ECONOMIC GEOLOGY RESEARCH UNIT

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URANIUM AND THORIUM CONTENTS OF ARCHAEAN GRANITOIDS FROM THE BARBERTON MOUNTAIN LAND, SOUTH AFRICA

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*(Economic Geology Research Unit,
University of the Witwatersrand, Johannesburg)*

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ABSTRACT

One hundred and eighty-five samples of granitic rocks from the Barberton Mountain Land have been analysed for U and Th by instrumental epithermal neutron activation analysis. The samples, which cover the full range of granitic *(sensu lato)* composition and vary in age from 3,5 to 2,5 Ga. The granitoids have been subdivided into three categories, termed magmatic cycles. Each magmatic cycle is considered to have evolved progressively through time by reworking of pre-existing crustal material.

The U and Th contents increase progressively from the first magmatic cycle to the third magmatic cycle. The geometric mean U value for the entire sample population is 1,8 ppm (range : 0,13 to 19,2 ppm) and for Th is 8,8 ppm (range : 0,78 to 47,6 ppm), yielding a mean Th/U ratio of 4,8. Correction of the raw data for alpha-decay results in significantly different U and Th abundances in the granites at their time of emplacement, a consideration which has a marked effect on the Th/U ratio (corrected Th/U ratio = 3,5).

Comparison of U, Th, Zr, and Ce contents indicates that the mineralogical siting of U and Th may differ from one magmatic cycle to the other. Furthermore, there are indications that some granitic bodies are relatively enriched in U whereas others appear to have suffered U loss.

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I. INTRODUCTION

Although a considerable amount of geochemical information has now been accumulated from Archaean granitoid terranes world-wide, a comprehensive set of data on U and Th contents of these rocks is still lacking. By contrast, granitoids of younger age have been extensively studied for their radio-element distribution, principally for a better understanding of uranium and thorium geochemistry, but also for heat flow and isotopic studies and in the assessment of their economic potential. The Archaean granite-greenstone terrane of the Barberton Mountain Land has been the object of detailed investigation over the past 20 years and the geological evolution of the area is now reasonably well understood. However, one gap in the knowledge of the area, results from a relative paucity of high-quality trace element data, particularly in the granitic basement. In order to correct this imbalance a large suite of samples, representative of most of the major granitic (*sensu lato*) bodies in the region, has recently been collected and analyzed for 21 selected trace elements by neutron activation techniques. The purpose of this paper is to present the U and Th data and to discuss its significance in terms of the evolution of the granites in the area.

II. GEOLOGICAL SETTING

Mapping of the granitic terrane both to the north and the south of the Barberton greenstone belt has delineated a diverse suite of granitic rock types (Anhaeusser *et al.*, 1983). These granitic rocks have been broadly sub-divided into three categories termed "magmatic cycles" (Anhaeusser and Robb, 1981). The earliest cycle commenced approximately 3500 Ma ago and is characterized by leucocratic biotite trondhjemite and hornblende tonalite, as well as complex bimodal gneiss and migmatite. The tonalites and trondhjemites are pervasively foliated and their mode of emplacement is regarded as being diapiric. These rocks are commonly believed to have been derived by the partial melting of volcanic precursors similar to the metabasalts found in the lowermost successions of greenstone belts (Robb and Anhaeusser, 1983).

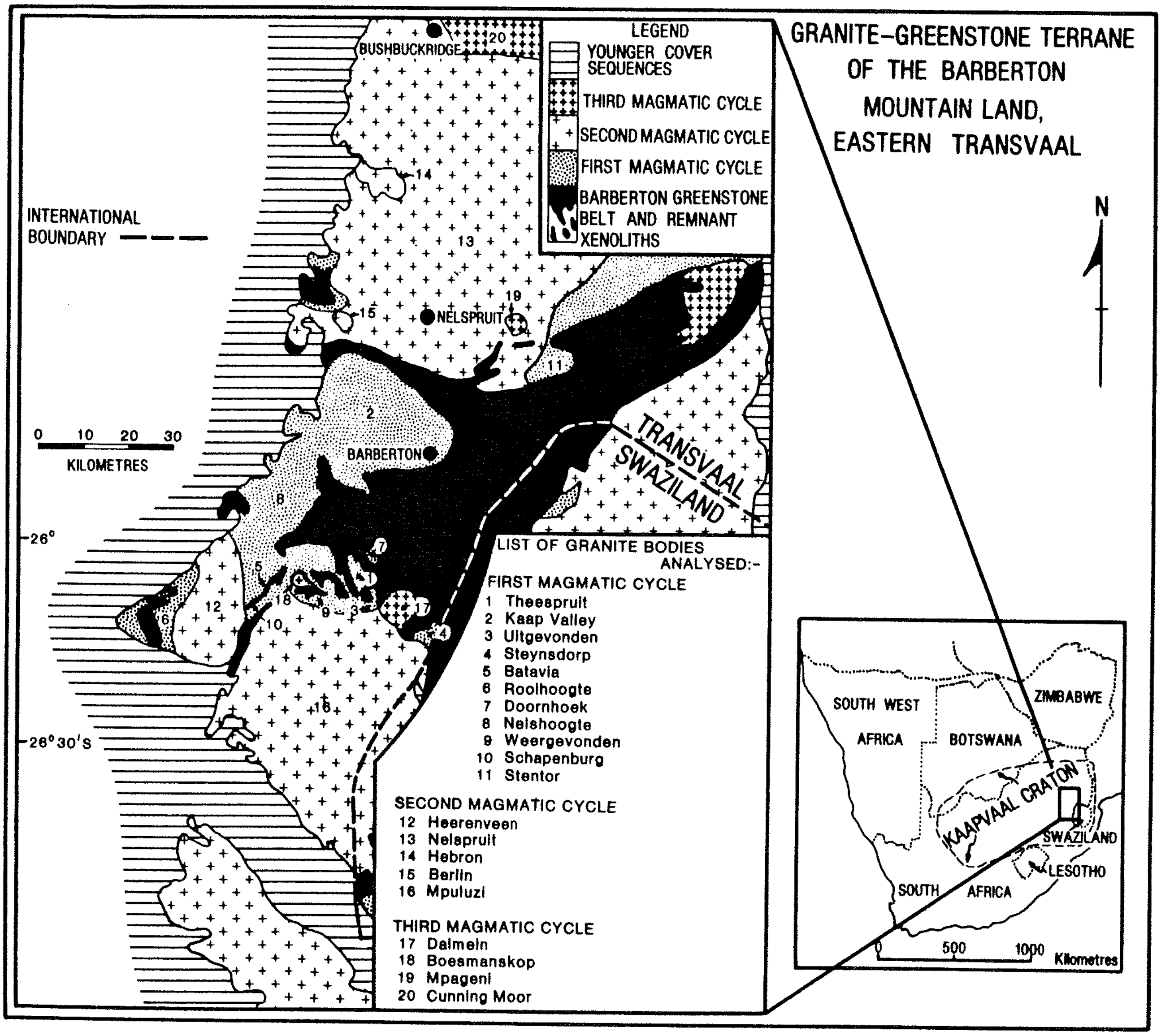

Figure 1. Geological sketch map of the Barberton Mountain Land showing the granitoid bodies sampled.

