CHEMICAL CONTROLS ON TETRAHEDRAL SITE OCCUPANCY IN AMPHIBOLES

Thesis for the Degree of M. S.

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STUART D. HANSON

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ABSTRACT

CHEMICAL CONTROLS ON TETRAHEDRAL
SITE OCCUPANCY IN AMPHIBOLES

By

Stuart D. Hanson

Amphiboles from an amphibolite in Ontario, Canada, show a range of compositional variation that is independent of the metamorphic grade as reflected in the plagioclase compositions. Compositional changes in the amphibole include increases in Mg and Si associated with decreases in Al and Fe. As there are only two mineral phases present in the amphibolite, amphibole and plagioclase, the Mg in the amphibole is the bulk Mg in the rock. The changes in composition suggest that the variation observed is not directly associated with changes in temperature or pressure, but is a direct function of the bulk chemical composition of the rock.

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Stuart D. Hanson

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INTRODUCTION

The purpose of this thesis is to determine whether there exists a systematic relationship governing the relative Al and Si occupancy of the tetrahedral site in amphiboles. The ionic radius of Al allows the cation to coordinate with oxygen in either the octahedral or tetrahedral mode. The amphibole structure is composed of double chain tetrahedral layers with cation layers in between. The cations occur in a layer of three good octahedral sites labeled M_1, M_2, and M_3, a less perfect six- or eight-fold M_4 site and a single twelve-fold A site on the other side of the tetrahedral layer. Al, as one of the smaller cations, has the option in terms of size of occupying the tetrahedral, or the M_1, M_2, or M_3 octahedral sites (Gibbs, 1966).

Harry (1950) proposed that a systematic relationship between Al and Si in amphibole tetrahedral sites may exist, and that it may serve as an effective geothermometer in metamorphic rocks. Using amphibole analyses selected from the literature, Harry observed an increase in the Si as temperature increased. Engel and Engel (1962 a, b) found no such relationship in amphiboles when passing from the amphibolite facies to the granulite facies. Dodge, Papike and Mays (1968) suggested that the relationship might hold as modified by pressure, and Leake (1965) suggested that composition and associated minerals may also affect tetrahedral site occupation.

The major thrust of this paper is to select an amphibole-bearing rock body whose composition is relatively constant and which has been subjected to a metamorphic gradient. In such a case the controls on Al, Si tetrahedral site occupancy can be determined. On this basis a series of basic metavolcanics in Tudor, Grimsthorpe and Cashel Townships, Ontario, Canada were selected. They are described by Lumbers (1968, 1969) as largely mafic flows originally basalt and andesite with minor inclusions of mafic pyroclastics and felsic flows. Lumbers defines a regional metamorphic gradient for the townships increasing from a greenshist facies in Tudor and Grimsthorpe Townships to an amphibolite facies in the central and northern parts of Cashel Township. He defined the gradient on the basis of zoned albite, oligoclase and andesine isograds which he extended from Limerick Township on the west into the amphibolite band. These isograds are truncated by the metamorphic aureole of the Westlemkoon Batholith in eastern Cashel Township. For the purposes of this study this appeared to be an ideal area, for apparently chemically similar rocks were subjected to a steep metamorphic gradient.

METHOD

Leake (1965) suggested that the nature of the associated minerals could affect the elemental variations within the amphibole structure. Therefore, in order to limit the number of possible

variables, amphibolites which contained only plagioclase and amphibole were selected for testing. The advantage of using these amphibolites is that all the elements are partitioned between only two mineral phases (amphibole and plagioclase) and such rocks are fairly common over the whole area. Whole rock, plagioclase, and amphibole major element analyses were done.

PROCEDURE

Samples were collected along the amphibolite belt in Cashel Township and at one location on the Jordan Road in Tudor Township south of Cashel Township. Seventy samples were thin-sectioned and examined microscopically to insure that the amphibole and plagioclase were the only primary phases present. From these, twenty-six thin sections containing only the two phases were polished for microprobe examination, and one hundred milligrams of each sample was also prepared for X-ray fluorescence analysis.

The polished sections were analyzed on an ARL EMX electron microprobe equipped with three channels. The beam was focused to a diameter of approximately 0.5 μ at an accelerating voltage of 15 kV and a specimen current of 0.020 μamps. The low specimen current precluded the volatilization of significant amounts of Na from the samples.

Nine thin sections from a geographic array approximately at right angle to Lumbers' 1968 isograds and covering the entire area

were analyzed. Five point analyses per grain were made on two grains of amphibole and two grains of plagioclase per sample. Point analyses were carried out for Si, Al, Mg, Fe, Ca, Na, using SiO_2, Al_2O_3, MgO, Fe_2O_3, albite and anorthite as standards. Two traverses were necessary for each grain to analyze for the six elements. Another continuous beam sweep was made of each grain for Al and Si in order to insure the homogeneity necessary to demonstrate equilibrium conditions.

The raw microprobe counts were then reduced to weight percent analyses using a modification of the Bence and Albee, Electron Probe Oxide Analysis Computer Program (Bence and Albee, 1968). The program, named Mprobe, was originally written as Specimen by Mrs. Kook Huber and debugged by L. G. Medaris and C. J. Bowser (all from the University of Wisconsin at Madison) using correction factors for ten elements and requiring count-ratio input instead of raw microprobe counts. The program was further modified and debugged by the author to accept raw microprobe data and calculate weight percent analyses of up to fifteen elements from an array of thirty-five elements using the Albee and Ray (1970) correction factors (Appendix).

As only three elements could be analyzed on the microprobe at one time, it was decided to run three traverses of each amphibole grain and two of each plagioclase grain. The first traverse in each case was

a continuous sweep of the entire grain which was recorded on an X-Y plotter. If the grains in one sample were consistently homogeneous, this traverse would demonstrate that the rock was at or near equilibrium. All samples measured were homogeneous with the exception of the two southernmost, CA 7-4 and CA 7-1. For the amphibole grains, the next two analyses consisted of five points, each fifty μ apart, with one traverse for Si, Mg and Fe and the other for Al, Ca and Na. The elements chosen are the major constituents of most amphiboles, and the larger the constituent percentage measured, the more accurate the weight percent analysis from the modified Bence and Albee program. These six elements comprised an average of 94.5 percent of each sample and were never less than 89.0 percent.

The use of Al, Ca and Na for plagioclase analysis also allowed a three way cross-check of the determination. The three final determinations were in each case $\pm$ 1 percent.

The fluorescence samples were run on a General Electric X-ray fluorescence unit in a helium atmosphere at 45 kV and 49.5 milliamps for 100 seconds for each of the elements Si, Al, Mg, Ca, and K. Calibration curves were prepared based on five U. S. Geological Survey standards run with the samples. Least square curves of best fit were calculated with the aid of the M.S.U. Agricultural Experimentation Station least squares (LS) program. The X-ray fluorescence of each sample provided a weight percent analysis of the bulk composition in terms of Si, Al, Mg, Ca, and K.

TABLE I

DATA

SAMPLE	AMPHIBOLE WEIGHT PERCENTS				PLAGIOCLASE	WHOLE ROCK WEIGHT PERCENTS		
Number	SiO_2	Al_2O_3	MgO	Fe_2O_3	Composition	MgO	SiO_2	Al_2O_3
CA 7-4	40.7	10.6	11.1	15.7	An_{25--62} zoned	4.4	47.5	16.0
CA 58	36.6	14.6	7.4	20.1	An_{26}	4.1	43.8	12.0
CA 19	35.0	15.1	7.0	19.0	An_{27}	1.8	48.6	15.4
CA 7-1	44.5	11.9	11.5	15.5	An_{30-67} zoned	4.0	46.4	16.2
CA 31	41.7	13.7	12.3	16.1	An_{30}	4.8	47.8	14.9
CA 29	39.4	17.3	9.3	17.6	An_{30}	3.1	48.5	15.2
CA 44	41.2	12.7	12.1	15.4	An_{33}	5.3	48.9	15.6
CA 22	39.5	14.4	9.9	15.4	An_{35}	4.0	46.8	14.3
CA 6	38.2	15.8	8.2	18.2	An_{54}	2.7	55.6	15.0

DISCUSSION OF DATA

Plagioclase compositions measured from An_{25} to An_{67}, and follow no consistent regional pattern (Figure 1). The homogeneity of the plagioclase grains indicates that the metamorphic grade is above that where the peristerites are stable. Lumbers' (1968) isograds were derived from a consistent regional pattern of plagioclase composition. The results reported here are inconsistent with such a pattern. It is possible that post-metamorphic intrusions such as the Westlemkoon Batholith to the east of the amphibolite metavolcanic belt had a more extensive aureole effect or that Lumbers' (1968) isograds are not accurate. The lack of a well defined metamorphic gradient precluded evaluation of the response of amphibole composition to a gradient. However, there do exist ranges of amphibole and plagioclase compositions (Table 1), and those ranges provide the opportunity to test the response of amphibole composition to slight changes in plagioclase composition in rocks with small bulk composition variation.

