ECONOMIC GEOLOGY

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URANIUM DISTRIBUTION AND REDISTRIBUTION
IN A SUITE OF FRESH AND WEATHERED
PRE-WITWATERSRAND AND WITWATERSRAND
CONGLOMERATES FROM SOUTH AFRICA

by

M. MEYER, R. SAAGER and V. KÖPPEL

UNIVERSITY OF THE WITWATERSRAND
JOHANNESBURG

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M. MEYER

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

R. SAAGER

(Mineralogisch-Petrographisches Institut, Universität zu Köln,
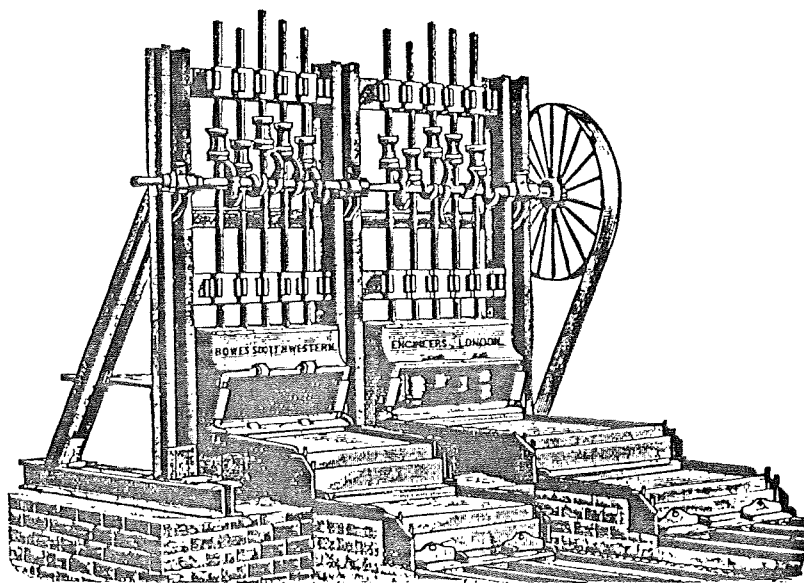*Zulpicherstrasse 49, D-5000 Köln, West Germany)*

and

V. KÖPPEL

(Institut für Kristallographie und Petrographie,
ETH, Zürich, Switzerland)

1886–1986

A CONTRIBUTION TO THE CELEBRATION OF THE
CENTENARY OF THE DISCOVERY OF THE
WITWATERSRAND GOLDFIELDS

URANIUM DISTRIBUTION AND REDISTRIBUTION IN A

SUITE OF FRESH AND WEATHERED PRE-WITWATERSRAND AND

WITWATERSRAND CONGLOMERATES FROM SOUTH AFRICA

ABSTRACT

        Uranium analyses of fresh and weathered Precambrian uraniferous conglomerate samples revealed that the weathered material experienced considerable uranium depletion. Alpha-spectrometric and gamma-spectrometric investigations as well as uranium leaching tests indicated that in weathered conglomerates only insignificant amounts of uranium are still available for supergene redistribution and that the bulk of the uranium was removed very early in the weathering history of the rocks. Consequently, gamma- and alpha-spectrometric measurements do not yield useful information on the original uranium content of the weathered rocks. However, lead isotope analysis gave good estimates of the fresh-rock uranium tenor of weathered conglomerates. This method is not sensitive to recent uranium redistribution and, in geological terranes similar to the ones investigated, it provides a helpful tool in the exploration for uranium deposits.

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I. INTRODUCTION

Under oxidizing low temperature conditions in the weathering environment the uranyl ion $(UO_2)^{2+}$ constitutes the dominant stable form of uranium. Depending upon prevailing Eh-pH conditions and on the availability of suitable cations the uranyl ion forms soluble or insoluble complexes and, accordingly, uranium can be depleted or concentrated in weathered rocks. This behaviour, which might affect the uranium content of weathered rocks, could influence geochemical exploration data to a considerable extent. For example, it is known that surface outcrops of Precambrian uraniferous quartz-pebble conglomerates carry lower uranium contents than their unweathered equivalents (Saager *et al.*, 1981; Meyer *et al.*, 1983).

The present study was undertaken to test the applicability of various analytical methods for the evaluation of the fresh-rock uranium grade of weathered conglomerates. One hundred and forty three samples from pre-Witwatersrand and Witwatersrand sedimentary rocks were studied by ore microscopy, gamma-spectrometry, alpha-spectrometry, neutron activation analysis, and lead isotope analysis.

II. SAMPLE DESCRIPTIONS

Thirteen samples were collected from supergenically altered conglomerate outcrops of three pre-Witwatersrand sedimentary deposits:

(i) the approximately 3300 Ma old Moodies Group of the Barberton Sequence;

(ii) the approximately 3000 Ma old Pongola Sequence; and

(iii) the Uitkyk Formation which is considered to be part of the Pietersburg Group. The age of the Uitkyk Formation, is uncertain and compares either with that of the Moodies or Dominion groups (i.e. 2800 Ma).