Granites of the second magmatic cycle were emplaced approximately 3200-3000 Ma ago and comprise large, multi-component, potassium-rich batholiths that were passively emplaced into the pre-existing trondhjemite-tonalite crust. Coarse-grained, relatively homogeneous, porphyritic granites, adamellites, and granodiorites form the bulk of these complexes which are also intruded by veins, as well as small plutons, of a medium-to fine-grained granodiorite phase isotopically coeval with the main complex. In the Mpuluzi batholith (Fig. 1) a prominent "hood" phase of homogeneous, medium-grained granodiorite-adamellite forms a carapace to the main intrusion. The margins of all the batholiths are characterized by potassium-rich migmatites and gneisses which represent zones of interaction between the batholith magma and the pre-existing crust. The batholiths of the second magmatic cycle are considered to have been derived by wide-spread partial melting of tonalite and trondhjemite gneisses (Robb *et al.*, 1983). Petrogenetic modelling suggests that the Heerenveen batholith (Fig. 1) resulted from a greater degree of partial melting than the Mpuluzi batholith, whereas the Nelspruit batholith to the north of the Barberton greenstone belt (Fig. 1) appears to have formed under conditions intermediate between the two (Anhaeusser and Robb, 1983).

The third magmatic cycle is characterized by the intrusion of discrete, granitic plutons which cross-cut all other Archaean rock types in the region, and vary in age between 3200 and 2500 Ma. Most of the plutons are homogeneous, coarse-grained, and porphyritic bodies that are adamellitic or granitic (*sensu stricto*) in composition, although one syenitic and one tonalitic pluton also fall within this category. The plutons of the third cycle have had diverse petrogenetic origins and cannot be characterized by a single model (Condie and Hunter, 1976; Robb, 1983). Generally, however, the late plutons are considered to have been derived by reworking of pre-existing crustal material such as that now represented in the study area.

III. ANALYTICAL METHODS AND SAMPLE POPULATION

One hundred and eighty-five samples were analyzed by instrumental epithermal neutron activation analysis at the Institute for Nuclear Chemistry, University of Cologne. A detailed description of the irradiation conditions and analytical procedures used, as well as an indication of the precision of the techniques employed, is presented in Meyer (1983).

The sample population analyzed was drawn mainly from the first and second magmatic cycles with only a minor contribution from the late plutons of the third cycle (Table I). A total of 93 samples from all individual tonalite-trondhjemite gneiss plutons of the first magmatic cycle were collected. Sixty-nine samples were selected from rocks of the second magmatic cycle. These are represented by the Heerenveen, Nelspruit and Mpuluzi batholiths, as well as the Hebron and Berlin granodiorite plutons, which form two of the isotopically coeval phases associated with the Nelspruit batholith. Only 23 samples were analysed from four of the late granite plutons of the third magmatic cycle. All the granitic bodies sampled for the purpose of this study are illustrated in Fig. 1.

IV. RESULTS

A. *U and Th Abundances*

The U and Th contents of the samples analyzed are summarized in Table I and presented graphically in Fig. 2 as a U versus Th scattergram. The U data range from 0,13 to 19,2 ppm and the Th values vary from 0,78 to 47,6 ppm. The highest U and Th contents are found in sample GM3 from the Mpuluzi batholith and in sample MPG4 from the Mpageni granite pluton. Samples TP55 and RH2 from the Theespruit and Rooihoogte trondhjemite plutons, respectively, carry the lowest U and Th contents.

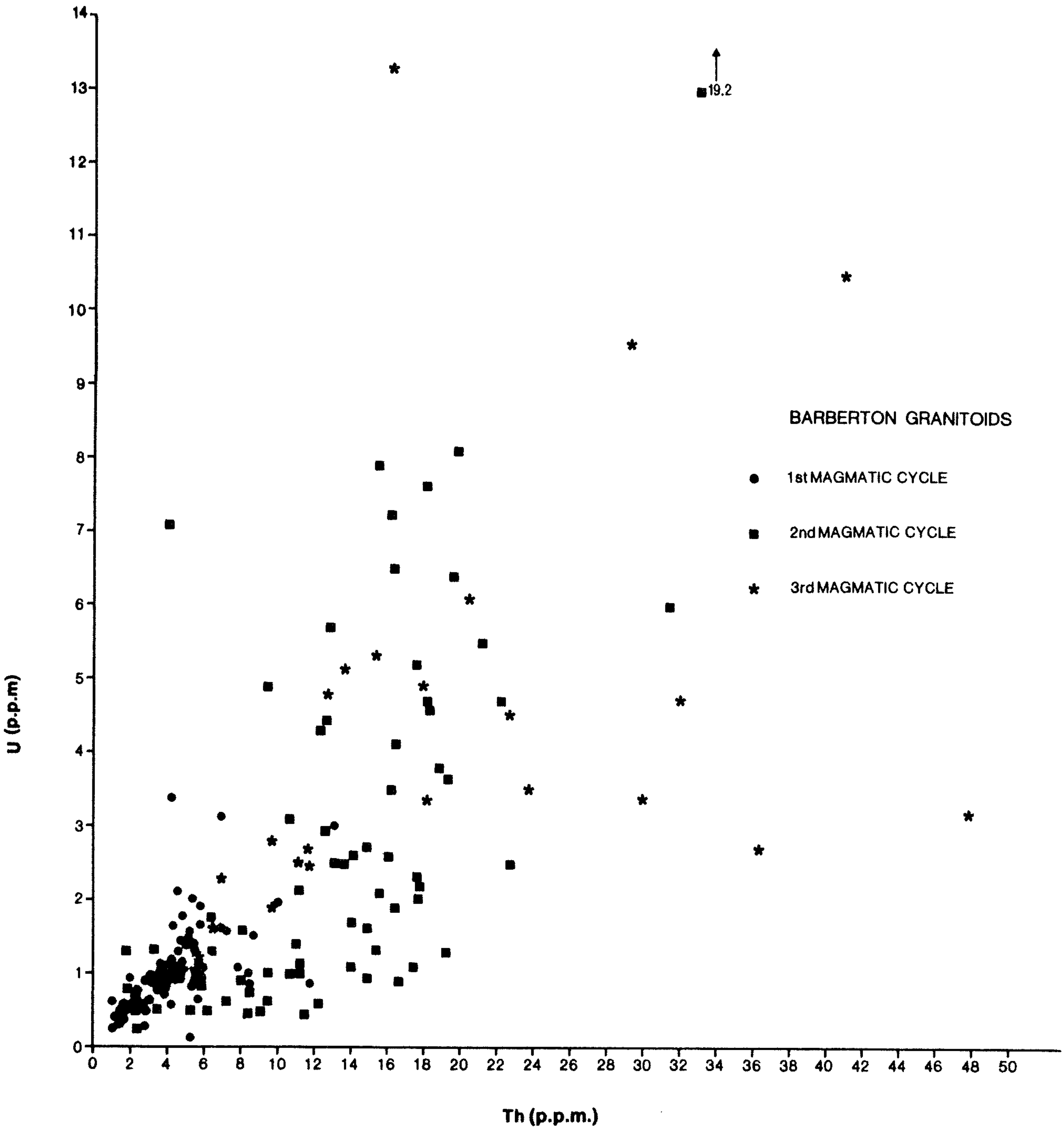

Figure 2. Plot of U versus Th for all individual samples analysed

B. <u>Distribution of U and Th</u>

The distribution of U and Th in rocks of each of the three magmatic cyles is shown in percentage histograms (Fig. 3). For all three populations the log-transformed U and Th data exhibit a quasi-normal distribution. Figure 3 also reveals a systematic increase of the median U and Th values from the first to the third magmatic cycle. This trend is also evident in the arithmetic and geometric mean U and Th values. As the data are log-normally distributed the geometric mean is regarded as the best measure of central tendency.

