -13 75 79 5 12 10 10	1 - 124 / 29 - 29 8 1 79 72 5 mm H 13 2 mm 6 - 10 64 - 62 6 9 57 61 7 - 3 91 97 5
-944 13 12 8 12 36 34 9 -6 255 -256 5 -7 56 -36 7 14¥ 0 -2 7 -4 110 -104 4 -5 53 83 8 16 47 -38 7 -24 0 -14 7	3 378 - 375 6 ### M 7 9 ### 24 0 5 85 - 83 4 - 218 18 4 8 48 11 7 143 - 145 4 - 19 78 - 80 6 69 0	-1 6 -14 53 -39 6 44 28 - -18 6 -12 103 -101 5 AAA H 6 1 27 7 -10 32 6 11 -10 64 -	23 8 -11 123 -123 4 64 0 15 5 AND -94 24 -23 8 84 19 -1 5 7 -74 0 29 8 10 118 -120	-64 0 -57 7 26 -18 14 13 14 15 -44 55 51 7 94 0 -6 8 3 51 -30 8
-3, 77 -80, 5, 184 0, 0, 7, 0, 42 -44, 10 -14 21 26 7, 20, 53 -33, 7, 24 14, 25 8 1, 32 -32 12 ANN H 6 6 ANN 4, 112 110 4	8 180 180 4 -17 43 -48 8 8 114 11 44 -43 8 -15 140 -148 3 104 18 13 75 69 5 -13 91 87 5 124 13	117 4 -8 163 164 4 -8 35 9 8 -8 31 -40 13 -6 41 -10 7 -4 334 337 5 -4 79 -	12 15 -5 117 117 4 124 0 -10 14 10 -3 133 131 4 14 61 -60 13 6 -1 82 -77 4 14 61 -60	7 0 91 97 5 -114 20 -26 9 -3 58 -53 7 7 2 48 -36 8 -94 0 2 8 -14 0 -6 9
3 45 -48 9 -22 51 -25 8 6 168 163 4 5 57 59 8 -20 52 31 7 8 30 -31 11 4 5 19 4 - 1 9 19 19 19 19 19 19 19 19 19	15 39 21 10 -11 34 31 11 14 58 17 159 154 4 -9 284 294 5 16 31 194 0 -29 8 -7 174 171 4 18 58	-61 8 -2 52 -51 7 -2 41 5 13 04 25 5 6 0 73 - 63 6 2 137 -139 4 2 53	29 10 1 39 -19 10 mm H 10 2 mm 1 6 3 75 -182 6 -16 35 24 1 19 8 5 35 37 12 -1144 1	6 49 26 8 -54 17 17 18 18 28 25 8 6 49 26 8 -54 12 1 7 18 18 13 4 18 1 1 64 27 -9 8 -34 16 -15 7 -1 63 -59 7
-9 52 45 8 -16 132 133 4 12 72 -70 6 -7 152 153 4 -14 57 63 6 14 38 11 11 -5 60 -44 7 -124 57 63 6 7 888 16 13 888	21 38 -30 11 -5 41 -28 8 20 0 ### H 7 3 ### -3 177 -187 4 ### H 8 -21 35 -19 11 -1 146 -141 4 -20 41	-36 8 ў 155-158 ў заа н 9 і 8 1.ааа 6 ў 9 50 8 1 223-2 -39 10 8 50 -36 8 34 0 -	0 444 7 61 -55 5 -12 36 17 1 5 4 9 32 9 12 -10 36 -19 6 8 11 28 -11 -54	
-3M 0 -1 8 -10 236 -225 4 -18 43 -12 9 -1 102 -99 5 -8 65 65 -16M 23 28 7 1M 2N -29 5 -8 65 97 -19M 23 -18 7	-19 173 -177 4 1 137 -128 4 -18 29 -17 59 54 6 3 86 -85 5 -16 43 -154 16 -11 8 5 35 36 11 -14 53	-34 14 10 54 -61 6 5 36 - -42 8 12 124 125 4 7 114 1 -52 8 14 28 -18 13 9 174 1	11 9 13-4 6 12 8 -6 159 -165 12 4 15-4 0 -3 8 -4 11 27 18 4 19 8 4 1 27	6 52 53 6 7 50 -42 7 6 39 47 10 AND H 11 9 AND 2 39 -27 11 -11 -11 -11
ana n 6 0 ana -4 49 -58 6 -12 54 63 6 0 230 237 4 -2 64 63 -10 33 -10 12 2 375 339 6 0 135 132 4 -8 66 -53 6	-13 272 275 5 7 138 135 4 -12 28 -11 141 142 4 9 90 83 5 -10 60 -9 124 123 3 114 12 10 7 -8 244	5 13 16 47 34 8 11 89 53 6 ### H 8 8 ### 13 60 259 4 -20M 26 3 9 15 106 -1	11 5 -17 65 -58 6 0 54 61 10 6 -15 73 56 5 2 91 94 15 5 -134 16 22 7 44 0 22	0 0 38 36 10 -94 18 -19 8 5 2 54 -47 7 -74 27 -39 8 9 4 36 -40 11 -54 9 70 8