Plots of Si and Al weight percent in amphibole versus plagioclase composition showed no systematic change (Figure 2). This lack of relationship between Al and Si and the plagioclase composition indicates that the amounts of Al and Si in amphibole is independent of plagioclase composition. Although these rocks do not exhibit a wide range in whole rock composition, this suggests that

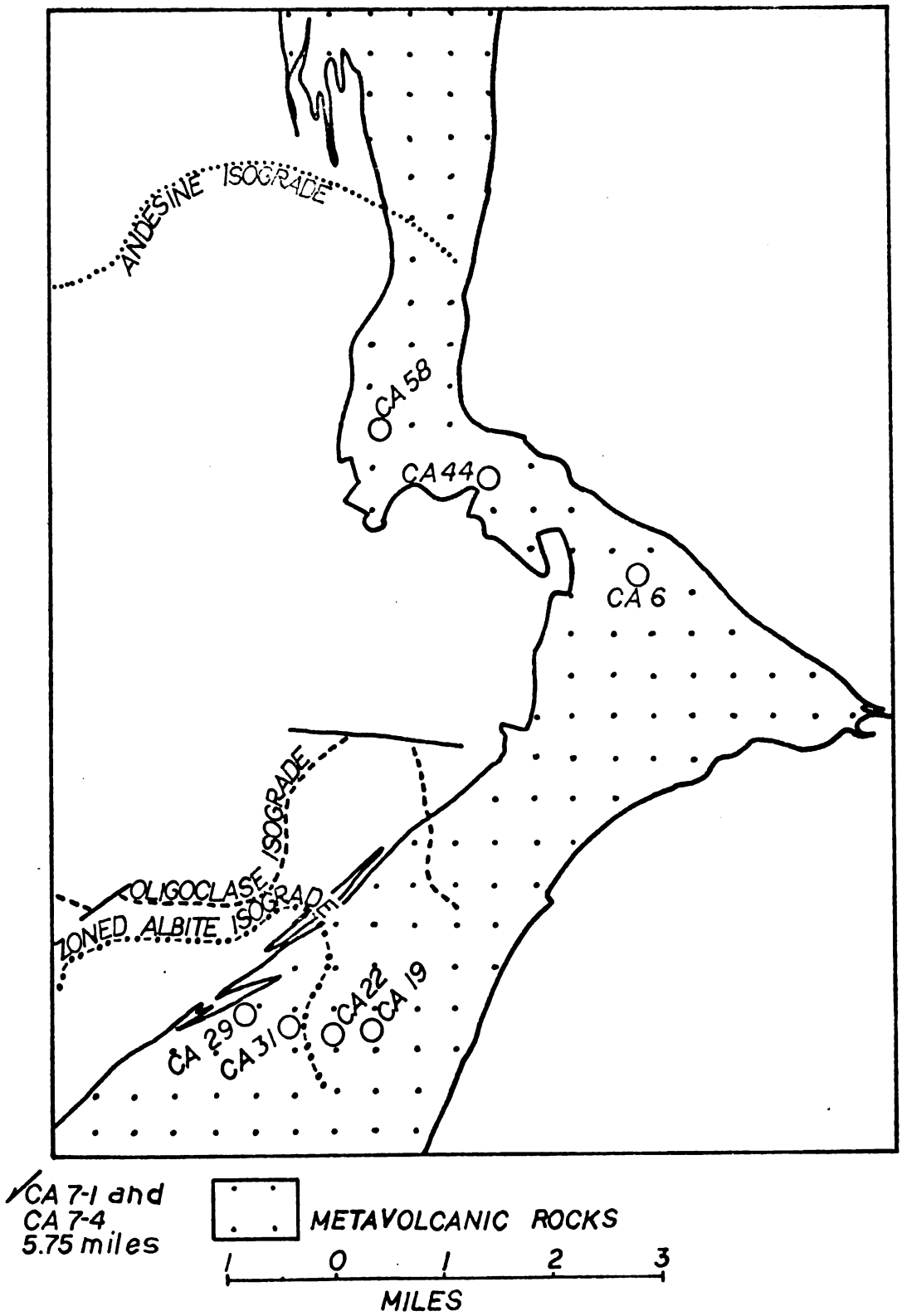

Figure 1. Metavolcanic zone in Cashel Township, Ontario, Canada.

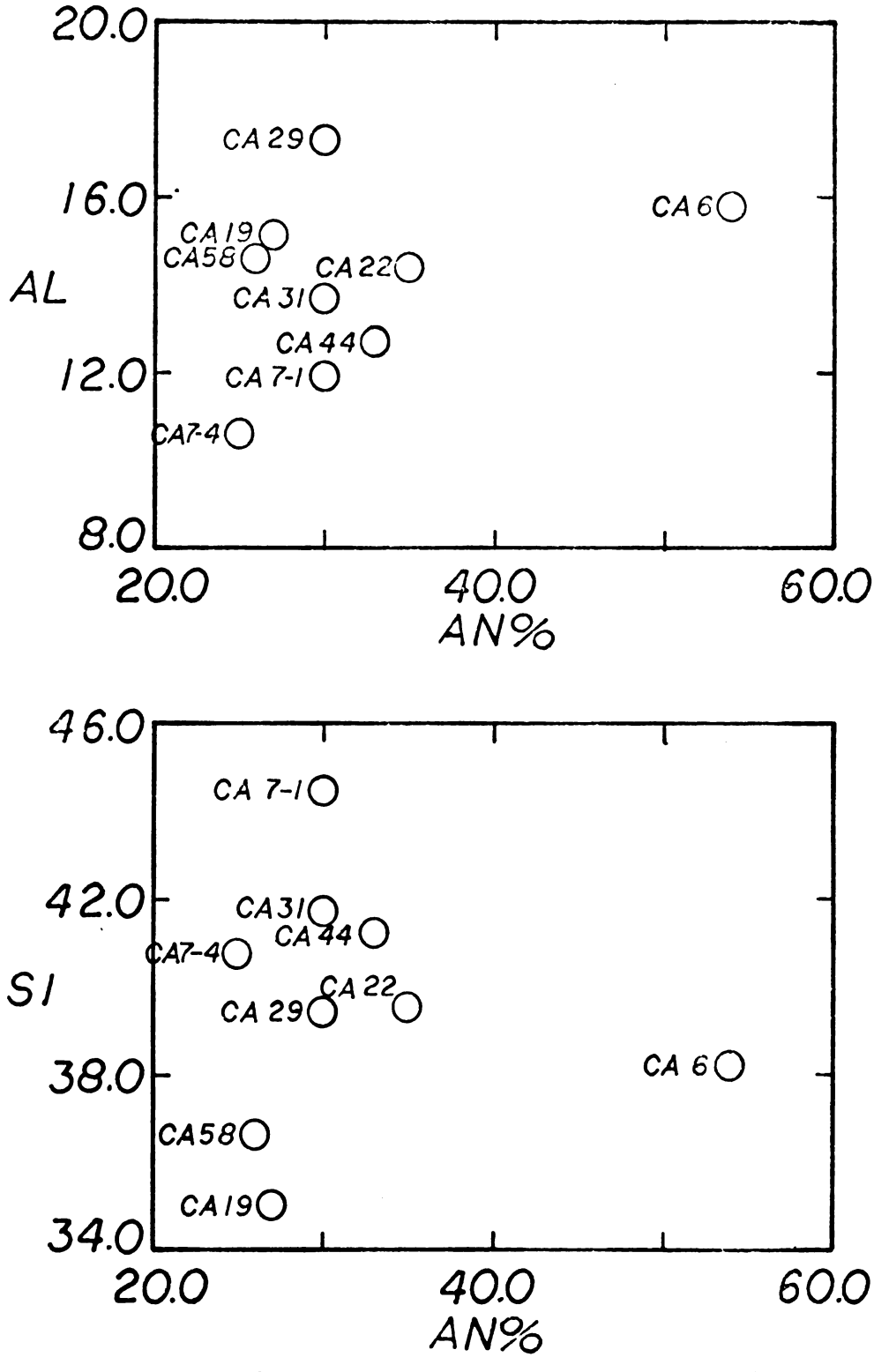

Figure 2. Graphs of Si and Al in terms of weight percent vs. plagioclase composition in terms of anorthite percent.

compositional variation that does exist might be the significant control on amphibole components. From the chemical data, the following relationships are evident:

1. As amphibole Mg increased, whole rock Mg increased (Figure 3);
2. As amphibole Si increased, amphibole Mg increased (Figure 4);
3. As amphibole Mg increased, amphibole Al showed a general decrease (Figure 5);
4. As amphibole Mg increased, amphibole Fe decreased (Figure 6).

The plagioclase composition of the samples exhibits no systematic relationship to any other variable. The amphibole Al versus amphibole Mg plot suggests that two populations with Mg increasing with Al exist; however, this conclusion is not supported by the other diagrams associating Al and Mg with other elements.