TABLE I

Th AND U CONTENTS IN GRANITOIDS FROM THE BARBERTON MOUNTAIN LAND

	Th	U
1ST MAGMATIC CYCLE		
THEESPRUIT		
TP3A	8,40	,84
TP4D	3,06	,69
TP5	5,84	,92
TP6	5,50	1,04
TP10A	4,83	1,04
TP14	5,46	1,33
TP15	3,27	,95
TP18	5,75	,98
TP21	3,45	,97
TP22	4,14	1,18
TP23	5,23	,92
TP25	4,49	1,09
TP30	3,75	,71
TP34	5,34	,83
TP36	5,62	1,16
TP38	3,58	,81
TP39A	3,28	,89
TP41	4,19	1,13
TP44	5,03	1,45
TP45	2,61	,90
TP47	4,99	1,38
TP49B	6,92	1,59
TP50	2,77	,82
TP52	1,25	,51
TP53	3,57	1,07
TP54	4,41	,86
TP55	4,61	,13
WN13	4,95	1,52
KAAP VALLEY		
LKV2	2,27	,67
LKV4	3,01	,94
LKV11	1,84	,51
LKV13	1,98	,90
LKV14	1,50	,46
LKV15A	1,22	,37
LKV23	1,32	,31
LKV25	2,91	,50
LKV28	3,87	,73
LKV29	2,88	,50
SKV2	4,16	,64
SKV5	3,59	1,15
SKV9	2,05	,61
SKV26	1,52	,41
SKV33	2,89	,57
NW2	1,58	,42
UITGEVONDEN		
AP10	4,79	1,76
AP13	9,98	1,94
AP14	5,57	1,92
AP16	5,69	1,07
AP17	5,07	1,55
AP19	4,68	,93
STEYNSDORP		
SP1A	4,67	1,45
SP2	5,64	,64
SP8	7,93	1,11
SP9	4,84	1,16
SP14	4,26	1,67
SP15	5,77	1,65
BATAVIA		
BT3D	11,70	,86
BT6	1,24	,42
BT9	2,74	,29
BT13	1,03	,23
BN3	1,48	,55
ROOIHOOGTE		
SY1	1,79	,37
SY3	2,39	,74
SM2	1,89	,54
RH2	,78	,38
DOORNHOEK		
DK7	6,84	3,15
DK17	13,10	3,00
DK22	4,56	2,13
DK36	5,36	1,99
WEERGEVONDEN		
LC2	2,39	,80
LC21	1,69	,57
LC27	8,66	1,51
LC35	4,47	1,32
LC36	8,38	,99
WN5	3,90	,89
WN9	4,03	,87
WN14A	3,76	1,05
SCHAPENBERG		
JV2	4,34	1,11
JV18	6,97	1,61
JV21	7,16	1,59
JV22	5,27	1,37
ET12	2,27	,28
STENTOR		
K4	13,20	2,50
K7	11,10	1,04
S43	,95	,54
KM6	15,40	1,33
KM10	14,00	1,66
KR2	11,20	1,09
KR8	9,55	,62
BL4	1,89	,78
HB7	8,14	1,58
NELSHOOGTE		
S60	2,06	,57
TS16	1,00	,62
2ND MAGMATIC CYCLE		
HEERENVEEN		
BC1	2,69	,61
BC4	2,38	,49
BC8	12,90	5,71
BC16	3,46	,48
BC17	12,70	2,94
BC19	14,90	2,71
BC32	3,25	,88
BC34	3,32	1,32
BC38	10,70	3,09
BC39	18,10	4,69
ET1	19,80	8,15
CB2B	2,40	,24
JV27	18,90	3,80
HEBRON		
C49	8,45	2,34
C39A	5,73	1,00
NELSPRUIT		
N38	14,90	1,60
N11A	17,60	2,32
N14	8,02	,91
N17	6,38	1,33
N21	9,47	1,02
N46A	16,10	2,59
K43	14,90	,96
S42	12,20	,61
B7	16,20	3,48
C39	3,87	,80
D28	6,10	,46
A31B	14,10	1,08
A31	7,23	,62
BERLIN		
N42	17,10	5,64
N44	20,10	2,73
MPULUZI		
BB5	31,70	5,97
BB9A	9,47	4,77
BB10	17,70	2,02
BB12	13,60	2,50
MF1	12,70	4,37
MF3	11,50	2,15
MF6	16,30	6,50
MF10	9,12	,48
MF12	16,40	1,86
OK12	7,35	,59
OK14	21,10	5,53
OK16	11,60	,44
TB3	16,40	4,12
TB8	8,45	,77
TB11	6,30	1,75
LR3	16,10	7,24
LR4A	18,30	4,58
LR5	17,50	1,10
LR6	19,40	1,30
GM1A	15,50	7,94
GM3	33,40	19,20
GM4	15,60	2,08
GM6	16,60	,87
GM7	19,30	3,73
JG1A	22,80	2,46
JG3	19,60	6,42
JG8	22,10	4,67
JG11	11,00	1,39
JG12	8,21	,39
JG14	19,30	1,31
AP11	17,80	7,68
AP12	4,12	3,37
L03	17,80	2,16
L05	10,80	,98
JV36	4,06	7,09
JV7	1,73	1,27
TS7	17,50	5,24
SP3	14,10	2,59
SP7	12,40	4,31
3RD MAGMATIC CYCLE		
DALMEIN		
DP1	18,20	3,35
DP6	12,80	4,83
DP10	7,00	2,30
CUNNINGMOOR		
F6	5,12	,97
F14	2,64	,68
BOESMANSKOP		
BK12	11,70	2,45
BK17	11,10	2,50
BK24	20,15	6,06
BK25	13,60	5,10
LC12	11,70	2,66
LC22	15,30	5,32
MY1	6,57	1,63
KZ1	41,30	10,50
KZ2	17,80	4,87
BT12	29,20	9,61
ND7	16,20	13,30
MPAGENI		
MPG1	9,81	1,91
MPG4	47,60	3,21
MPG5	22,70	4,53
MPG7	29,80	3,44
MPG9	36,20	2,72
MPG10	23,70	3,47
MPG12	31,90	4,73

(ALL VALUES IN PARTS PER MILLION)