One hundred and thirty samples originate from the approximately 2700 Ma old West Rand Group (formerly known as the Lower Witwatersrand Succession). In addition to weathered surface material (78 samples), fresh core material (52 samples) was available. Figure 1 provides a geologic sketch map of the northeastern part of South Africa and shows the 9 sample localities.

All the samples investigated were subjected to lower greenschist facies metamorphism and can best be described as metaconglomerates.

Microscopic studies of weathered conglomerates revealed the total absence of non-refractory uranium minerals and indicated that pyrite, which in these samples was originally the most abundant detrital mineral, is altered to limonite/hematite aggregates. Uranium-bearing minerals still present include allogenic zircon and monazite. From the work carried out by Thiel *et al.* (1979) on similar rocks it is known that uranium is also present as adsorptions on carbonaceous matter, on secondary Fe-Ti oxyhydroxides, and on phyllosilicates.

The fresh borehole core samples, in contrast, contain unaltered pyrite and other sulphides, with uraninite, brannerite, and uraniferous leucoxene being the main non-refractory uranium hosts.

III. RESULTS AND DISCUSSION

The analytical methods employed in this study are depicted in Fig. 2. Additional information has been provided by Meyer *et al.* (1983).

Figure 3 shows the ranges and geometric as well as arithmetic mean values of the uranium contents obtained by *neutron activation analysis* of weathered conglomerates, fresh conglomerates, and fresh quartzites. The graph reveals the conspicuous difference in uranium concentration which exists between fresh and weathered conglomerates and it also demonstrates that quartzites possess generally lower uranium concentrations than conglomerates.

A. Gamma- and Alpha- Spectrometry

A comparison of uranium contents obtained by gamma spectrometry with uranium contents measured in the same conglomerate samples by means of neutron activation analysis indicates discordancy for practically all samples (Fig. 4). This points to the widespread occurrence of radioactive disequilibrium in the weathering zone of the conglomerates, i.e. it shows that the equilibrium between the measured gamma emitter (^{214}Bi) and its parent nuclide ^{238}U is disturbed. Such disequilibrium is the result of differences in mobility which the various members of the ^{238}U decay series possess in meteoric waters, e.g. ^{234}U is more mobile than the other two long-lived nuclides ^{238}U and ^{230}Th.

In the case of radioactive disequilibrium caused by the disturbance of one of the members of the decay chain, 95 per cent equilibrum will be restored after about four times the half-life of the disturbed member. If ^{234}U was disturbed this takes about one million years.

^{238}U, ^{234}U and ^{230}Th are strong alpha emitters and, therefore, alpha-spectrometry has proved to be a suitable method to study recent uranium redistribution processes, i.e. processes which took place within the last one million years.
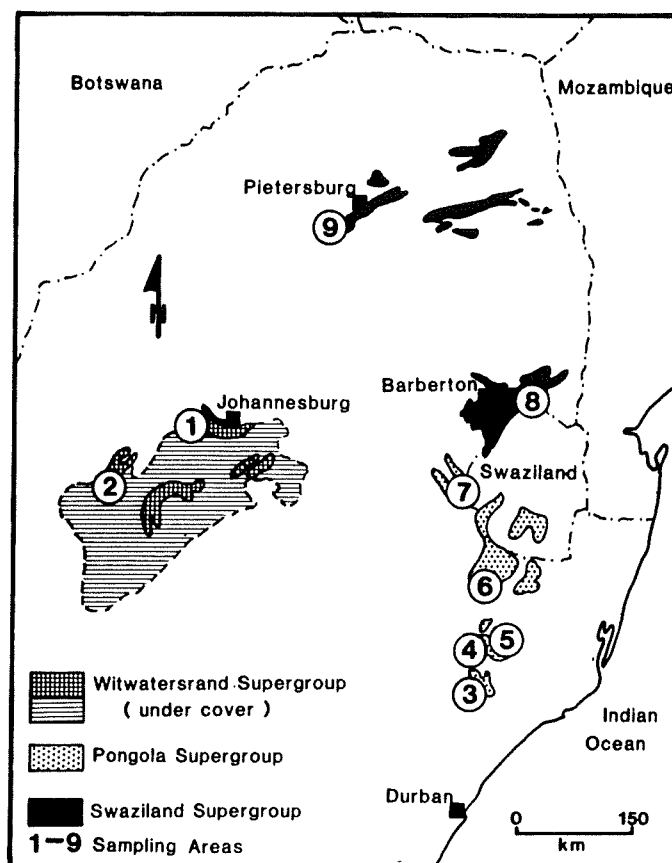
Figure 1. *Geologic sketch map of the northeastern portion of South Africa
showing sample localities of early Precambrian conglomerates.*