to compensate for the systematic errors indicated by Fig.3. Crystallographers are accustomed to regarding the set of thermal parameters as a sink for absorbing errors; however, in the present case the compensation for errors must involve the coordinates as well, since differences in thermal parameters could not possibly compensate for the form of the curve in Fig.3. It is in fact surprising, in view of Fig.3, that the two sets of molecular parameters agree as well as they do.

Rigid-body analysis

By the method of Schomaker & Trueblood (1968) as embodied in a program by Johnson (1965) a leastsquares rigid-body analysis of the thermal parameters* U_{ij} of the S, C, and N atoms of the 6-mercaptopurine

^{*} The U_{ij} 's, derived from the B_{ij} 's of Table 4, are the components of the tensors of mean-square vibrational displacement referred to a Cartesian coordinate system.

molecule was performed, with unit weight for each U_{ij} . The unusually low value, 0.0009 Å² for the standard deviation of an observation of unit weight, which is in this case the root-mean-square ΔU_{ij} corrected according to the number of degrees of freedom, shows that the fit to the rigid-body model is excellent.

In the rigid-body analysis the origin was chosen^{\dagger} so as to make S symmetric and minimize the trace of T, and the constraint that the trace of S be zero was applied. The detailed description of the rigid-body motion resulting from the analysis is given in Table 6, including the description of the motion preferred by Schomaker & Trueblood, in terms of 3 independent screw librations (helical motions) about non-intersecting axes and 3 reduced translations.

The root-mean-square amplitudes about the principal axes of librational motion are 3.13° , 2.54° , and 2.46° for axes K=1, 2, and 3 respectively. Axis 1 lies only 4° from the molecular plane; axis 2, 13° ; axis 3, 76° .

[†] The choice was actually made by letting the program find this origin in a preliminary calculation starting with an arbitrary origin.

Fig.4 shows graphically how well the calculated and observed ellipsoids agree. The left side of the Figure shows, at the top, the observed 50% probability ellipsoids (see Johnson, 1965) viewed in the direction perpendicular to the best plane through the nonhydrogen atoms and, at the bottom, the same ellipsoids viewed at 60° from the perpendicular. For each nonhydrogen atom the three principal r.m.s. vibrational amplitudes (Å) are given, and for each hydrogen atom the single r.m.s. amplitude corresponding to the isotropic thermal parameter B is given. The right side of the Figure shows corresponding views of the calculated ellipsoids from the rigid-body analysis, with the calculated r.m.s. amplitudes. Calculated anisotropic ellipsoids are included for the hydrogen atoms. The plus sign (+) in each molecular drawing marks the center of gravity of the molecule, and the cross (\times) marks the origin used in the rigid-body calculation. The center of gravity and the origin are respectively -0.001 Å and +0.090 Å from the best plane, the latter deviation being toward the molecule related to the reference molecule by inversion through the center at $\frac{1}{4}, \frac{3}{4}, \frac{1}{2}$ (see Fig.3 of Sletten *et al.* and Fig.6 below).