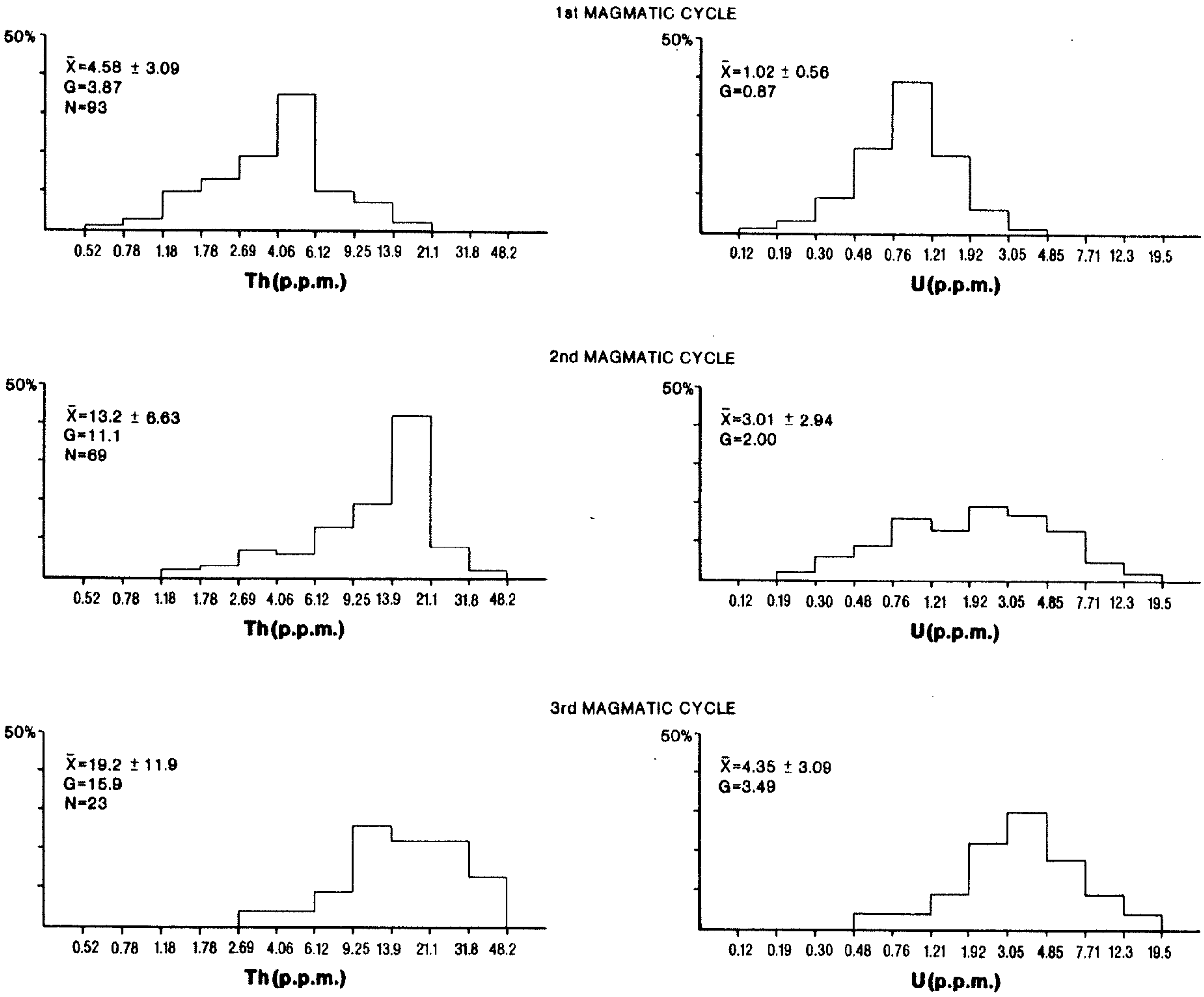

Figure 3. *Percentage histograms showing U and Th distribution in the three magmatic cycles*

C. *Th/U Ratios*

Figure 4 shows the distribution of decay-corrected Th/U ratios in rocks of the three magmatic cycles. As the principal U and Th isotopes possess different half-lives for their alpha-decay (i.e. $t_{1/2}$ ^{238}U = 4,51 x 10^9 years; $t_{1/2}$ ^{232}Th = 1,39 x 10^{10} years), and because of the widely differing ages of the granitoids in the study area (i.e. circa 3,5 - 2,5 Ga, Barton *et al.*, 1983; Barton, 1983) the absolute abundances of Th and U, and particularly the Th/U ratios, are markedly affected. This is emphasized in Fig. 4 where both the corrected and uncorrected Th/U ratios are compared. The decay-corrected geometric mean Th/U ratios are seen to be between 25% (third magmatic cycle) and 30% (first and second magmatic cycles) lower than the uncorrected values. The Th/U data for both sets of values are strongly positively skewed and it is apparent that the ratios derived from rocks in all three magmatic cycles exhibit lognormal distributions. The mean (G, $\bar{x}$) Th/U ratios of the first and third magmatic cycles are very similar whilst the granitoids of the second cycles possess the highest mean (G, $\bar{x}$) values and the highest standard deviations from the arithmetic means (Fig. 4).

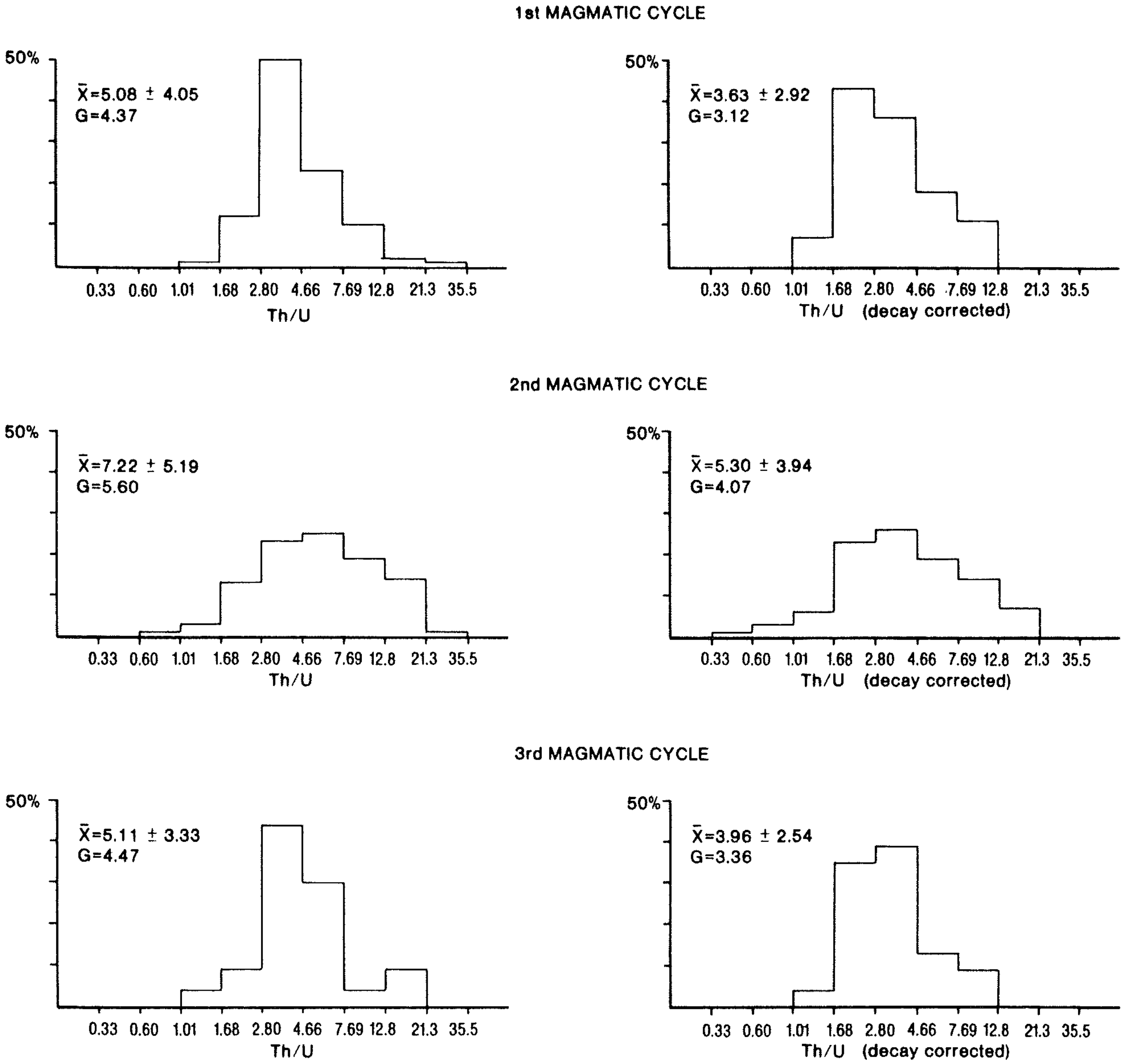

Figure 4. Percentage histograms showing distribution of Th/U ratios in the three magmatic cycles. Raw data (left hand side) are compared to the data corrected for alpha-decay of ^{232}Th and ^{238}U.