West Rand Group:	(1) Klerksdorp area
	(2) West Rand area
Pongola Sequence:	(3) Copper's Store
	(4) Speedwell
	(5) Denny Dalton
	(6) Gunsteling
	(7) Redcliff
Moodies Group:	(8) Bearded Man
Uitkyk Formation:	(9) Mount Robert

The activity ratios of $^{234}U/^{238}U$ and $^{230}Th/^{234}U$ for weathered and fresh conglomerate samples from the West Rand Group are plotted in Fig. 5. Two fresh samples taken from borehole core possess ratios of one and, consequently, are in a state of radioactive equilibrium. In contrast to this, all weathered conglomerates exhibit radioactive disequilibrium. Using the diagram of Thiel *et al.* (1983) it may be concluded that the uranium in these samples underwent a complex redistribution history with more than one stage of uranium loss and/or gain.

B. Uranium Leaching Tests

As a consequence of the data obtained from neutron activation analysis and the gamma- and alpha-spectrometric studies, uranium leaching tests on fresh and weathered samples were performed (Fig. 6). These tests show that in fresh conglomerates between 48 and 90 per cent of the total uranium content is leachable by hot HNO_3 while in weathered samples only 4,4 to 18,5 per cent (average 7,7%) of the uranium can be removed.

Assuming that the leaching experiments simulate natural supergenic processes the tests indicate that, in weathered samples, only small, almost insignificant, amounts of uranium are available for redistribution detectable by alpha-spectrometry.

Tests on fresh samples indicate that significant amounts of uranium can be removed by leaching. For weathered samples, which were originally uranium rich, this implies that they lost the bulk of their uranium content during their first exposure to weathering. In the investigated terranes the onset of weathering must have occurred more than one million years ago. The large-scale uranium loss caused by the first period of weathering is, therefore, undetectable by alpha-spectrometry.

Analytical Methods Used

γ-spectrometry	indirect U and Th assays via ^{214}Bi and ^{208}Tl
α-spectrometry	direct U and Th assays and determination of ^{238}U, ^{234}U and ^{230}Th activities
Pb-isotope analysis	determination of Pb-isotope ratios (^{208}Pb/^{204}Pb, ^{207}Pb/^{204}Pb, ^{206}Pb/^{204}Pb)
Neutron activation analysis	determination of U and Th and other major and trace elements

^{234}U, ^{230}Th, ^{214}Bi, and ^{206}Pb members of ^{238}U decay chain

^{208}Tl and ^{208}Pb members of ^{232}Th decay chain

^{207}Pb member of ^{235}U decay chain

^{204}Pb non-radiogenic lead

Figure 2. Analytical methods used in this study.

From the above it may be concluded that uranium redistribution in the secondary environment is not governed by simple systematic mechanisms. The present uranium contents of weathered conglomerates are, therefore, not related to their fresh-rock uranium tenor and cannot be used to evaluate the potential of such rocks for the presence of uranium mineralization.

C. Lead Isotope Analysis

Lead isotope analysis might provide a means of estimating the fresh-rock uranium content of weathered rocks as this method is insensitive to recent uranium redistribution.
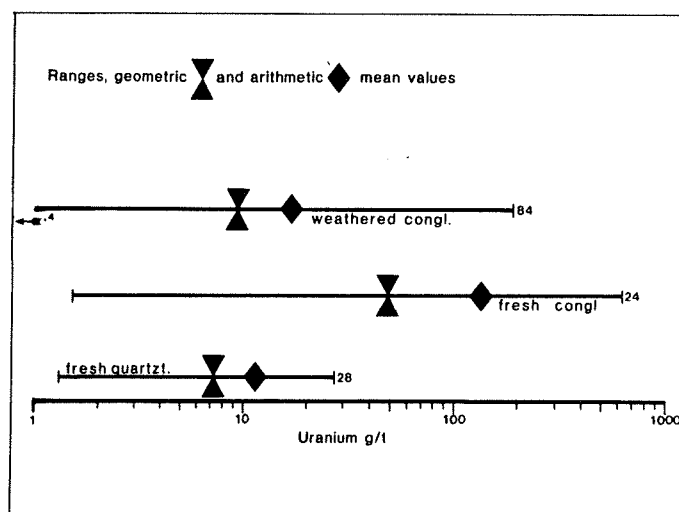

Figure 3. Ranges, geometric and arithmetic mean uranium values of various rock types investigated. Figures on the right end of the bars give number of samples analysed.

The original uranium contents of a sample can be calculated from its lead or thorium content and from its lead isotope ratios (^{206}Pb/^{204}Pb, ^{207}Pb/^{204}Pb, and ^{208}Pb/^{204}Pb), if the following assumptions are made:

(1) the U-Th-Pb system was closed from the time of conglomerate formation until onset of weathering, i.e. until recent times;

(2) thorium and lead were immobile in the weathering environment;

(3) the time t of sedimentary deposition of the conglomerates is known; and

(4) an appropriate common lead correction is chosen.