Table 6. Description of the motion of the 6-mercaptopurine molecule from the Schomaker–Trueblood rigid-body analysis

The Cartesian coordinate system to which the tensor components below are referred is oriented as follows: base vector **i** is along the unit normal **n** to the least-squares best plane through the 10 nonhydrogen atoms of the molecule; base vector **k** is along $\mathbf{n} \times \mathbf{m}$, where **m** is the vector from C(5) to C(4); base vector **j** is along $\mathbf{k} \times \mathbf{i}$.

		Crys	tal coordina	tes		Ca	rtesian coo	ordinate	s (Å)
Components of n Centroid Origin for calculatio	n (see text)	x -0.03408 0.32651 0.31195	y 0·10912 0·56911 0·57309	z -0.021 0.469 0.489	185 983 915		Y 0.0 -0.32 0.0	70	Z 0·0 0·0118 0·0
-		Tenso	or elements	x 105 (st	andard e	rrors × 1	02)		
Tensor L (rad. ²) T (Å ²) S (rad. × Å)	11 186 (9) 2308 (56) -6 (10)	22 219 (17) 1735 (33) 39 (16)	33 276 (2374 (-33 (16) 31) 18)	$ \begin{array}{r} 12 \\ -2 (1) \\ 76 (3) \\ -21 (1) \end{array} $	3) 8) 1)	13 9 (12) 116 (34) -9 (12)	4 25 2	23 3 (12) 9 (28) 7 (22)
						Helio	al axis K	or princi	ipal axis K
Helical motions about no R.m.s. amplitude (radians) (degrees) Pitch (Å rad. ⁻¹)	on-intersecting	; axes				1 0.054 3.13 -0.130	17 ()	2 0·0444 2·54° 0·172	3 0·0429 2·46° 0·028
Orientation angles of l	nelical axes to	Cartesian base	vectors		i j k	85·6° 118·0° 28·4°	10 15 11	2•8° 0•2° 6•4°	13·6° 99·3° 99·8°
Displacement (Å) of ea	ach helical axi	s K from the ot	her two helic	al axes J	$\begin{array}{c} J J=1 \\ 2 \\ 3 \end{array}$	0.028 0.052	3 · 2 —	0·156 0·079	-0·167 0·045
Reduced translation R.m.s. amplitude (Å)						0.158	3	0.151	0.127
Orientation angles of p	orincipal axes	to Cartesian bas	se vectors		i j k	65·3° 104·4° 29·1°	15 10 6	3·3° 6·9° 9·9°	80·3° 157·5° 110·1°

Of course, only for the nonhydrogen atoms are the calculated and observed ellipsoids directly comparable; and for these the agreement seems remarkably good, consistent with the low value of the standard deviation of an observation of unit weight quoted above. The differences between the calculated and the observed ellipsoids for atoms C(4), C(5), and C(6) which appear in the drawings are misleading, because for each of these atoms both the calculated and observed ellipsoids are nearly ellipsoids of revolution, with only one principal-axis direction well determined.

Rigid-body analyses were also performed on the sets of thermal parameters U_{ij} from least-squares refinement calculations (6) and (7) (see Table 2). The descriptions of thermal motion resulting are quite close to the one already given from the U_{ij} of calculation (8). The standard errors of the components of L, T, and S are somewhat larger, however, than those in Table 6; and the standard errors of fit are higher, both about 0.0011 Å². This suggests that the parameters of calculation (8) really are more reliable than those of (6) and (7).

Molecular structure

Perhaps the most interesting result so far as molecular structure is concerned is the finding that the tautomeric

form of the 6-mercaptopurine molecule in the crystal is that of the thione of formula I, with hydrogen atoms on N(1) and N(7). So far as the five-membered ring is concerned, this structure corresponds to that of purine (Watson, Sweet & Marsh, 1965).