The data show that all available Mg went to the amphibole which is necessitated by the fact that amphibole and plagioclase are the only phases present. As Mg, Fe and some of the Al occupy the octahedral sites in an amphibole, Mg's increase at the expense of the Al indicates that it must be preferentially accepted into that site. As total whole rock Mg increases in the bulk composition, it also increases in the amphibole. These amphiboles have a lower total charge in the octahedral site. This lower charge could be

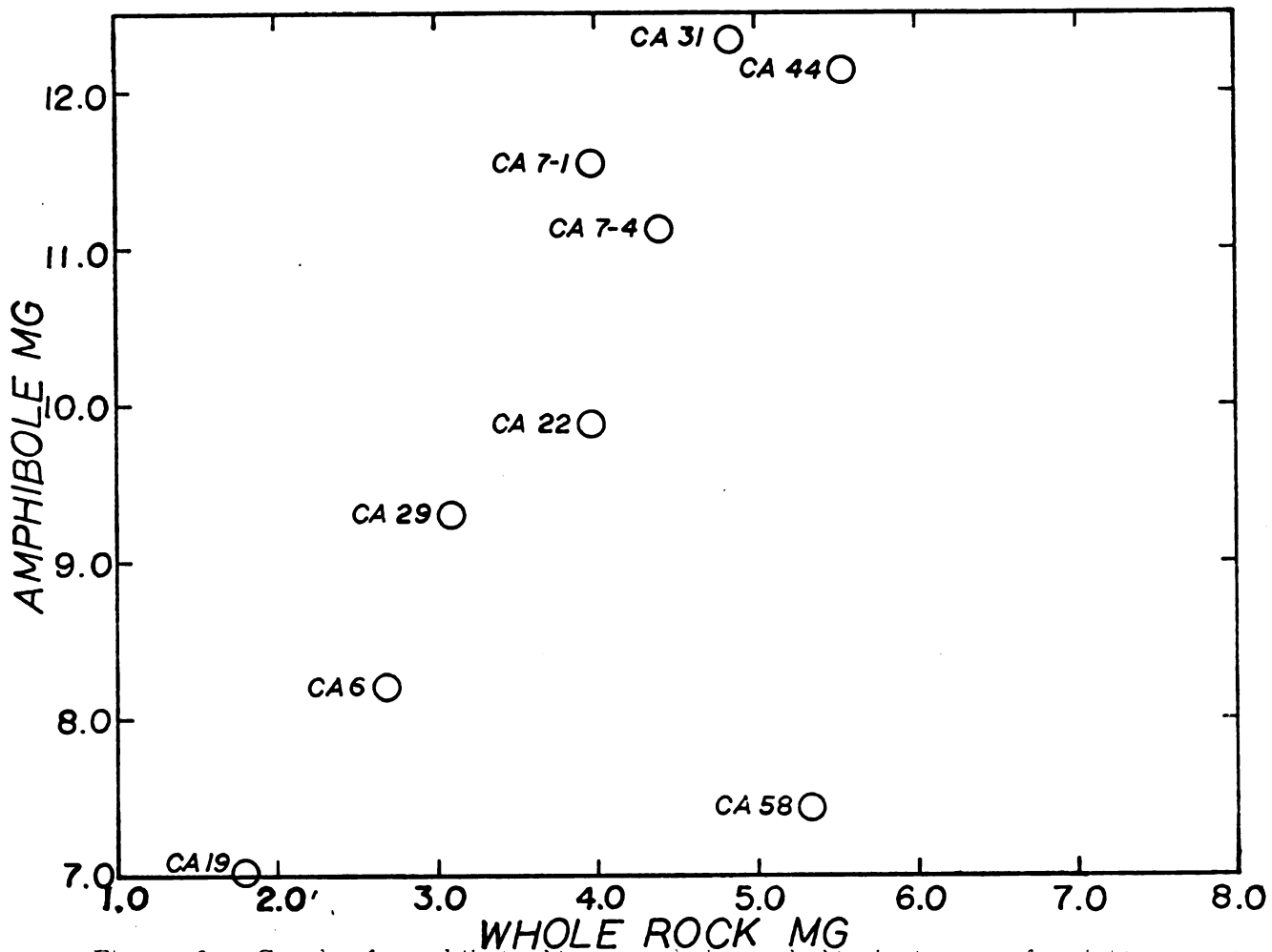

Figure 3. Graph of amphibole Mg vs. whole rock Mg in terms of weight percent.

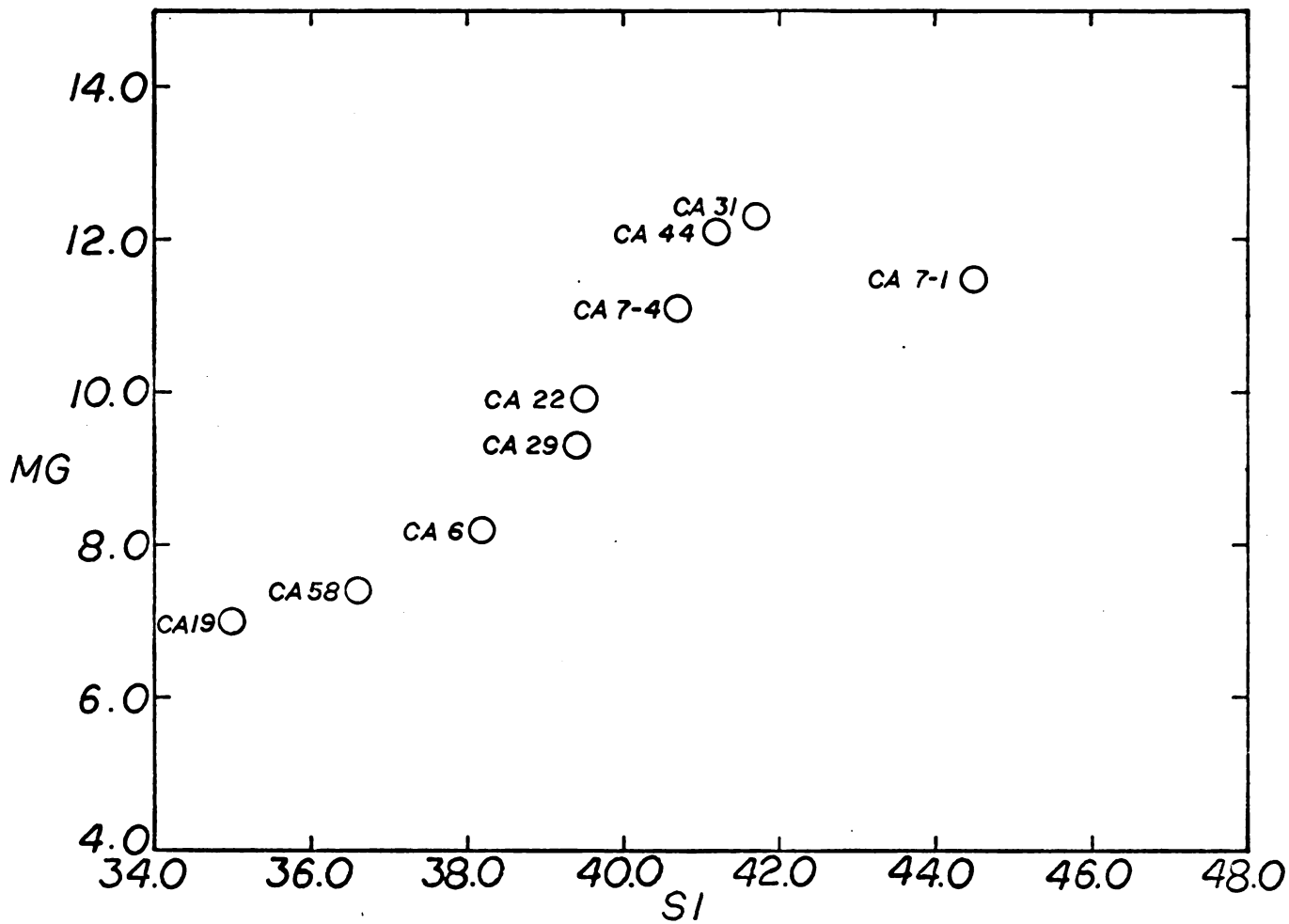

Figure 4. Graph of amphibole Mg vs. amphibole Si in terms of weight percent.