D. Average U and Th Abundances

The decay-corrected geometric mean U and Th values for the individual granitoid bodies investigated are given in Table II, and are presented graphically as a U versus Th scattergram in Fig. 5. These data indicate that the tonalite-trondhjemite plutons of the first magmatic cycle carry the lowest mean U and Th contents whilst the mean values for the potassium-rich batholiths and plutons of the second and third magmatic cycles are generally higher and more variable, but are not significantly different from each other. The mean U and Th contents of the Cunning Moor, Hebron and Heerenveen bodies are more akin to the mean values of rocks from the first cycle plutons, this being a function of the relatively sodic composition of all three of these bodies.

TABLE II

GEOMETRIC MEAN Th, U, Zr AND Ce CONTENTS AND GEOMETRIC MEAN Th and U CONTENTS CORRECTED FOR ALPHA-DECAY FOR INDIVIDUAL GRANITE BODIES SAMPLED

		N	G Th(ppm)	G U(ppm)	Th/U	G Zr(ppm)	G Ce(ppm)	* Th(ppm)	* U(ppm)	* Th/U
1ST MAGMATIC CYCLE										
Theespruit	(TS)	28	4,27	0,92	4,64	126	29,20	5,08	1,56	3,26
Kaap Valley	(KV)	16	2,24	0,57	3,93	117	34,60	2,67	0,98	2,72
Uitgevonden	(UG)	6	5,74	1,47	3,90	235	67,60	6,75	2,37	2,85
Steynsdorp	(SD)	6	5,41	1,22	4,43	205	52,30	6,43	2,07	3,11
Batavia	(BT)	5	2,27	0,42	5,40	132	30,40	2,61	0,64	4,08
Rooihoogte	(RH)	4	1,58	0,49	3,22	152	72,90	1,86	0,79	2,35
Doornhoek	(DH)	4	6,84	2,52	2,71	148	40,50	8,01	4,11	1,95
Nelshoogte	(NH)	2	1,44	0,59	2,44	87	26,30	1,69	0,97	1,70
Weergevonden	(WG)	8	4,08	0,96	4,25	158	61,00	4,74	1,50	3,16
Schapenburg	(SB)	5	4,82	1,02	4,73	168	53,80	5,67	1,65	3,44
Stentor	(ST)	9	7,16	1,11	6,45	216	107,00	8,23	1,82	4,52
2ND MAGMATIC CYCLE										
Heerenveen	(HV)	13	7,03	1,67	4,21	136	58,40	8,17	2,65	3,08
Nelspruit	(NS)	13	10,30	1,15	8,96	116	94,20	12,10	1,85	6,54
Hebron	(HB)	2	6,97	1,53	4,56	149	73,60	8,20	2,51	3,27
Berlin	(BL)	2	18,50	3,93	4,71	277	238,00	21,80	6,44	3,39
Mpuluzi	(ML)	39	13,20	2,52	4,77	208	116,00	15,30	4,00	3,84
3RD MAGMATIC CYCLE										
Dalmein	(DM)	3	11,80	3,34	3,52	203	125,00	13,90	5,48	2,54
Boesmanskop	(BK)	11	15,80	4,76	3,32	326	167,00	18,20	7,33	2,48
Mpageni	(MG)	7	26,30	3,30	7,97	367	332,00	29,90	4,85	6,16
Cunning Moor	(CM)	2	3,67	0,81	4,53	246	49,60	4,22	1,25	3,38

G = Geometric mean
* = Geometric mean corrected for alpha-decay
N = Number of samples

A noticeable feature of Fig. 5 is that the mean U and Th values for the Nelspruit batholith and the Mpageni pluton do not correspond to the well-defined trend delineated by the other bodies. The two points for the Nelspruit and Mpageni bodies exhibit the greatest deviation from the regression line which defines the mean Th/U ratio (Th/U = 3,00 r = 0,949) for all the Barberton granitoids, with the exception of the two errant points. The unusually high Th/U ratios of the Nelspruit batholith (6,59) and the Mpageni pluton (6,16) point either to a significant U loss or Th gain, and this aspect is discussed in more detail in the following section.

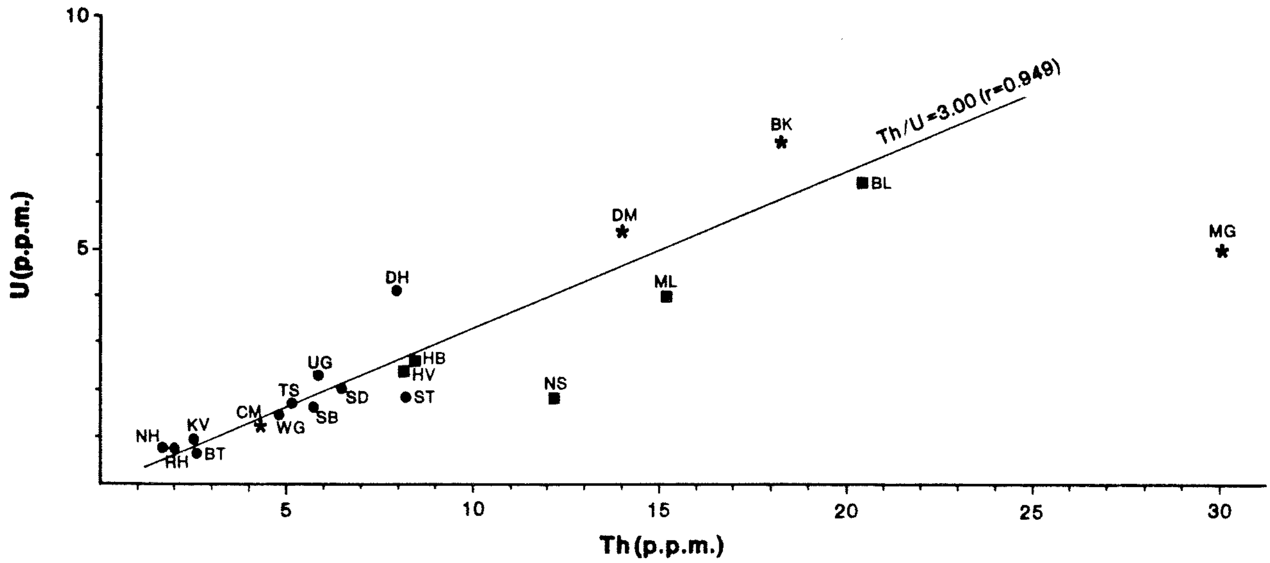

Figure 5. Plot of geometric mean U and Th contents for the individual granitoid bodies sampled. Abbreviations referred to in Table II.

V. DISCUSSION

It is evident, particularly in Fig. 5, that the U and Th contents of the Barberton granitoids exhibit a general increase from the rocks of the first magmatic cycle through to those of the third cycle. As mentioned earlier, it is considered that the magmatic cycles defined in the region are essentially related by a process of consecutive reworking of pre-existing crustal material. Consequently, it is felt that the U and Th distribution described above can best be explained by referring this pattern to the existing model for the petrogenetic evolution of the terrane. In the discussion that follows, only decay-corrected U and Th values are considered.