The finding of the thione form is in agreement with results from spectroscopic studies. Apparently the only



Fig.4. Comparison of observed thermal ellipsoids (left side of Figure) for the nonhydrogen atoms of the 6-mercaptopurine molecule with ellipsoids calculated (right) from the rigid-body analysis according to the Schomaker-Trueblood method. Top views are in the direction of the normal to the best plane through the nonhydrogen atoms; bottom views are in the direction 60° from the normal. The principal r.m.s. vibrational amplitudes are shown. The symbol + marks the centroid of the molecule and the symbol x marks the origin used in the rigid-body analysis.

physical evidence for a thiol tautomeric form for 6mercaptopurine is in the report (Mason, 1957) that 2-, 6-, and 8-mercaptopurines in the solid state show weak infrared bands in the region of the S-H stretching frequency, but this report included the cautionary remark that these bands are not unequivocal evidence for the thiol forms. The weight of the evidence from ultraviolet spectroscopy (Elion, 1957; Mason, 1957) of neutral and acid solutions of 6-mercaptopurine and various monomethyl derivatives is that a thione form predominates. Similarly, an infrared study of a chloroform solution of 7-methyl-6-mercaptopurine (Mason, 1957) suggests that 6-mercaptopurine, which is itself not soluble in chloroform, exists partly in thione form. Katritsky & Ambler (1963) make the generalization that, 'most six-membered heteroaromatics with mercapto groups α or γ to a ring nitrogen exist predominantly in the thione form.' For 6-mercaptopurine the thione form has usually been formulated as in II, with a hydrogen atom on N(9) instead of on N(7), because of the similarity of the ultraviolet spectra of the parent compound and its 1-methyl and 9-methyl derivatives (Elion, 1957). In spite of the generally contrary evidence from spectroscopy, the thiol formula, III, has continued to appear frequently in the chemical and medical literature, consistent with the established use of the misnomer 6-mercaptopurine.

Bond lengths and valence angles as calculated directly from the parameters of least-squares calculation (8) are listed in Table 7; those in the 6-mercaptopurine molecule are also shown in Fig. 5. The librational ten-

O - H(11)

0.837 (18)



Fig. 5. Bond lengths (Å) and valence angles (°) in the 6-mercaptopurine molecule.

sor from the rigid-body analysis was used to correct each bond length between heavy atoms for the effects of librational motion in the way indicated by Busing & Levy (1964), equivalent to the prescription of equation (22) of Schomaker & Trueblood (1968). The corrected bond lengths are included in Table 7. Although the corrections are small, from 0.0025 to 0.0039 Å, the average value of 0.0031 Å is 2.2 times the average standard error of the uncorrected lengths. Since the fit to the rigid-body model is exceptionally good, the corrections should be quite reliable.

Table 7. Bond lengths and angles in the crystal structure of 6-mercaptopurine monohydrate, with standard errors For the bonds in the 6-mercaptopurine molecule not involving hydrogen atoms corrected values computed from the results of a rigid-body analysis are included (italic numerals).

Bond ler	ngths (Å)	Angles (°))
N(1)-C(2)	1.3583 (13)	C(6) - N(1) - C(2)	124.74 (10)
C(2)-N(3)	1.3088 (16)	C(2) - N(3) - C(4)	124.97 (10) 113.52 (08)
N(3)-C(4)	1·3133 1·3609 (13)	N(3) - C(4) - C(5) C(4) - C(5) - C(6)	123·91 (09) 121·65 (09)
C(4) C(5)	1.3641	C(5) - C(6) - N(1)	111.18 (08)
C(4)-C(3)	1.3971	N(1) - C(6) - S	120.04 (08)
C(5)-C(6)	1·4050 (14) <i>1·4077</i>	C(6)—C(5)–N(7) C(4)—C(5)–N(7)	132·52 (08) 105·83 (09)
C(6)-N(1)	1·3720 (13)	C(5) - N(7) - C(8)	106.17 (08)
C(6)-S	1.6786 (09)	C(8) - N(9) - C(4)	104.16 (09)
C(5)-N(7)	1.6825 1.3729 (14)	N(9)—C(4)-C(5) N(9)—C(4)-N(3)	110·38 (09) 125·71 (08)
N(7)-C(8)	<i>1-3759</i> 1-3523 (15)	C(6) - N(1) - H(1) H(8) - C(8) - N(9)	119·9 (9) 123·4 (8)
$C(\theta)$ $N(\theta)$	1.3550	H(1) - N(1) - C(2)	115.3 (9)
	1.3359	H(1) - C(2) - H(2) H(2) - C(2) - N(3)	113·0 (8) 121·9 (7)
N(9)–C(4)	1·3675 (14) <i>1·3702</i>	C(5)N(7)-H(7) H(7)N(7)-C(8)	126·3 (10) 127·5 (10)
N(1)-H(1) C(2)-H(2)	0.880 (15) 0.956 (12)	N(7) - C(8) - H(8) H(10) - O - H(11)	$123.1(8)^{(14)}$
N(7)-H(7)	0.874 (15)	II (10)-0 II (11)	101 9 (14)
$C(\delta) - H(\delta)$	0.934 (13)		