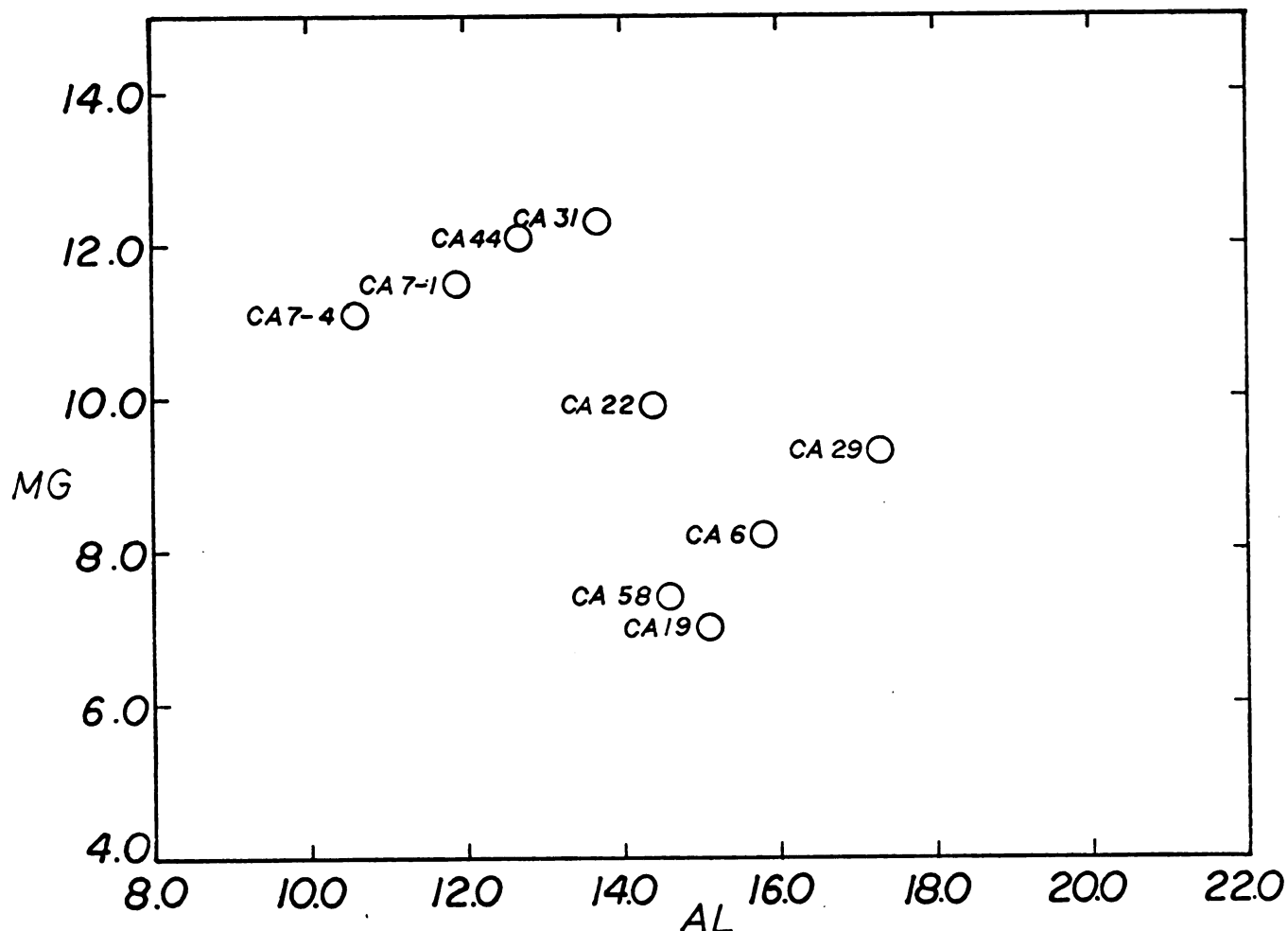

Figure 5. Graph of amphibole Mg vs. amphibole Al in terms of weight percent.

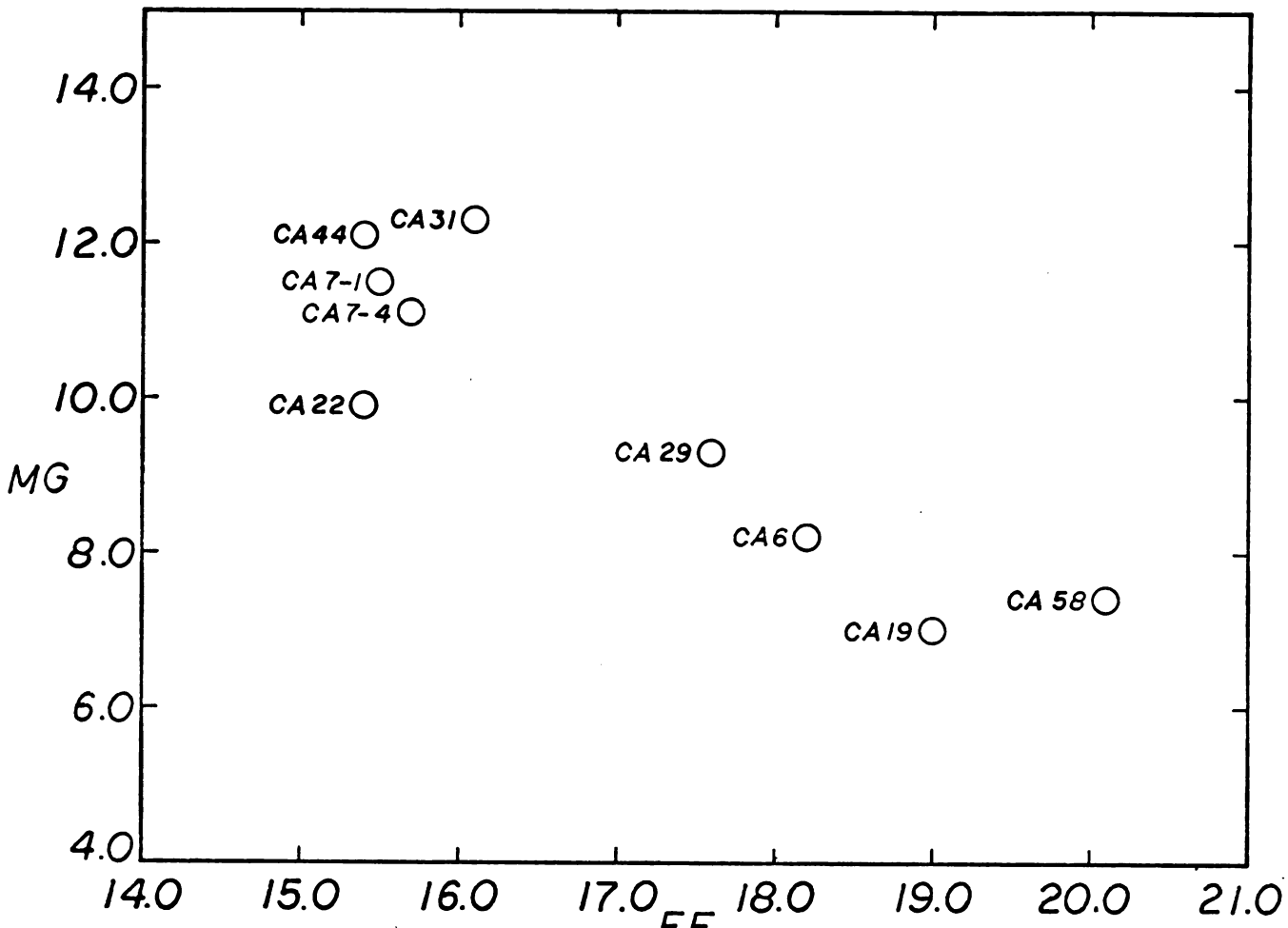

Figure 6. Graph of amphibole Mg vs. amphibole Fe in terms of weight percent.

balanced by an increase of Ca in the A sites or by an increase of Si in the tetrahedral sites. The increase in Si associated with the increase in Mg in the amphibole and the constantly high Ca levels demonstrate that the latter is the case. The closer spatial association of the tetrahedral sites and the octahedral sites and the twelve-fold coordination of the A sites compared with the six-fold coordination of the octahedral sites also suggest that a charge gain in the tetrahedral site could balance an octahedral charge loss with less structural strain than could the A site substitution.

CONCLUSION

The increase of Mg in amphibole in the amphibolite is a direct function of the bulk Mg content of the rock. This increase, associated with the decrease of Fe and Al, suggests that Mg is accepted into the octahedral site preferentially with respect to Fe and Al. That this increase in Mg is also associated with an increase in Si while Ca remains constant indicates that the tetrahedral sites are required to make up the octahedral site charge deficits associated with an increase of Mg. Therefore, the major conclusion of this paper is that the Si content of the tetrahedral site in amphiboles--the Al/Si ratio--is not directly associated with temperature and pressure conditions during cooling or during later metamorphic events. The amphibole composition of these plagioclase amphibole rocks is a direct function of their bulk rock composition. It seems

by pressure or temperature. It is also worthwhile to point out here the importance of using whole rock analyses along with individual mineral analyses, for without this type of analysis pairing, an erroneous conclusion could have been made.