If time t is not known it can be chosen by assuming an age (t_{ass}) on geologic reasoning or by calculting an age with the help of lead-lead ($t_{207Pb/206Pb}$) or thorium-lead ($t_{208Pb/232Th}$) isotopic data.
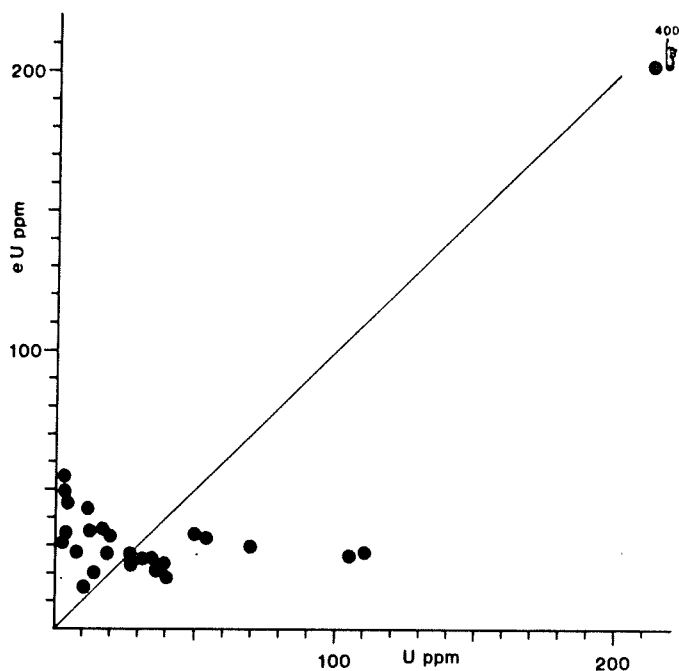
Figure 4. Comparison of uranium values obtained by gamma-spectrometry (eU) with uranium values obtained by neutron activation analyses (U) of weathered conglomerate samples.

Fresh-rock uranium contents of weathered conglomerates can be calculated by the following two equations:

$$(1) \quad ^{238}U \quad = \frac{(^{206}Pb/^{204}Pb)_m - (^{206}Pb/^{204}Pb)_t}{(e^{\lambda_8 t} - 1)} \times \quad ^{204}Pb$$

where: $(^{206}Pb/^{204}Pb)_m$ = measured isotopic ratio of weathered rock

$(^{206}Pb/^{204}Pb)_t$ = isotopic ratio of rock at time t (common lead correction)

λ_8 = decay constant of ^{238}U

^{204}Pb = measured non radiogenic lead in rock

t = time of conglomerate formation

It should be noted that equation (1) implies that the lead isotopes were homogenized close to the time of conglomerate formation or that the crystallization of the detrital uranium carriers does not differ appreciably from time t. Furthermore, equation (1) shows that the calculated uranium concentration is affected by possible recent lead gains or losses. In the present study the common lead correction was based on the lead evolution model of Stacey and Kramers (1975). However, the inaccurate choice of $(^{206}Pb/^{204}Pb)_t$ has only a minor influence on the final result because most of the measured samples possess some radiogenic lead. Of more importance is the right choice of t, as the calculated uranium values are sensitive to small changes of t. Meyer *et al.* (1983) showed that a change from 2700 to 3000 Ma will decrease the calculated uranium concentration by 12 per cent.

$$(2) \quad ^{238}U = \,^{232}Th \times (^{206}Pb/^{208}Pb)_r \times \frac{e^{\lambda_2 t} - 1}{e^{\lambda_8 t} - 1}$$

where: $(^{206}Pb/^{208}Pb)_r$ = isotopic ratio of radiogenic ^{206}Pb and ^{208}Pb generated in the sample since time t

λ_2 = decay constant of ^{232}Th

^{232}Th = measured thorium content of rock

In this case the calculated uranium value is influenced by recent thorium gains or losses, but it is not affected by recent changes in the lead content of the rock. The choice of time t is less critical in the second equation than in the first and a change of t from 2700 to 3000 Ma will decrease the calculated uranium content by only 2 per cent.

In both equations uranium does not occur on the right hand side and, therefore, recent uranium losses, caused by weathering, have no effect on the calculated uranium values. However, the reliability of the calculated uranium values depends on the closed system behaviour for U-Th-Pb between the time t and the onset of weathering. Existence of open system conditions can be detected by comparing the assumed age of the rock with its calculated $^{207}Pb/^{206}Pb$ and $^{208}Pb/^{232}Th$ ages.

U-234/U-238

O borehole samples
● surface samples

radioact. equil.