Such small corrections, though applicable in principle to bonds C(2)-H(2), C(8)-H(8), N(1)-H(1), and N(7)-H(7), have no practical meaning for these bonds because of the larger standard errors of the apparent bond lengths and, more important, because of the usual large systematic error of shortening which is observed for these bonds and for bonds in the water molecule. The orientations of the thermal ellipsoids for H(2) and H(8) obtained in least-squares calculation (7) (see Fig. 1) suggest that as a gross approximation one might be justified in using the 'riding' model (Busing & Levy, 1964) for correcting the lengths of C(2)-H(2) and C(8)-H(8). However, the corrections calculated from the parameters of least-squares calculation (7) turn out to be only 0.019 and 0.016 Å; the bond lengths are still far from the known C-H internuclear distance of about 1.08 Å.

It is worth emphasizing that for 6-mercaptopurine monohydrate the use of anisotropic thermal parameters, successful in a formal mathematical sense, has not led to the pleasing kind of result reported by Delaplane & Ibers (1967) from a study of oxalic acid dihydrate, namely, that 'normal O-H bond lengths were obtained after appropriate corrections for thermal motion'. Delaplane & Ibers suggest that a possible cause for the short bond lengths O-H, C-H, and so forth,

usually obtained from X-ray analyses is the use of the isotropic model for hydrogen vibration. The experience with 6-mercaptopurine offers no support for this view; thus, the relevant comparison, 6-7, in Table 3 shows an average absolute difference in lengths of bonds involving hydrogen which is less than the average standard error of the bond lengths. The very close agreement between the C-H, N-H, and O-H bond lengths derived from this analysis and those derived from the parallel analysis of Sletten, Sletten & Jensen (1969) is a strong argument for the view (see Stewart, Davidson & Simpson, 1965) that the centroids of the hydrogen electron distributions are shifted from coincidence with the proton positions as a result of chemical bonding.

The agreement in the values of the angles involving hydrogen atoms between this study and that of Sletten et al. is remarkably good (see Table 7 in this paper and Table 2 of Sletten et al.). Not one of the differences is as much as the corresponding standard error. Moreover, the standard errors of the angles are not large, being only about 1° except for the H-O-H angle, which involves two hydrogen atoms. It appears, therefore, that the directions, as distinct from the lengths, of the C-H, N-H, and O-H bonds have been determined to a degree of precision as good as that often

Table 8. Distances of atoms from best least-squares planes in the 6-mercaptopurine molecule

Distances in boldface type indicate the atoms included in each least-squares plane calculation. The Table includes the distances to the molecular centroid and the origin used in the rigid-body analysis and the distances to atoms involved in hydrogen bonding with atoms in the 6-mercaptopurine molecule; e.g. O [H(1)] means the oxygen atom to which H(1) is hydrogen bonded. The last column gives the standard deviations of position of each atom in the direction of the plane normals (essentially the same for planes a, b and c). The equations of the three planes are:

(a)	7.18355x -	·6·51266y+	-2.03908z =	—0∙40435 Å

(b)	$7 \cdot 37213x - 6 \cdot 45951y + 2 \cdot 00582z = -0 \cdot 32746$	
(c)	$7 \cdot 16989x - 6 \cdot 54277y + 1 \cdot 90682z = -0 \cdot 48057$	

.109	09 <i>x</i> –	0.2471	iy +	1.20097	2z = -0	4000
------	---------------	--------	------	---------	---------	------

Displacement nom plane (A)