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APPENDIX

```
      PROGRAM MPROBE                                                    A    1
C                                                                       A    2
C                                                                       A    3
C     THIS PROGRAM WILL ACCEPT RAW ARL MICROPROBE DATA AND OUTPUT A DECK A    4
C     SUITABLE FOR INPUT INTO PROGRAM FORMULA.  IT CAN HANDLE           A    5
C     UP TO FIFTEEN OF THE ELEMENTS LISTED IN DATA STATEMENT ELE AT A   A    6
C     TIME.                                                             A    7
C                                                                       A    8
C                                                                       A    9
      DIMENSION NELE(15), CRAT(15), MEL(35), NEN(15), BB(15), A(35,35)  A   10
      DIMENSION CODE(3), AX(35,35)                                      A   11
      DIMENSION CORC(15), C(15), B(15), W(35), FACT(35), ATMPER(15), WX( A   12
     135)                                                               A   13
      DIMENSION STD(15), ELE(35), XEL(15), CR(15), UZ(15), BN(15), R(15)A   14
     1, ID(8), FTR(35)                                                  A   15
      DATA ((ELE(L),L=1,35)=3H  C,3H  F,3H NA,3H MG,3H AL,3H SI,3H  P,3HA   16
     1  S,3H CL,3H  K,3H CA,3H TI,3H CR,3H MN,3H FE,3H CO,3H NI,3H CU,3HA   17
     2 ZN,3H RB,3H SR,3H  Y,3H ZR,3H BA,3H LA,3H CE,3H PR,3H ND,3H SM,3HA   18
     3 GD,3H DY,3H ER,3H HF,3H TH,3H  U)                                A   19
      DATA (((CODE(I),I=1,3)=6HCASE 1,6HCASE 2,6HCASE 3)                A   20
      DATA ((W(I),I=1,35)=1.15,5.73,1.67,1.29,1.17,1.08,1.06,1.03,1.10,1A   21
     1.06,1.03,1.05,1.11,1.15,1.14,1.16,1.14,1.19,1.19,1.41,1.34,1.26,1,A   22
     224,1.46,1.42,1.42,1.42,1.44,1.47,1.52,1.54,1.57,1.58,1.63,1.65)   A   23
      DATA ((WX(I),I=1,35)=1.18,8,97,1.98,1.44,1.24,1.12,1.09,1.04,1.12,A   24
     11.06,1.03,1.07,1.09,1.12,1.11,1.14,1.11,1.16,1.17,1.36,1.29,1.24,1A   25
     2.20,1.37,1.34,1.33,1.33,1.34,1.37,1.41,1.44,1.46,1.47,1.56,1.56)  A   26
      DATA ((FACT(I),I=1,35)=28.011,18,998,30,989,40,304,50.981,60.076,7A   27
     10.972,80.062,35,453,47,102,56,079,79,899,76,485,70,937,71,846,74.9A   28
     233,74,709,79,539,81,369,93,469,103,619,112.904,123.219,153,339,162A   29
     3.919,164.119,164,906,168,239,174,349,181,249,186,499,191,259,210.4A   30
     489,264.037,270.029)                                               A   31
      DATA (MFACT=9,008)                                                A   32
C                                                                       A   33
C                                                                       A   34
C     ARRAY FTR CONTAINS CORRECTION FACTORS FOR THE STANDARD USED,      A   35
C     ANY CHANGE IN THE STANDARD MUST BE ACCOMPANIED BY AN APPROPRIATE  A   36
C     CHANGE IN THE CORRECTION FACTOR,  1,0 FACTORS INDICATE A PURE     A   37
C     OXIDE STANDARD                                                    A   38
C                                                                       A   39
C                                                                       A   40
      DATA ((FTR(J),J=1,35)=1,0,1,0,0,1046,1.0,1.0,1.0,1.0,1.0,1.0,1.0,0,128A   41
     10,0.1803,1.0,1.0,1.0,1.0,0.7685,1.0,1.0,1.0,1.0,1.0,1.0,1.0,1.0,1.0,A   42
     20,1.0,1.0,1.0,1.0,1.0,1.0,1.0,1.0,1.0,1.0,1.0,1.0,1.0,1.0)        A   43
C                                                                       A   44
C                                                                       A   45
C                          DATA INPUT                                   A   46
C                                                                       A   47
C     FIRST CARD - NUMBER OF POINTS ANALYSED IN COLUMNS 1 THROUGH 4,    A   48
C     THE NUMBER OF ELEMENTS ANALYSED FOR AT EACH POINT IN COLUMNS 5    A   49
C     THROUGH 8, THE VOLTAGE AT WHICH THE MICROPROBE WAS RUN (EITHER 15 A   50
C     OR 20 KV) IN COLUMNS 9 THROUGH 12, THE CASE NAME (CASE 1, CASE 2, A   51
C     OR CASE 3, SEE EXPLANATION) IN COLUMNS 13 THROUGH 18, AND THE     A   52
C     ELEMENT SYMBOLS IN EACH 3 COLUMN GROUP FROM 19 THROUGH 63.  ALL   A   53
C     DATA MUST BE RIGHT JUSTIFIED.                                     A   54
C                                                                       A   55
C     SECOND CARD - ANY ALPHANUMERIC IDENTIFICATION IN COLUMNS 1 THROUGH A   56
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C     32 AND THE BACKGROUND VALUES FOR THE POINT ANALYSED IN FOUR          A   37
C     COLUMN GROUPINGS FROM COLUMN 33 ON.  A SECOND CARD MAY BE USED IF    A   38
C     NECESSARY.                                                           A   39
C                                                                          A   40
C                                                                          A   41
C     THIRD CARD - RAW COUNTS WITH ONE DECIMAL POINT IN EIGHT COLUMN       A   42
C     GROUPINGS FOR UP TO FIFTEEN VALUES.  A SECOND CARD MAY BE USED IF    A   43
C     NECESSARY.                                                           A   44
C                                                                          A   45
C     FOURTH CARD - CORRECTED STANDARD VALUES WITH ONE DECIMAL POINT IN    A   46
C     EIGHT COLUMN GROUPINGS FOR UP TO FIFTEEN VALUES.  A SECOND CARD      A   47
C     MAY BE USED IF NECESSARY.                                            A   48
C                                                                          A   49
C     FIFTH CARD - H2O OR OH WEIGHT PER CENT IN FIRST EIGHT COLUMNS WITH   A   50
C     FOUR DECIMAL PLACES IF KNOWN (I.E. CARD FIVE ONLY IN CASE TWO).      A   51
C                                                                          A   52
C     CARDS 2, 3, 4, AND 5 (IF INCLUDED) ARE REPEATED FOR EACH POINT       A   53
C     ANALYSED.                                                            A   54
C                                                                          A   55
C                                                                          A   56
      READ 65, ((A(I,J),J=1,35),I=1,35)                                    A   57
      READ 65, ((AX(I,J),J=1,35),I=1,35)                                   A   58
      PRINT 51                                                             A   59
      READ 67, MPT,NE,KV,CASE,(XEL(L),L=1,NE)                              A   60
      NUM=0                                                                A   61
      DO 9 JTR=1,MPT                                                       A   62
      IF (NE-10) 1,1,2                                                     A   63
    1 READ 69, (ID(I),I=1,8),(BN(K),K=1,NE)                                A   64
      READ 68, (CR(I),I=1,NE)                                              A   65
      READ 68, (STD(I),I=1,NE)                                             A   66
      GO TO 3                                                              A   67
    2 READ 71, (ID(I),I=1,8),(BN(K),K=1,NE)                                A   68
      READ 71, (CR(I),I=1,NE)                                              A   69
      READ 71, (STD(I),I=1,NE)                                             A   70
    3 DO 4 N=1,NE                                                          A   71
    4 CR(N)=CR(N)-BN(N)                                                    A   72
      DO 6 JK=1,NE                                                         A   73
      DO 5 JL=1,35                                                         A   74
      IF (XEL(JK)-ELE(JL)) 5,6,5                                           A   75
    5 CONTINUE                                                             A   76
    6 JZ(JK)=JL                                                            A   77
      DO 7 JM=1,NE                                                         A   78
      LR=JZ(JM)                                                            A   79
    7 P(JM)=(CR(JM)/STD(JM))*FTR(LR)                                       A   80
      DO 8 I=1,3                                                           A   81
      IF (CASE-CODE(I)) 8,10,8                                             A   82
    8 CONTINUE                                                             A   83
    9 CONTINUE                                                             A   84
      IF (NUM-MPT) 10,50,50                                                A   85
   10 ISWITCH=1                                                            A   86
      IF (ISWITCH-4) 11,50,11                                              A   87
   11 TOT=0.0                                                              A   88
      PRINT 52, CASE                                                       A   89
      NUM=NUM+1                                                            A   90
      TOTWT=0.0                                                            A   91
      WTP=0.0                                                              A   92
```

```
      DO 12 I=1,NE                                                    A  113
      BB(I)=0.0                                                       A  114
   12 C(I)=0.0                                                        A  115
      DO 15 J=1,NE                                                    A  116
      CRAT(J)=R(J)                                                    A  117
      CONC(J)=CRAT(J)                                                 A  118
      DO 13 K=1,35                                                    A  119
      IF (XEL(J)-ELE(K)) 13,14,13                                     A  120
   13 CONTINUE                                                        A  121
   14 TOTWT=TOTWT+CRAT(J)                                             A  122
   15 NEM(J)=K                                                        A  123
      PRINT 55, (ID(I),I=1,6)                                         A  124
      PRINT 56, (XEL(I),I=1,NE)                                       A  125
      PRINT 57, (CRAT(I),I=1,NE)                                      A  126
      IF (ISWITCH-3) 16,36,16                                         A  127
   16 IF (ISWITCH-2) 18,17,18                                         A  128
   17 READ 53, WTR                                                    A  129
      PRINT 59, WTR                                                   A  130
      WATPER=WTR/WFACT                                                A  131
      PRINT 64, WATPER                                                A  132
      TOTWT=TOTWT+WTR                                                 A  133
   18 N=0                                                             A  134
   19 DO 26 M=1,NE                                                    A  135
      I=NEM(M)                                                        A  136
      DO 22 K=1,NE                                                    A  137
      J=NEM(K)                                                        A  138
      IF (KV-15) 20,20,21                                             A  139
   20 BB(M)=BB(M)+A(I,J)*CONC(K)                                      A  140
      GO TO 22                                                        A  141
   21 BB(M)=BB(M)+AX(I,J)*CONC(K)                                     A  142
   22 CONTINUE                                                        A  143
      IF (KV-15) 23,23,24                                             A  144
   23 BB(M)=BB(M)+W(I)*WTR                                            A  145
      GO TO 25                                                        A  146
   24 BB(M)=BB(M)+WX(I)*WTR                                           A  147
   25 B(M)=1./TOTWT*BB(M)                                             A  148
   26 CONTINUE                                                        A  149
      DO 27 M=1,NE                                                    A  150
      CONC(M)=CRAT(M)*B(M)                                            A  151
      TOT=TOT+CONC(M)                                                 A  152
      BB(M)=0.0                                                       A  153
   27 CONTINUE                                                        A  154
      PRINT 60, (CONC(I),I=1,NE)                                      A  155
      I=1                                                             A  156
   28 IF (ABSF(CONC(I)-C(I))-.0001) 29,30,30                          A  157
   29 I=I+1                                                           A  158
      IF (I-(NE+1)) 28,33,33                                          A  159
   30 TOT=TOT+WTR                                                     A  160
      TOTWT=TOT                                                       A  161
      TOT=0.0                                                         A  162
      DO 31 I=1,NE                                                    A  163
   31 C(I)=CONC(I)                                                    A  164
      N=N+1                                                           A  165
      IF (N-10) 19,32,32                                              A  166
   32 PRINT 61                                                        A  167
      GO TO 9                                                         A  168
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```
      33 SUM=0.0                                                          A  169
         DO 34 M=1,NE                                                     A  170
      34 SUM=SUM+CONC(M)                                                  A  171
         SUM=SUM+WTR                                                      A  172
         PUNCH 66, (ID(I),I=1,8)                                          A  173
         PUNCH 64, (CONC(KLA),KLA=1,NE),WTR                               A  174
         PRINT 58, SUM                                                    A  175
         DO 35 M=1,NE                                                     A  176
         I=IEM(M)                                                         A  177
      35 ATMPER(M)=(CONC(M)/FACT(I))*1000.0                               A  178
         PRINT 63, (ATMPER(I),I=1,NE)                                     A  179
         GO TO 9                                                          A  180
      36 N=1                                                              A  181
      37 DO 44 M=1,NE                                                     A  182
         I=IEM(M)                                                         A  183
         DO 44 K=1,NE                                                     A  184
         J=IEM(K)                                                         A  185
         IF (KV-15) 39,38,39                                              A  186
      38 RB(M)=RB(M)+A(I,J)*CONC(K)                                       A  187
         GO TO 40                                                         A  188
      39 RB(M)=RB(M)+AX(I,J)*CONC(K)                                      A  189
      40 CONTINUE                                                         A  190
         IF (KV-15) 41,41,42                                              A  191
      41 RB(M)=RB(M)+W(I)*WTR                                             A  192
         GO TO 43                                                         A  193
      42 RB(M)=RB(M)+WX(I)*WTR                                            A  194
      43 B(M)=1./TOTWT*RB(M)                                              A  195
      44 CONTINUE                                                         A  196
         DO 45 M=1,NE                                                     A  197
         CONC(M)=CRAT(M)*B(M)                                             A  198
         TOT=TOT+CONC(M)                                                  A  199
         RB(M)=0.0                                                        A  200
      45 CONTINUE                                                         A  201
         PRINT 60, (CONC(I),I=1,NE)                                       A  202
         IF (N-4) 46,47,30                                                A  203
      46 N=N+1                                                            A  204
         TOTWT=TOT                                                        A  205
         TOT=0.0                                                          A  206
         GO TO 37                                                         A  207
      47 IF (TOT-1.00) 48,49,49                                           A  208
      48 WTR=1.00-TOT                                                     A  209
         PRINT 59, WTR                                                    A  210
         WATPER=WTR/WFACT                                                 A  211
         PRINT 64, WATPER                                                 A  212
         N=N+1                                                            A  213
         TOTWT=1.00                                                       A  214
         TOT=0                                                            A  215
         GO TO 37                                                         A  216
      49 PRINT 59, WTR                                                    A  217
         GO TO 30                                                         A  218
      50 PRINT 62, NUM                                                    A  219
C                                                                         A  220
      51 FORMAT (1H1,7X,58H MICROPROBE ANALYSES OF MINERALS WITH UNKNOWN CO A  221
        1MPOSITIONS//)                                                    A  222
      52 FORMAT (1H0,9X,A6)                                               A  223
      53 FORMAT (10F8.4)                                                  A  224
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   54 FORMAT (10F8.4/5F8.4)                                                A 225
   55 FORMAT (8X,19A4,/)                                                   A 226
   56 FORMAT (2X,8HELEMENTS,5X,15(A5,3X))                                  A 227
   57 FORMAT (2X,10HCOUNTRATIO,2X,15(F8.4))                                A 228
   58 FORMAT (2X,3HSUM,1X,F8.4)                                            A 229
   59 FORMAT (2X,5HWATER,7X,F8.4)                                          A 230
   60 FORMAT (2X,8HCONCENT.,4X,15(F8.4))                                   A 231
   61 FORMAT (17X,34H THE ABOVE VALUES FAIL TO CONVERGE)                   A 232
   62 FORMAT (1H0,9X,33HTOTAL NUMBER OF MINERALS ANALYSED,I4)              A 233
   63 FORMAT (2X,12HREL.AT.PERC./3X,6HX 1000,5X,15(F8.4))                  A 234
   64 FORMAT (2X,12HWTR.AT.PERC.,F8.4)                                     A 235
   65 FORMAT (87(14F5.2/),7F5.2)                                           A 236
   66 FORMAT (8A4)                                                         A 237
   67 FORMAT (3I4,A6,15A3)                                                 A 238
   68 FORMAT (10F8.1)                                                      A 239
   69 FORMAT (8A4,10F4.2)                                                  A 240
   70 FORMAT (8A4,10F4.2/5F4.2)                                            A 241
   71 FORMAT (10F8.1/5F8.1)                                                A 242
      END                                                                  A 243
```