Th-230/U-234

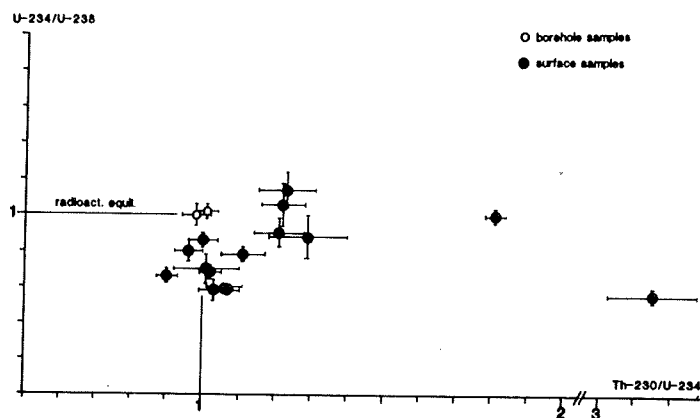*Figure 5. Alpha-activity ratios for weathered surface samples and fresh borehole samples.*

URANIUM LEACHING EXPERIMENTS

total U content
leachable U
( ) percentage of leachable U

fresh borehole samples

(47.9)

(90.1)

weathered surface samples

(5.7)

(6.3)

(18.5)

(4.4)

(6.5)

(5.1)

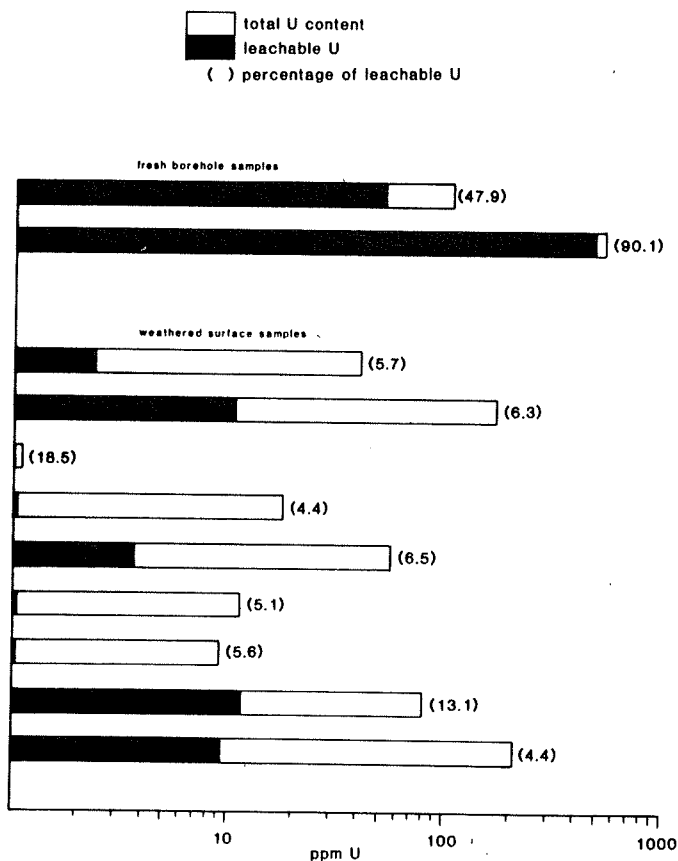(5.6)

(13.1)

(4.4)

10   100   1000
ppm U

Figure 6. Comparison of total uranium content with leachable uranium content of two fresh and nine weathered conglomerate samples.

Figure 7 reveals that for only a few samples is the assumption of closed system behaviour fulfilled. Most samples from the West Rand Group, for example, display open system behaviour and show the following trend:

assumed sedimentary age > lead-lead age > thorium-lead age

This trend suggests that the lead isotopic composition was influenced after deposition of the West Rand Group sediments, possibly by the intrusion of the Bushveld Igneous Complex, and that either lead was lost or thorium gained by the system.