	(a)	(<i>b</i>)	(c)	σ
S	-0.0230	-0.0537		0.0004
N(1)	0.0291	0.0100		0.0011
C(2)	0.0040	-0.0025		0.0014
N(3)	-0.0170	-0.0052	0.0120	0.0012
C(4)	-0.0128	0.0048	0.0002	0.0011
C(5)	-0.0028	0.0025	-0.0002	0.0011
Cíó	0.0047	-0.0097	0.0127	0.0010
N(7)	0.0125	0.0290	0.0002	0.0012
C(8)	0.0103		0.0000	0.0012
N(9)	-0.0049	0.0310	-0.0001	0.0013
HÌIÌ	0.092	0.061		0.014
H(2)	0.044	0.032		0.012
H(7)	0.055		0.034	0.014
H(8)	0.004		-0.012	0.012
O [H(1)]	0.016	0.11		
O [H(2)]	0.52	0.20		
O [S]	2.83	2.78		
O [N(9)]	0.20		0.52	
N(3) [H(7)]	-0.02		-0.09	
H(7) [N(3)]	-0.31	-0.29		
H(10) [N(9)]	0.28		0.29	
H(11) [S]	1.94	1.89		
$\frac{1}{4}, \frac{3}{4}, \frac{1}{2}$	1.665			
$\frac{1}{4}, \frac{1}{4}, \frac{1}{2}$	-1.592			
Centroid	-0.001			
Rigid-body origin	0.090			

reported for bonds C-C, C-N, etc. in less precise work. To complete the specification of atomic coordinates in the crystal, it seems appropriate to compute corrected coordinates for the hydrogen atoms. This may be done for a C-H bond, for example, by 'moving' the hydrogen atom along the bond direction away from the carbon atom the appropriate distance to correct for the systematic error in bond length. The adjusted hydrogen coordinates given in Table 4 were obtained by this procedure. The lengths assumed for the internuclear separations were: N(1)-H(1), 1.06; N(7)-H(7), 1.04; O-H(11), 1.01; O-H(10), 0.97; C(2)-H(2), 1.08 Å. (The first three values are the expected lengths in Table 4 of Sletten et al.) These corrected coordinates are surely to be preferred to the uncorrected ones for calculation of van der Waals contacts and the description of hydrogen bonds.

The equations of the least-squares best planes were calculated by the method of Schomaker, Waser, Marsh & Bergman (1959) for (a) the 10 atoms of the 6-mercaptopurine molecule excluding hydrogen atoms, (b) the atoms of the six-membered ring only, and (c) the atoms of the five-membered ring only. The three equations and the distances (with standard errors) of various atoms from each plane are given in Table 8. Equation (a) is very close to the equation given by Sletten *et al.* for the same 10 atoms. Moreover, comparison of the distances of the atoms from plane (a) with those in Table 3 of Sletten *et al.* shows that the details of the distortion from exact coplanarity of the atoms are nearly the same in the two structures.

The separate calculations (b) and (c) for the two different rings provide a better basis for understanding the distortions from planarity. Thus, the atoms of the six-membered ring are much closer to being coplanar than their distances from the overall plane (a) might suggest without careful scrutiny; and the five atoms of the other ring are nearly perfectly coplanar. The major departures from coplanarity result from bending about the bond C(4)–C(5) and from bending of the external bonds C–S, N–H, and C–H. The angle between the normals to planes (b) and (c) is 1·16°. The sulfur atom is out of plane (b) by 0·054 Å, and the bond C(6)–S makes an angle of 1·50° with plane (b).

When the equations of planes (b) and (c) above were recomputed with the coordinates from least-squares refinement calculation (6), the fit in each case was slightly poorer than for the coordinates from calculation (8) used above. Thus, the sums of the squares of the deviations from the planes for the atoms included in the best-plane calculations were 6.38×10^{-6} and 2.90×10^{-4} instead of 1.41×10^{-7} and 2.56×10^{-4} Å² for (a) and (b) respectively. Similarly, the sums are 3.70×10^{-6} and 2.81×10^{-4} Å² for planes calculated from the coordinates of least-squares refinement (7). Since it seems improbable that a closer approach to coplanarity of the atoms in each ring would occur accidentally, this is another indication that the coor-

Table 9. Parameters of the hydrogen bonds in the crystal structure of 6-mercaptopurine monohydrate The interaction C(2)-H(2)···O is included. The distances and angles in italics correspond to the adjusted coordinates for the hydrogen atoms (see text).