1.00	5.19	1.51	1.18	1.09	1.03	1.02	1.00	1.07	1.04	1.01	1.06	1.09	1.13	
1.14	1.14	1.19	1.17	1.18	1.34	1.27	1.23	1.19	1.42	1.39	1.39	1.39	1.41	
1.44	1.49	1.51	1.54	1.55	1.57	1.58	1.43	1.00	1.89	1.40	1.23	1.11	1.07	
1.02	1.08	1.03	1.00	1.05	1.07	1.10	1.10	1.12	1.09	1.14	1.15	1.41	1.33	
1.27	1.23	1.39	1.36	1.36	1.35	1.37	1.41	1.45	1.48	1.50	1.51	1.56	1.57	
2.84	2.29	1.01	1.05	1.51	1.31	1.21	1.13	1.17	1.09	1.05	1.09	1.10	1.14	
1.15	1.15	1.13	1.18	1.18	1.60	1.51	1.42	1.36	1.42	1.40	1.40	1.39	1.41	
1.44	1.49	1.51	1.53	1.54	1.60	1.61	3.93	3.12	1.18	1.00	1.62	1.39	1.27	
1.18	1.21	1.12	1.08	1.11	1.13	1.16	1.15	1.18	1.15	1.20	1.21	1.69	1.59	
1.43	1.42	1.47	1.44	1.43	1.43	1.45	1.48	1.52	1.55	1.57	1.58	1.67	1.67	
4.93	3.66	1.25	1.03	1.00	1.43	1.29	1.19	1.21	1.11	1.06	1.10	1.11	1.14	
1.13	1.15	1.13	1.18	1.18	1.74	1.63	1.51	1.44	1.42	1.40	1.39	1.39	1.41	
1.44	1.48	1.51	1.53	1.54	1.61	1.60	5.75	4.11	1.35	1.09	1.04	1.00	1.34	
1.22	1.24	1.13	1.08	1.11	1.12	1.16	1.15	1.17	1.14	1.19	1.20	1.30	1.24	
1.55	1.49	1.45	1.43	1.42	1.42	1.44	1.47	1.51	1.53	1.56	1.56	1.66	1.66	
7.49	4.51	1.39	1.11	1.05	0.99	1.00	1.25	1.25	1.13	1.08	1.10	1.11	1.14	
1.13	1.15	1.12	1.17	1.18	1.27	1.21	1.18	1.15	1.42	1.39	1.39	1.38	1.40	
1.45	1.47	1.51	1.52	1.52	1.61	1.60	8.60	4.85	1.46	1.16	1.08	1.02	1.01	
1.01	1.28	1.15	1.09	1.11	1.12	1.15	1.14	1.16	1.13	1.18	1.19	1.32	1.25	
1.21	1.17	1.45	1.42	1.41	1.41	1.43	1.46	1.50	1.52	1.54	1.55	1.67	1.66	
31.46	3.26	1.19	0.98	0.95	0.91	0.92	0.90	1.00	1.26	1.16	1.13	1.10	1.12	
1.10	1.11	1.08	1.13	1.13	1.18	1.13	1.09	1.07	1.43	1.39	1.38	1.36	1.37	
1.40	1.42	1.44	1.46	1.46	1.77	1.74	3.24	4.87	1.48	1.15	1.07	1.00	0.98	
0.95	1.01	1.00	1.20	1.15	1.12	1.14	1.12	1.13	1.10	1.14	1.14	1.26	1.20	
1.16	1.12	1.45	1.41	1.40	1.38	1.39	1.41	1.44	1.46	1.47	1.47	1.43	1.44	
3.10	5.75	1.65	1.26	1.14	1.05	1.03	0.99	1.05	0.99	1.00	1.17	1.14	1.16	
1.19	1.15	1.19	1.16	1.17	1.33	1.26	1.21	1.18	1.49	1.45	1.43	1.42	1.43	
1.45	1.48	1.49	1.51	1.51	1.50	1.51	3.12	7.20	1.83	1.34	1.18	1.06	1.03	
0.93	1.02	0.96	0.92	1.00	1.09	1.11	1.09	1.10	1.07	1.11	1.11	1.32	1.25	
1.19	1.15	1.29	1.27	1.27	1.35	1.36	1.38	1.40	1.42	1.43	1.43	1.41	1.42	
4.18	8.70	2.14	1.50	1.28	1.12	1.06	1.00	1.03	0.96	0.91	0.90	1.00	1.03	
1.08	1.09	1.05	1.09	1.09	1.38	1.30	1.23	1.18	1.21	1.23	1.23	1.23	1.25	
1.28	1.39	1.43	1.41	1.40	1.37	1.37	5.25	9.74	2.39	1.63	1.37	1.17	1.09	
1.00	1.03	0.94	0.90	0.89	0.94	1.00	0.99	1.07	1.03	1.07	1.06	1.40	1.32	
1.24	1.19	1.19	1.16	1.16	1.18	1.20	1.24	1.28	1.30	1.37	1.37	1.35	1.35	
6.07	2.32	2.62	1.76	1.46	1.23	1.13	1.04	1.05	0.96	0.92	0.91	0.88	0.98	
1.04	1.02	1.04	1.08	1.07	1.47	1.38	1.29	1.23	1.21	1.18	1.18	1.17	1.18	
1.25	1.29	1.32	1.34	1.38	1.38	1.38	6.96	2.34	2.86	1.89	1.53	1.27	1.16	
1.05	1.05	0.96	0.91	0.90	0.87	0.86	0.94	1.00	0.98	1.06	1.05	1.51	1.41	
1.31	1.25	1.19	1.16	1.15	1.14	1.14	1.18	1.25	1.29	1.31	1.36	1.35	1.35	
8.17	2.50	3.18	2.47	1.65	1.35	1.22	1.10	1.10	0.99	0.94	0.94	0.91	0.90	
0.84	0.97	1.00	1.04	1.08	1.60	1.49	1.38	1.31	1.23	1.20	1.19	1.18	1.19	
1.24	1.24	1.31	1.34	1.35	1.41	1.40	9.50	2.51	1.98	2.18	1.72	1.38	1.23	
1.09	1.08	0.96	0.91	0.91	0.89	0.88	0.83	0.80	0.90	1.00	1.00	1.62	1.50	
1.31	1.31	1.18	1.15	1.14	1.14	1.15	1.16	1.19	1.20	1.27	1.29	1.35	1.34	
11.11	2.62	2.45	2.36	1.83	1.45	1.28	1.12	1.10	0.97	0.92	0.92	0.90	0.90	
0.85	0.83	0.75	0.93	1.00	1.69	1.57	1.44	1.35	1.18	1.15	1.15	1.14	1.15	
1.17	1.19	1.20	1.20	1.27	1.36	1.35	21.82	3.51	1.20	0.93	0.88	0.82	1.57	
1.39	1.25	1.03	0.94	0.92	0.90	0.91	0.90	0.91	0.89	0.92	0.92	1.00	1.60	
1.74	1.60	1.13	1.09	1.08	1.08	1.08	1.10	1.12	1.13	1.15	1.16	1.37	1.33	
18.14	4.06	1.32	1.00	0.94	0.86	1.65	1.46	1.30	1.06	0.97	0.95	0.92	0.94	
0.92	0.93	0.91	0.94	0.94	1.04	1.00	0.96	1.65	1.16	1.12	1.11	1.10	1.11	
1.12	1.15	1.14	1.17	1.18	1.41	1.37	14.66	4.45	1.38	1.05	0.97	0.89	0.88	
1.35	1.33	1.08	0.99	0.96	0.94	0.96	0.94	0.95	0.93	0.96	0.97	1.09	1.04	
1.02	0.97	1.19	1.15	1.15	1.13	1.14	1.16	1.18	1.20	1.21	1.22	1.46	1.42	
16.34	4.96	1.48	1.11	1.02	0.92	0.91	1.41	1.37	1.11	1.02	0.98	0.96	0.97	
0.96	0.97	0.94	0.98	0.98	1.12	1.07	1.03	1.00	1.21	1.18	1.17	1.16	1.16	
1.18	1.20	1.22	1.23	1.24	1.49	1.45	7.38	2.12	1.95	1.73	1.61	1.26	1.15	
0.99	0.95	0.84	0.78	0.82	0.90	0.93	0.91	0.90	0.87	0.89	0.89	1.48	1.36	
1.27	1.18	1.01	0.98	0.97	0.97	0.98	1.11	1.13	1.12	1.12	1.11	1.11	1.10	
7.07	2.27	1.96	1.74	1.59	1.26	1.14	0.99	0.97	0.85	0.79	0.80	0.84	0.94	