For the *fresh samples* from borehole cores it is safe to assume that the U-Th-Pb system was not disturbed by supergene weathering. Therefore, the samples from the West Rand Group must have suffered their lead loss or thorium gain between time t and the onset of weathering (10 to 100 Ma ago). Depending upon whether a lead loss or a thorium gain occurred, the equations either yield minimum uranium (equation 1) or maximum uranium (equation 2) values. Minimum and maximum uranium values for a number of fresh core samples were accordingly calculated and compared with uranium values directly determined by neutron activation analyses (Fig. 8). The graph indicates that the directly measured uranium concentrations generally lie between the two calculated values and thereby support the above assumption. It is noteworthy that the mean of the two calculated values generally does not deviate by more than a few per cent from the measured values and in only one case was the deviation found to be close to 20 per cent.
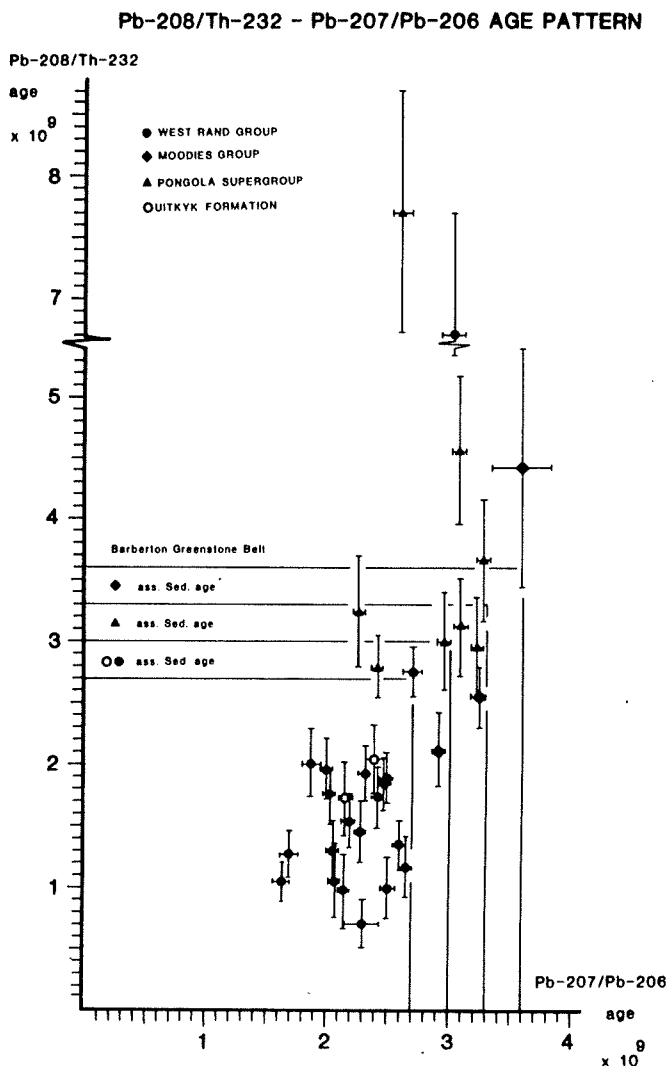
Figure 7. $^{208}Pb/^{232}Th$ and $^{207}Pb/^{206}Pb$ ages obtained for the various samples investigated.

The history of the U-Th-Pb system of weathered samples is generally more complex because it normally is subjected to a recent disturbance. A comparison of the ages (t_{ass}, $t_{207Pb/206Pb}$, $t_{208Pb/232Th}$) will often indicate how the system was disturbed and, consequently, the age pattern is a helpful guide in evaluating the calculated original uranium concentrations. Six patterns can be distinguished:

(1) $t_{ass} \sim t_{207Pb/206Pb} \sim t_{208Pb/232Th}$

The measured and assumed ages are concordant which indicates that the U-Th-Pb system was closed between the time of lead isotope homogenization and the onset of weathering, and that the time of lead isotope homogenization was not much different from the time of conglomerate deposition. It further shows that thorium and lead were not affected by supergene alteration. The calculated uranium values are, therefore, close to the fresh-rock uranium contents of the conglomerates;

(2) $t_{ass} > t_{207Pb/206Pb} \sim t_{208Pb/232Th}$

The calculated ages are concordant, but lower than the assumed age. This may indicate that the two measured ages were completely reset or that t_{ass} is wrong. Therefore, the best estimate for the calculated uranium value is obtained by inserting the measured ages into the two equations;

(3) $t_{ass} > t_{207Pb/206Pb} > t_{208Pb/232Th}$

This pattern indicates open system behaviour with a thorium gain or a lead loss possibly also during weathering. In view of the fact that lead has higher supergene mobility than thorium, equation (2) using the lead-lead age should be employed for the calculation of the fresh-rock uranium content;

(4) $t_{ass} \sim t_{207Pb/206Pb} < t_{208Pb/232Th}$

This pattern points to severe disturbances of the Th-Pb system. Because of the concordancy between t_{ass} and $t_{207Pb/206Pb}$ a lead gain seems to be unlikely and, therefore, only a thorium loss should be assumed and equation (1) used for the calculation of the fresh-rock uranium tenor;