A. U and Th Distribution in Terms of Magmatic Processes

In an attempt to define the relationships between U and Th contents and the petrogenetic evolution of the Barberton granitoid terrane, plots of U and Th versus Rb-Sr whole rock ages and U and Th versus initial $^{87}Sr/^{86}Sr$ ratios (R) have been plotted (Fig. 6a-d).

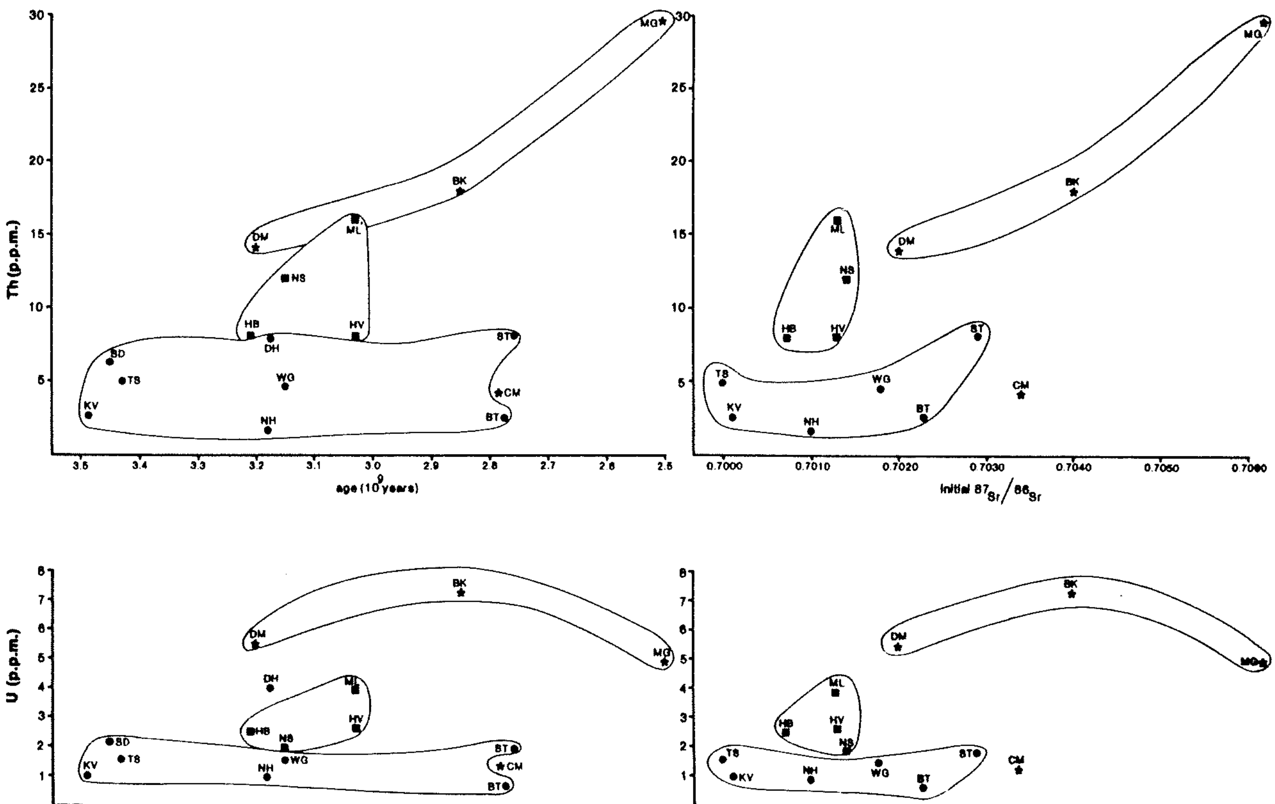

Figure 6. Plots of geometric mean Th and U contents for individual granite bodies sampled versus initial $^{87}Sr/^{86}Sr$ ratios (c and d) and Rb-Sr whole rock ages (a and b).

In the plots of R_0 versus U and Th (Fig. 6c, d) three broad fields, corresponding to the components of the three magmatic cycles, have been defined. In both plots, the tonalite-trondhjemite gneiss plutons of the first cycle define a flattish evolutionary trend where U and Th contents remain low over a considerable spread of R_0. This accords with the suggestion that these rocks were derived from primitive basaltic precursors (Robb and Anhaeusser, 1983; Barton, 1983) with the evolution of their Sr-isotopic ratios being controlled by vectors of shallow slope (i.e. controlled by low Rb/Sr ratios). Similar trends are evident in the plots of U and Th versus age, where it is clear that the rocks of the first magmatic cycle evolved over a considerable time span (i.e. circa 700 Ma). In spite of this prolonged evolution the U and Th contents remained relatively constant, indicating a measure of similarity in the process whereby these various bodies were formed. It is not, however, clear whether certain of the younger ages obtained for rocks of the first magmatic cycle reflect actual ages of emplacement or dates of tectonic/metamorphic resetting.

Rocks of the potassium-rich batholiths of the second magmatic cycle are all characterized by higher mean U and Th contents but with R_O's and ages of emplacement occupying more restricted ranges than the rocks of the first cycle. Consequently, only the more primitive members of the first magmatic cycle (i.e. the Kaap Valley, Theespruit and Nelshoogte plutons) could represent the type of material from which the magmas of the second cycle formed. The U and Th contents of the potassium-rich batholiths are consistent with a derivation by partial melting of tonalitic-trondhjemitic precursors (Robb *et al*., 1983). If the younger ages of the first cycle rocks are interpreted as reflecting a resetting episode then any of the tonalite-trondhjemite plutons defined in Fig. 6 could have constituted the source from which the potassium-rich batholiths were derived.

Three of the late granitoid bodies representing the third magmatic cycle define fields in Fig. 6a-d that are characterized by the highest U and Th contents. The Th data for these three bodies (Dalmein, Boesmanskop and Mpageni plutons) show a systematic increase with respect to both age and R_O. The higher Th and U contents are again consistent with a derivation by reworking of pre-existing crustal material. As mentioned earlier, however, the rocks of the third magmatic cycle cannot be simply unified in terms of a single petrogenetic model (Robb, 1983) and it is conceivable that rocks of either the first or the second magmatic cycles could represent the parental material. An exception to the general trend is afforded by the Cunning Moor tonalite which, although classified as a third cycle component in terms of field relationships, has an affinity with the first magmatic cycle, at least in terms of U and Th contents. This suggests that the Cunning Moor body may have been derived by similar processes to those responsible for the generation of rocks of the first magmatic cycle, but at a time relatively late in the geotectonic evolution of the region.

Although the above discussion holds true in terms of the broad trends, there are minor discrepancies which are noteworthy. In Fig. 6b it is apparent that the Doornhoek pluton of the first magmatic cycle is markedly enriched in uranium by comparison with the other gneiss plutons. It is pertinent to note that the R_O of this body is also extremely high (R_O = 0.7169; Barton *et al* ., 1983) and it is suggested that this reflects an open-system alteration in the trace element chemistry of this unit. The Th data in Fig. 6a and 6c confirms the previous suggestion that the Heerenveen, Nelspruit and Mpuluzi batholiths formed, respectively, by progressively decreasing degrees of partial melting of a tonalite-trondhjemite parent (Anhaeusser and Robb, 1983). In contrast the U data (Fig. 6b,d) does not reflect the systematic trend evident in the Th plots, and the Nelspruit batholith, specifically, appears to be depleted in uranium. This trend is also clearly illustrated in Fig. 5, where the high mean Th/U ratio of the Nelspruit batholith again points to significant U loss. Finally, U loss in the Mpageni pluton of the third magmatic cycle, is also apparently indicated in Fig. 6b and d. In contrast to the Th data, where a systematic increase in the sequence Dalmein-Boesmanskop-Mpageni is evident, the U content of the Mpageni pluton appears to be concomitantly low by a factor of approximately 2x. Questions related to the apparent loss of uranium, and also to the specific mineralogical siting of both U and Th in the Barberton granitoids, are discussed in the following section.