0.92 0.92 0.89 0.91 0.90 1.53 1.40 1.29 1.21 1.03 1.00 1.00 0.99 1.00
1.09 1.13 1.15 1.15 1.14 1.14 1.13 7.44 2.32 2.10 1.58 1.65 1.30 1.17
1.01 0.98 0.85 0.80 0.81 0.84 0.91 0.91 0.92 0.88 0.91 0.90 1.58 1.44
1.33 1.24 1.01 1.00 1.00 0.99 1.01 1.02 1.11 1.14 1.15 1.14 1.16 1.14
7.84 2.38 2.13 1.64 1.55 1.34 1.20 1.04 1.00 0.87 0.81 0.78 0.84 0.86
0.89 0.93 0.89 0.81 0.91 1.63 1.48 1.36 1.27 1.02 0.99 1.01 1.00 1.01
1.03 1.11 1.15 1.16 1.15 1.17 1.15 8.21 2.40 2.26 1.52 1.59 1.37 1.22
1.04 1.00 0.87 0.81 0.78 0.83 0.85 0.88 0.91 0.88 0.90 0.90 1.66 1.51
1.32 1.28 1.01 0.98 1.00 0.99 1.00 1.02 1.04 1.11 1.13 1.13 1.17 1.15
8.97 2.46 1.05 1.60 1.48 1.43 1.27 1.07 1.01 0.87 0.81 0.78 0.78 0.83
0.82 0.87 0.86 0.89 0.88 1.59 1.57 1.44 1.32 0.97 0.95 0.96 0.96 0.98
1.00 1.02 1.04 1.09 1.12 1.17 1.14 9.98 2.53 1.01 1.69 1.40 1.36 1.31
1.09 1.02 0.87 0.80 0.77 0.72 0.77 0.80 0.81 0.82 0.86 0.86 1.65 1.49
1.49 1.36 0.95 0.93 0.93 0.92 0.95 0.98 1.00 1.01 1.03 1.07 1.16 1.14
10.82 2.60 1.03 0.83 1.47 1.26 1.25 1.12 1.04 0.88 0.80 0.77 0.73 0.77
0.7 0.80 0.78 0.84 0.84 1.54 1.40 1.42 1.30 0.96 0.93 0.92 0.90 0.91
0.95 0.96 1.01 1.01 1.05 1.17 1.14 11.68 2.68 1.04 0.84 1.55 1.21 1.30
1.16 1.06 0.88 0.81 0.78 0.73 0.72 0.75 0.74 0.77 0.80 0.82 1.48 1.34
1.34 1.35 0.95 0.93 0.91 0.90 0.90 0.91 0.95 0.96 1.00 1.01 1.18 1.15
14.99 3.05 1.14 0.89 0.85 1.36 1.21 1.08 1.13 0.93 0.84 0.80 0.75 0.74
0.71 0.68 0.72 0.72 0.79 1.10 1.46 1.35 1.24 0.97 0.94 0.93 0.91 0.91
0.92 0.93 0.93 0.97 1.00 1.22 1.18 35.33 4.84 4.65 1.20 1.12 0.96 0.94
0.85 0.85 0.85 0.94 0.91 0.91 0.88 0.85 0.85 0.82 0.84 0.84 1.15 1.07
1.03 0.98 1.08 1.05 1.07 1.05 1.07 1.05 1.05 1.05 1.05 1.04 1.00 0.99
39.59 5.17 1.73 1.25 1.16 0.98 0.96 0.86 0.85 0.80 0.82 0.92 0.90 0.89
0.86 0.85 0.82 0.84 0.84 1.18 1.10 1.05 1.00 1.10 1.07 1.05 1.03 1.05
1.07 1.06 1.05 1.05 1.04 1.01 1.00