	Distances (Å)			
$X - H \cdots Y$	$\overline{X \cdots Y}$	Х-Н	$\mathbf{H}\cdots \mathbf{Y}$	Angle (°)
N(1)-H(1)····O	2.758	0.88	1.88	172.4
N(7)-H(7)····O	2.910	1·06 0·87	1·70 2·04 1.87	171·6 174·2 173-7
OH(11)····S	3.379	0.84 0.97	2·56 2·43	166·3 165·6
$\mathbf{O} \longrightarrow \mathbf{H}(10) \cdots \mathbf{N}(9)$	2.800	0·82 1·01	1·99 1·80	170·9 170·0
$C(2)-H(2)\cdots O$	3.331	0·96 <i>1·08</i>	2·39 2·26	170·4 169·9

Fig.6. Stereoscopic view of the crystal structure of 6-mercaptopurine monohydrate. The direction is of view close to $-\mathbf{b}$; \mathbf{c} is nearly horizontal; \mathbf{a} is nearly vertical. The parallelepiped outlined has dimensions a, b, c and is centered on the point $\frac{1}{2}, 0, \frac{1}{2}$.

dinates from refinement calculation (8) are indeed more reliable than those from (6) or (7).

Molecular packing and hydrogen bonding

A stereoscopic drawing of the crystal structure is shown in Fig. 6 to complement the projection drawings of Sletten, Sletten & Jensen (1969).

In the stacking of the molecules along the *b* axis, the concave side of the reference molecule (see Fig. 3 of Sletten *et al.*) is in contact with the molecule related by the symmetry center at $\frac{1}{4}, \frac{3}{4}, \frac{1}{2}$ (Fig. 4 of Sletten *et al.*), the spacing between the two overall average planes being 3.33 Å.

The description of the hydrogen bonds is given in Table 9. The C-H···O interaction is included, although according to Donohue (1968) it would not be considered a hydrogen bond in the usual sense. The corrected distances and angles in Table 9 were calculated using the corrected hydrogen parameters given in Table 4 (see explanation above under heading *Molecular structure*). The corrected angles are not significantly different from the uncorrected. The uncorrected hydrogen bond parameters are, of course, very close to those of Sletten *et al.*

Table 8 includes the distances from planes (a), (b), and (c) to the various atoms linked to the 6-mercaptopurine reference molecule by hydrogen bonds. As Sletten *et al.* point out, the departures from planarity of the molecule are not solely determined by the disposition of the hydrogen bonds; for example, the sulfur atom is 0.054 Å out of least-squares plane (b) in the direction opposite to that in which it is hydrogen bonded to the water molecule.

The computer programs used in the various calculations are as follows:

- Least-squares determination of cell parameters: part of the program package for the computer-controlled diffractometer (Busing *et al.*, 1968).
- Absorption corrections: program modified by R. D. Ellison & H.A. Levy from *ORABS* (Wehe, Busing & Levy, 1962).
- Fourier syntheses: a version of the program $F \emptyset R$ -DAPER of A.Zalkin modified by G.Brunton; program COMFO by G.M.Brown.
- Least-squares refinement of structure parameters: program XFLS, modified by R.D. Ellison, H.A. Levy & H. Yakel from ORFLS (Busing, Martin & Levy, 1962).
- Bond lengths and angles: program *ORFFE* II by C.K. Johnson and modified version by R.D.Ellison & H.A.Levy.
- Rigid-body analysis: program ORSBA of C.K.Johnson.
- Least-squares best plane: program *BSPLAN*, modified by G.M.Brown from a program of W.C.Hamilton.

Table of F_o 's and F_c 's: program *EDIT* of G. M. Brown. Drawings: program *ORTEP* (Johnson, 1965).