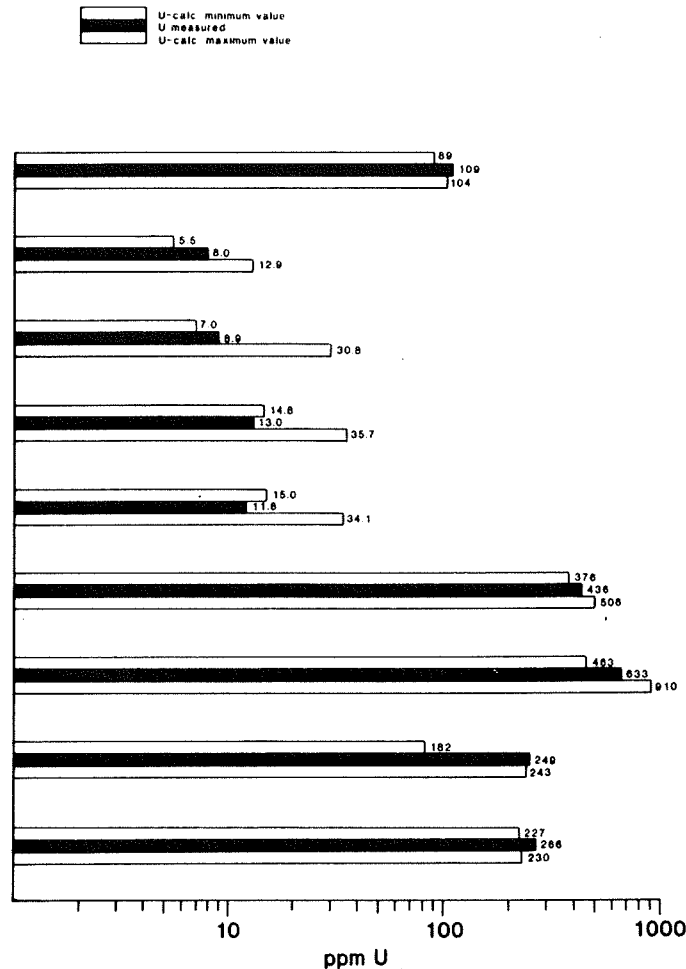**Measured & Calculated U Concentrations For Borehole Samples**

Figure 8. Comparison of calculated minimum and maximum uranium contents with measured uranium content of nine fresh conglomerate samples.

(5) $t_{ass} \sim t_{207Pb/206Pb} > t_{208Pb/232Th}$

Severe disturbances of the Th-Pb system are also reflected by this pattern. It is, however, uncertain whether a lead loss or thorium gain (recently or in the past), or both, occurred. Consequently, only equation (1) can be used which yields a minimum calculated uranium value; and

(6) $t_{207Pb/206Pb} < t_{ass} < t_{208Pb/232Th}$

This pattern reveals severe disturbance of the U-Th-Pb system before onset of weathering, and a recent thorium loss or lead gain cannot be ruled out. Assuming that lead has a higher supergene mobility than thorium, calculation of the fresh-rock uranium content must be carried out with equation (2) and using t_{ass} for time t.

Most of the samples from the West Rand Group follow age pattern (3) and those from the Pongola Sequence age pattern (1). The age patterns of the samples from the Uitkyk Formation and from the Moodies Group vary more randomly.

Figure 9 shows, for a number of weathered samples, the fresh-rock uranium contents (best estimates calculated according to the rules discussed in the above 6 cases), the actual measured uranium contents (measured by neutron activation analysis), and the differences between the two uranium values. These differences are equal to the theoretical uranium losses suffered by the samples during weathering. It should be noted that only one sample deviates from the general pattern and shows a theoretical uranium gain.

The recognized theoretical uranium losses closely resemble the uranium losses inflicted by leaching of fresh samples (Fig. 6). In addition, in Table I the measured uranium contents of fresh borehole core samples are compared with calculated uranium values of weathered conglomerate samples. Each of the sample pairs compared originate from one particular stratigraphic horizon.
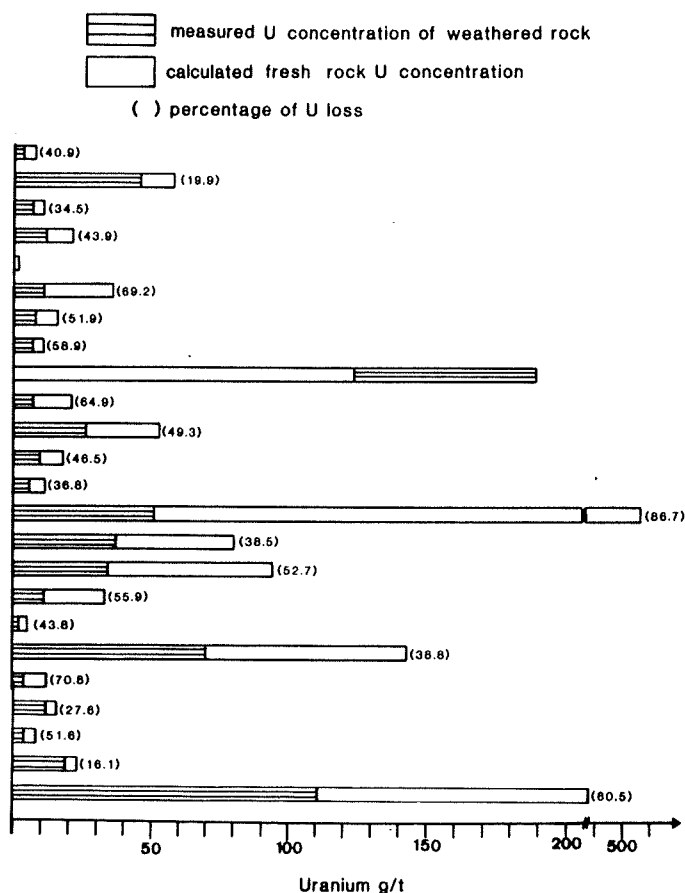
Fresh Rock U Tenor For Weathered Samples

measured U concentration of weathered rock

calculated fresh rock U concentration

( ) percentage of U loss

(40.9)
(19.9)
(34.5)
(43.9)

(69.2)
(51.9)
(58.9)

(64.9)
(49.3)
(46.5)
(36.8)
(86.7)
(38.5)
(52.7)
(55.9)
(43.8)
(38.8)
(70.8)
(27.6)
(51.6)
(16.1)
(60.5)

50   100   150   200   500

Uranium g/t

Figure 9.   Comparison of calculated uranium contents with measured uranium contents of 24 weathered conglomerate samples.