1.00	8.26	1.76	1.29	1.14	1.05	1.03	1.00	1.08	1.04	1.01	1.05	1.07	1.11
1.09	1.12	1.09	1.14	1.15	1.28	1.21	1.18	1.14	1.33	1.31	1.30	1.30	1.31
1.34	1.38	1.41	1.43	1.44	1.49	1.50	1.55	1.00	2.32	1.62	1.35	1.18	1.12
1.05	1.11	1.04	1.00	1.04	1.05	1.08	1.07	1.09	1.07	1.12	1.12	1.38	1.31
1.25	1.21	1.31	1.28	1.27	1.27	1.28	1.31	1.35	1.37	1.39	1.41	1.49	1.49
3.45	3.02	1.01	2.27	1.75	1.46	1.31	1.20	1.23	1.10	1.05	1.08	1.08	1.11
1.10	1.12	1.09	1.14	1.14	1.64	1.54	1.44	1.38	1.33	1.31	1.30	1.29	1.31
1.33	1.37	1.39	1.41	1.42	1.53	1.52	4.92	4.43	1.26	1.00	1.92	1.58	1.40
1.27	1.29	1.15	1.09	1.11	1.12	1.14	1.13	1.15	1.12	1.17	1.17	1.76	1.65
1.53	1.46	1.39	1.36	1.35	1.34	1.36	1.38	1.42	1.44	1.46	1.47	1.61	1.60
6.47	5.48	1.38	1.06	1.00	1.67	1.46	1.30	1.31	1.15	1.09	1.11	1.10	1.13
1.12	1.14	1.11	1.15	1.16	1.86	1.73	1.59	1.51	1.35	1.33	1.32	1.31	1.33
1.35	1.38	1.41	1.43	1.43	1.57	1.55	7.62	6.23	1.51	1.15	1.05	1.00	1.53
1.36	1.36	1.18	1.12	1.13	1.12	1.15	1.13	1.15	1.12	1.17	1.17	1.21	1.16
1.66	1.58	1.39	1.36	1.35	1.34	1.36	1.38	1.42	1.44	1.46	1.46	1.63	1.61
10.32	7.05	1.59	1.18	1.07	1.00	1.00	1.40	1.39	1.18	1.12	1.12	1.11	1.13
1.11	1.13	1.11	1.15	1.15	1.19	1.14	1.11	1.09	1.35	1.33	1.32	1.31	1.32
1.34	1.37	1.40	1.41	1.42	1.59	1.56	11.89	7.61	1.69	1.24	1.11	1.03	1.02
1.09	1.43	1.21	1.14	1.13	1.12	1.14	1.13	1.14	1.11	1.16	1.16	1.24	1.18
1.15	1.12	1.38	1.35	1.35	1.33	1.35	1.37	1.40	1.42	1.44	1.45	1.65	1.62
47.09	4.68	1.31	1.00	0.94	0.90	0.91	0.89	1.00	1.44	1.29	1.19	1.13	1.33
1.10	1.11	1.09	1.11	1.11	1.07	1.03	1.01	0.99	1.41	1.36	1.34	1.32	1.33
1.33	1.35	1.35	1.37	1.37	1.86	1.79	3.99	7.62	1.73	1.25	1.12	1.02	1.00
0.96	1.03	1.00	1.35	1.23	1.16	1.16	1.13	1.13	1.09	1.13	1.13	1.19	1.14
1.10	1.07	1.43	1.40	1.38	1.35	1.35	1.36	1.37	1.38	1.39	1.39	1.34	1.34
3.78	9.20	1.97	1.40	1.22	1.10	1.06	1.01	1.08	1.00	1.00	1.26	1.18	1.18
1.15	1.16	1.12	1.16	1.15	1.28	1.22	1.17	1.14	1.49	1.43	1.41	1.39	1.39
1.39	1.41	1.42	1.43	1.43	1.41	1.42	3.88	12.17	2.29	1.56	1.31	1.15	1.08
1.02	1.07	0.98	0.93	1.00	1.14	1.14	1.11	1.11	1.07	1.11	1.10	1.31	1.24
1.18	1.14	1.21	1.19	1.19	1.33	1.33	1.33	1.35	1.35	1.36	1.36	1.34	1.34
5.46	15.24	2.81	1.32	1.48	1.25	1.16	1.07	1.10	0.99	0.94	0.92	1.00	1.03
1.13	1.12	1.09	1.11	1.10	1.42	1.33	1.25	1.20	1.16	1.16	1.16	1.16	1.17
1.20	1.36	1.35	1.36	1.34	1.32	1.31	6.97	17.22	3.18	2.03	1.62	1.34	1.21
1.10	1.11	0.99	0.93	0.91	0.95	1.00	0.99	1.11	1.06	1.09	1.08	1.47	1.38
1.29	1.23	1.14	1.11	1.11	1.12	1.14	1.17	1.20	1.22	1.34	1.32	1.32	1.31
8.19	3.21	3.55	2.24	1.76	1.43	1.28	1.15	1.15	1.01	0.96	0.94	0.90	0.99
1.00	1.02	1.08	1.11	1.10	1.56	1.46	1.36	1.30	1.16	1.13	1.13	1.12	1.13
1.18	1.22	1.24	1.25	1.34	1.35	1.34	9.55	3.27	3.96	2.45	1.89	1.51	1.33
1.18	1.17	1.02	0.95	0.94	0.90	0.89	0.95	1.00	0.98	1.10	1.08	1.63	1.52
1.41	1.34	1.15	1.12	1.11	1.10	1.10	1.12	1.18	1.21	1.23	1.33	1.34	1.32
11.35	3.52	4.46	2.72	2.07	1.64	1.43	1.25	1.23	1.06	0.99	0.97	0.93	0.92
0.88	0.99	1.01	1.04	1.12	1.76	1.63	1.51	1.43	1.19	1.16	1.15	1.13	1.14
1.16	1.18	1.24	1.26	1.28	1.40	1.38	13.46	3.57	4.18	2.94	2.20	1.70	1.47
1.26	1.22	1.04	0.97	0.95	0.91	0.09	0.86	0.84	0.92	1.00	1.00	1.81	1.68
1.54	1.45	1.14	1.11	1.10	1.09	1.10	1.11	1.13	1.14	1.20	1.22	1.35	1.33
15.97	3.77	3.33	3.23	2.38	1.82	1.55	1.32	1.26	1.06	0.99	0.96	0.92	0.92
0.88	0.86	0.80	0.95	1.00	1.92	1.77	1.62	1.52	1.15	1.12	1.11	1.10	1.10
1.14	1.13	1.14	1.15	1.20	1.38	1.35	34.02	5.57	1.46	1.06	0.97	0.89	2.10
1.84	1.61	1.23	1.10	1.03	0.98	0.97	0.95	0.95	0.93	0.95	0.95	1.00	1.93
2.18	1.99	1.18	1.13	1.12	1.10	1.10	1.10	1.12	1.12	1.13	1.13	1.53	1.47
27.54	6.52	1.61	1.15	1.04	0.94	2.19	1.92	1.66	1.26	1.12	1.05	0.99	0.99
0.96	0.97	0.94	0.97	0.97	1.04	1.00	0.96	2.04	1.19	1.15	1.14	1.12	1.12
1.12	1.13	1.14	1.15	1.16	1.56	1.49	21.99	7.22	1.69	1.20	1.07	0.96	0.94
1.73	1.69	1.27	1.13	1.06	1.00	1.00	0.98	0.98	0.95	0.98	0.98	1.08	1.04
1.00	0.98	1.22	1.18	1.16	1.14	1.14	1.14	1.16	1.16	1.17	1.18	1.61	1.53
24.40	9.10	1.83	1.28	1.13	1.00	0.97	1.80	1.73	1.30	1.16	1.07	1.01	1.01
0.99	0.99	0.96	0.99	0.99	1.12	1.07	1.02	1.00	1.23	1.19	1.17	1.15	1.15
1.16	1.17	1.18	1.18	1.19	1.63	1.56	10.45	3.05	2.63	2.31	2.12	1.61	1.43
1.19	1.12	0.95	0.96	0.88	1.01	1.03	1.01	0.99	0.94	0.95	0.94	1.73	1.57
1.46	1.36	1.01	0.97	0.97	0.96	0.96	1.16	1.18	1.16	1.14	1.12	1.16	1.13
10.04	3.31	2.67	2.34	2.11	1.61	1.42	1.19	1.14	0.96	0.88	0.87	0.88	1.05

1.02 1.00 0.95 0.97 0.95 1.79 1.63 1.50 1.40 1.03 1.00 0.99 0.98 0.99
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