I am most grateful to Professor L. H. Jensen, Dr E. Sletten, and Dr J. Sletten for supplying me with their data and results and especially for delaying their publication until I could finish this work.

References

- BROWN, G. M. (1967). Abstr. Amer. Cryst. Ass. p. 26. Summer Meeting, Minneapolis, Minnesota.
- BUSING, W. R., ELLISON, R. D., LEVY, H. A., KING, S. P. & ROSEBERRY, R. T. (1968). The Oak Ridge Computer-Controlled X-ray Diffractometer. Report ORNL-4143, Oak Ridge National Laboratory, Tennessee.
- BUSING, W. R. & LEVY, H. A. (1957). Acta Cryst. 10, 180.
- BUSING, W. R. & LEVY, H. A. (1964). Acta Cryst. 17, 142.
- BUSING, W. R., MARTIN, K. O. & LEVY, H. A. (1962). ORFLS, A Fortran Crystallographic Least-squares Program. Technical Memorandum 305, Oak Ridge National Laboratory, Tennessee.
- CADY, H. H. & LARSON, A. C. (1965). Acta Cryst. 18, 485.
- CRUICKSHANK, D. W. J. (1956). Acta Cryst. 9, 915.
- DELAPLANE, R. G. & IBERS, J. A. (1967). Amer. Cryst. Ass. Abstr. p. 52. Winter Meeting, Atlanta, Georgia.
- DONOHUE, J. (1968). Structural Chemistry and Biology, p. 443. Eds. A. RICH & N. DAVIDSON. San Francisco: Freeman.
- ELION, G. B. (1957). Ciba Foundation Symposium on the Chemistry and Biology of Purines, p. 39. Eds. G. E. W. WOLSTENHOLME & C. M. O'CONNOR. London: Churchill.
- FRITCHIE, C. J. (1966). Acta Cryst. 20, 27.
- FURBERG, S. & JENSEN, L. H. (1968). Private communication.
- HAMILTON, W. C. (1961). Acta Cryst. 14, 185.
- HAMILTON, W. C. (1965). Acta Cryst. 18, 502.
- HOOGSTEEN, K. (1956). Nature, Lond. 178, 379.
- Howells, E. R., Phillips, D. C. & Rogers, D. (1950). Acta Cryst. 3, 210.
- JEFFREY, G. A. & CRUICKSHANK, D. W. J. (1953). Quart. Rev. Chem. Soc. (London), 7, 335.
- JOHNSON, C. K. (1965). ORTEP, A Fortran Thermal-Ellipsoid Plot Program for Crystal-Structure Illustrations. Report 3794, revised, Oak Ridge National Laboratory, Tennessee.
- KATRITSKY, A. R. & AMBLER, A. P. (1963). *Physical Methods in Heterocyclic Chemistry*, 2, p. 161. Ed. A. R. KATRITSKY. New York: Academic Press.
- LEVY, H. A. (1966). Unpublished work.
- MASON, S. F. (1957). Ciba Foundation Symposium on the Chemistry and Biology of Purines, p. 60. Eds. G. E. W. WOLSTENHOLME & C. M. O'CONNOR. London: Churchill.
- SCHOMAKER, V. & TRUEBLOOD, K. N. (1968). Acta Cryst. B24, 63.
- SCHOMAKER, V., WASER, J., MARSH, R. E. & BERGMAN, G. (1959). Acta Cryst. 12, 600.
- SLETTEN, E., SLETTEN, J. & JENSEN, L. H. (1969). Acta Cryst. B25, 1330.
- STEWART, R. F., DAVIDSON, E. R. & SIMPSON, W. T. (1965). J. Chem. Phys. 42, 3175.
- WATSON, D. G., SWEET, R. M. & MARSH, R. E. (1965). Acta Cryst. 19, 573.
- WEHE, D. J., BUSING, W. R. & LEVY, H. A. (1962). A Fortran Program for Calculating Single Crystal Absorption Corrections. Technical Memorandum 229, Oak Ridge National Laboratory, Tennessee.
- WILSON, A. J. C. (1942). Nature, Lond. 150, 151.
- ZACHARIASEN, W. H. (1965). Trans. Amer. Cryst. Ass. 1, 33.