B. *Mineralogical Effects on U and Th Distribution*

The effects of mineralogy on U and Th distributions in the Barberton granitoids are examined in plots of Ce versus Th and Zr versus U (Fig. 7a, 7b). This association of elements is selected because petrographic observations indicate that zircon, apatite and allanite are the most abundant accessory mineral phases in which U and Th are likely to be hosted. In both cases a significant positive correlation between these elements is observed, although the data for the rocks of the first and second magmatic cycles define regression curves of different slopes. It is also clearly evident that a general increase in U and Th, as well as Zr and Ce, occurs in progressing from units of the first magmatic cycle through to the third cycle.

Although the plots in Fig. 7 suggest a relationship between U and Th contents and the abundance of accessory mineral phases such as zircon, apatite and allanite, as well as minor phases such as sphene and monazite, the effects of mineralogy are, in fact, best illustrated in plotting ratios such as Ce/Th, Ce/U, Zr/Th, and Zr/U against either U or Th (Fig. 8a-d). In the plots of Zr/Th and Ce/Th v Th (Fig. 8a, b) a clear cut distinction between the tonalite-trondhjemite gneiss plutons of the first cycle and the components of the second and third cycles is emphasized, with regression curves through the two populations being quite distinct. The contrast between the steep slopes defined by the first magmatic cycle and the shallow-to-horizontal trend of the second and third cycles is probably the result of differing mineralogical sitings for Th. The first cycle trends display variable Zr/Th and Ce/Th ratios which imply that Th contents can increase without their being a concomitant increase in either Zr or Ce. Consequently, a significant amount of Th must also be hosted in minerals other than the accessory phases, namely biotite and hornblende. Mineral analyses by fission track techniques indicate Th contents of up to 65 ppm in biotites and 40 ppm in hornblendes from the Kaap Valley and Theespruit plutons (Vorwerk, 1984). The second and third cycle trend illustrates a much greater degree of constancy in the Zr/Th and Ce/Th ratios, indicating that even substantial increases in Th contents are matched by concomitant increases in both Zr and Ce. Hence, the distribution of Th in this case can be accounted for almost entirely by the abundances of accessory minerals such as those listed previously. It is also apparent, from the consistently lower Zr/Th and Ce/Th ratios of the second and third cycle rocks relative to the first cycle plutons, that the dominant accessory minerals in the latter suite will generally contain higher Th contents than equivalent phases in the less evolved granitoids.

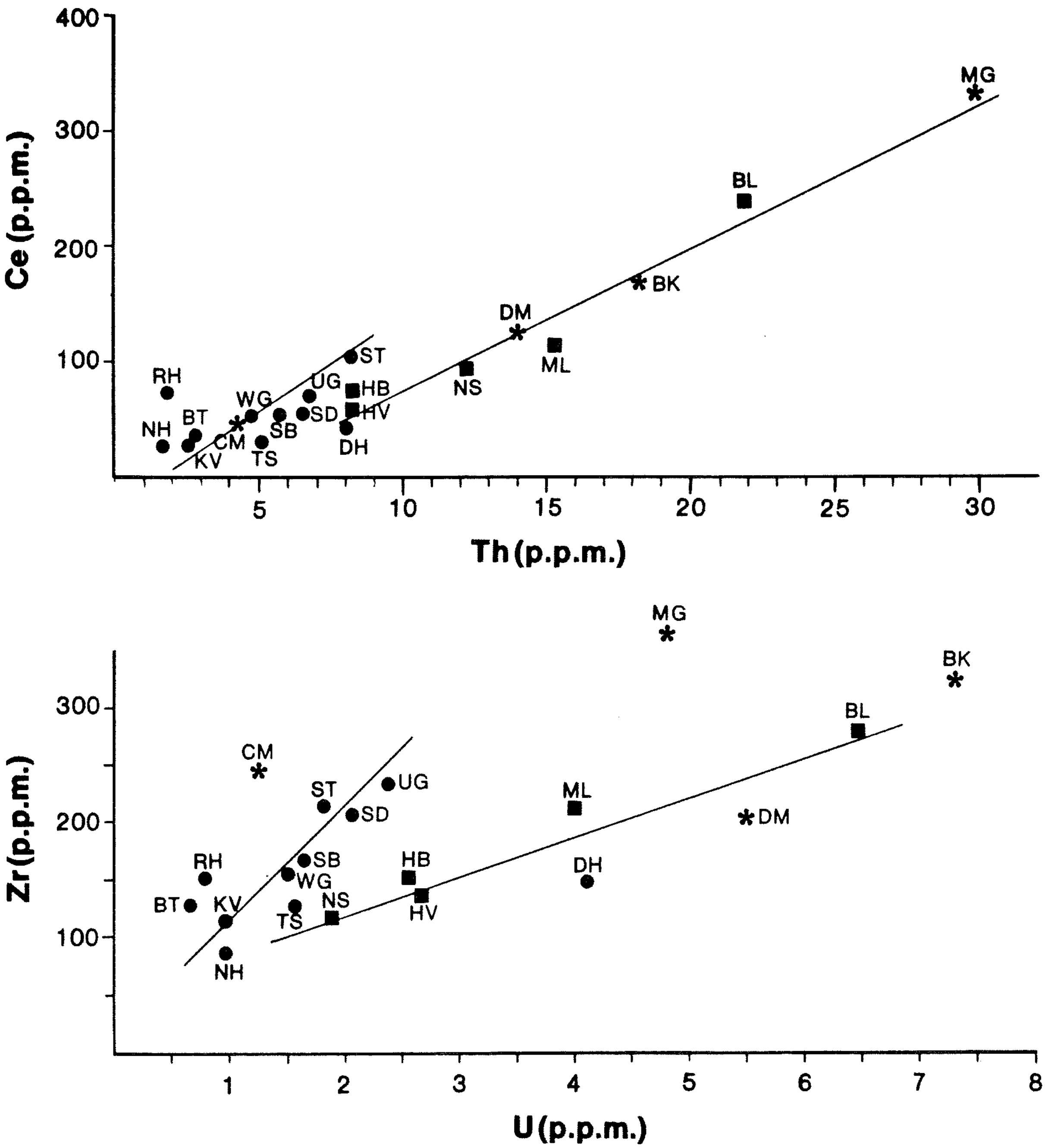

Figure 7. Plots of geometric mean Ce versus Th (a) and Zr versus U (b) contents for individual granite bodies sampled