TABLE I

COMPARISON OF CALCULATED AND MEASURED URANIUM CONTENTS OF CORRESPONDING WEATHERED AND FRESH CONGLOMERATE SAMPLES

Weathered Sample	U (NAA) ppm	U (calc) ppm	Fresh Sample	U (NAA) ppm	U (calc) ppm
UCZ 1	111	202	HB 310	109	104
TPQ 1	3,5	7,9	HB 316	8,0	12,9
			HB 313	11,8	15,0
LCZ 1	18,8	22,4	HB 24	8,9	7,0
LTQ 1	11,8	16,3	HB 27	13,2	14,9

The results shown in Table I and Fig. 9 are distinct indications that lead isotope analyses, in contrast to the earlier discussed methods, yield sensible estimates on the fresh-rock uranium tenor of weathered conglomerate samples and, thus, provide a useful tool for uranium exploration of deeply weathered, potentially uraniferous, conglomerates or similar sedimentary rocks.

IV. SUMMARY AND CONCLUSIONS

The comparison of uranium analyses of Precambrian uraniferous conglomerate samples originating from weathered outcrops and from fresh borehole core material reveals that the weathered samples underwent uranium depletion. In addition, ore microscopy showed that brannerite and detrital uraninite are the principal uranium minerals in fresh conglomerates, whereas weathered conglomerates only contain refractory uranium carriers, such as detrital zircon and monazite.

These observations agree with the well-established fact that uraninite and brannerite are unstable in the secondary environment where uranium is oxidized to the uranyl ion which readily forms soluble complexes and is redistributed by meteoric waters. Formation of H_2SO_4 by the decomposition of sulphide minerals further increases the mobility of uranium in the weathering zone.

If uranium redistribution occurred within the last one million years it can be studied by alpha-spectrometry. Such investigations were carried out on the weathered samples and revealed the presence of radioactive disequilibria. They were interpreted to be the results of complex uranium redistribution processes (several stages of uranium loss and/or gain) which occurred in the sampled conglomerate outcrops. Consequently, the results of gamma-spectrometric surveys undertaken over these conglomerate outcrops do not reflect their actual uranium content.

Uranium leaching tests, using hot HNO_3 attack, were performed on fresh and weathered conglomerate samples. The tests revealed that in fresh conglomerates the bulk of the uranium is leachable. This contrasts with the weathered conglomerates from which only insignificant proportions of the uranium could be removed by the acid. These results suggest that in weathered conglomerates only small amounts of uranium are still available for supergene redistribution and that the bulk of the uranium contained in the conglomerates was removed during an early stage of their weathering history, probably during their first supergene alteration. In the geological terrane studied these processes occurred more than one million years ago and, therefore, are not traceable by alpha-spectrometry.

Lead isotope analysis provides a method of calculating the fresh-rock uranium content of weathered rocks because the method is insensitive to recent uranium losses. However, the sedimentary age of the rocks studied must be known, and it must be assumed that the rocks formed a closed U-Th-Pb system until onset of weathering, and that thorium and lead are immobile in the weathering zone. Generally, the investigated conglomerate samples did not show closed U-Th-Pb system behaviour and this made it necessary to consider the age patterns for obtaining best estimates of the fresh-rock uranium contents.

The lead isotope method was tested on fresh conglomerate samples which did not suffer uranium losses and the calculated uranium values were found to be in good agreement with directly measured uranium contents. For a number of weathered samples the calculated fresh-rock uranium tenor indicates the occurrence of supergene uranium losses which are of the same magnitude as the uranium losses caused by experimental leaching of fresh conglomerate samples.

In addition, calculated fresh-rock uranium contents of weathered conglomerates were compared with the uranium contents of fresh samples collected from the identical sedimentary horizon as the weathered samples. These comparisons showed close agreement of the values.

It is concluded that uranium redistribution in the secondary environment is not governed by systematic processes. This means that the original uranium contents of weathered conglomerates cannot be estimated by simple extrapolations using gamma- and alpha-spectrometric methods. Lead isotope techniques proved to be the only method yielding usable estimates of the fresh-rock uranium contents of weathered conglomerates. The method, albeit expensive, is recommended for exploration purposes as it might help to cut drilling costs in deeply weathered terranes underlain by potentially uranium-bearing conglomerates.

ACKNOWLEDGMENTS

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