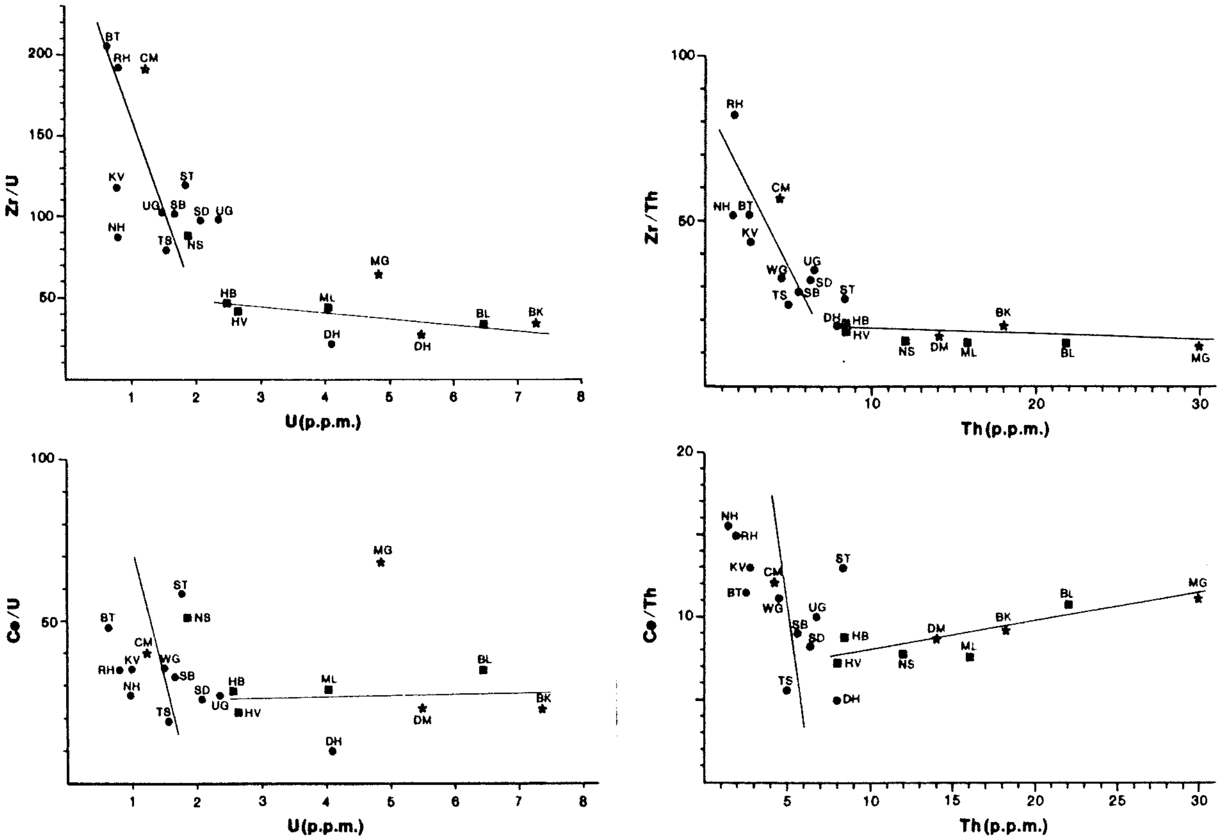

Figure 8. Plots of geometric mean Zr/Th and Ce/Th ratios versus Th (a and b), and Zr/U and Ce/U ratios versus U (c and d) for individual granite bodies sampled.

Plots of Zr/U and Ce/U versus U (Fig. 8c,d) exhibit similar trends and consequently, the conclusions pertaining to Th distributions apply equally to uranium. It is, however, apparent that, in terms of uranium distributions, certain of the granite bodies in the Barberton area do not conform to the established pattern, as defined in Fig. 8a and 8b. The Doornhoek pluton, for example, does not plot on the steep trend of the first magmatic cycle and is characterized by low Zr/U and Ce/U ratios (i.e. abnormally high U contents). The Doornhoek body is, therefore, enriched in U, but apparently not in Th, implying a decoupling of the one with respect to the other. This suggests that enrichment is not a magmatic process (i.e. where U and Th tend to behave similarly; Larsen and Gottfried, 1960; Gabelman, 1977), but possibly a hydrothermal concentration involving labile U.

The plots in Fig. 8c and 8d also indicate that the Nelspruit batholith and the Mpageni pluton are characterized by abnormally high Zr/U and Ce/U ratios, and as indicated previously, this suggests depletion of U in these two bodies. A detailed explanation of this phenomenon awaits further study, although at this stage two possible suggestions can be offered. In the case of the highly differentiated Mpageni granite, it is likely that the accessory mineral suite originally contained high U and Th contents. Consequently, the U-bearing accessory phases may have been subjected to significant radioactive damage resulting in lattice disturbances and permitting leaching of U by percolating fluids. In the case of the Nelspruit batholith this argument does not necessarily apply and U loss may be better explained by secondary processes. The Nelspruit batholith is, for the most part, underlain by a mature planation surface of late Cretaceous-early Palaeocene age (Lageat and Robb, 1984) which has been responsible for extended erosion and repeated exhumation of the batholith. It is possible, therefore, that this has resulted in significant leaching of labile U by surficial processes. In the case of the Nelspruit and Mpageni granites both the above suggestions point to a mineralogical siting of U whereby this element is perhaps more prone to remobilization than in the case of the other second and third magmatic cycle components.

VI. SUMMARY AND CONCLUSIONS

(1) Granitic rocks from the Barberton Mountain Land exhibit a range in U between 0.13 - 19.2 ppm and in Th between 0.78 - 47.6 ppm. The geometric mean for the total sample population is 1.8 ppm U and 8.8 ppm Th, yielding a mean Th/U ratio of 4.8. The distribution of the raw Th and U data, as well as the Th/U ratios, exhibits a distinct positive skew.

(2) Correction of the raw data for alpha-decay of ^{238}U and ^{232}Th yields U and Th abundances that are significantly different (up to 72% higher for U, and 19% higher for Th, at the time of emplacement) from the values measured at present. This is particularly relevant to the original Th/U ratios of Archaean granites and accounts for the difference between the mean uncorrected Th/U ratio of 4.8 compared to a figure of 3.5 at the time of granite formation.

(3) A general systematic increase in U and Th is evident in progressing from rocks of the first magmatic cycle through to the third magmatic cycle. This is consistent with previous ideas that interrelate the three granitoid cycles by progressive reworking of pre-existing crustal material.

(4) Trace element ratios (Zr/Th, Ce/Th, Zr/U, Ce/U) for tonalite and trondhjemite gneisses of the first magmatic cycle exhibit significant variation indicating that U and Th are not solely hosted in accessory phases such as zircon, apatite, allanite, sphene, and monazite, but also occur in other phases such as biotite and hornblende.

(5) By contrast, similar trace element ratios for the rocks of the second and third magmatic cycles are relatively invariable and indicate that increases in U and Th contents are matched by concomitant increase in Zr and Ce. It would appear, therefore, that most of the U and Th in these rocks is hosted in the accessory minerals listed above.

(6) Consistently lower Zr/Th, Zr/U, Ce/Th, Ce/U ratios in rocks of the second and third magmatic cycles, relative to the first cycle, indicate that the U and Th contents of individual accessory mineral phases are higher in more evolved granitoids.

(7) Certain granites from the study appear to be characterized by significant uranium loss (Nelspruit batholith and Mpageni pluton) whereas at least one body is apparently enriched in uranium (Doornhoek pluton).

The body of data presented here provides a comprehensive indication of U and Th abundances in typical Archaean granitoids. A further contribution of these data is that it raises a number of interesting questions which otherwise might have remained obscure. Clearly, additional work, such as fission track micro-mapping and U/Pb isotopic studies, is required before a complete understanding of the distribution and behaviour of the radio-elements in the Earth's primitive granitic crust is achieved.

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