APPENDIX 4 INTRAVAL (88)4

D R A F T

ALLIGATOR RIVERS ANALOGUE PROJECT

KOONGARRA URANIUM DEPOSIT

GENERAL BACKGROUND DESCRIPTION

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KOONGARRA URANIUM DEPOSIT

GENERAL BACKGROUND DESCRIPTION

1. INTRODUCTION

A. Pilot Group Identification

At the INTRAVAL Coordinating Group meeting in Stockholm in October 1987, Peter Duerden was appointed as the Pilot Group Leader, with Cezary Golian of ANSTO as a member.

B. Experimental Location

The Alligator Rivers region of the Northern Territory of Australia is located about 200 km east of Darwin, with the Koongarra uranium ore deposit being approximately 25 km south of Jabiru, the principal town. There are regular airline services from Darwin to Jabiru and sealed highways link Darwin with Jabiru with Koongarra, excepting the last 10 km (Figure 1).

C. Overall Objectives

The objective of the Alligator Rivers Analogue Project is to assist in the long-term prediction of the rate of transport of radionuclides through the geosphere. Specific aims will be to evaluate the significance of processes which may play an important part in radionuclide transport and to establish a radiochemical data base which may be used to validate radionuclide transport and geochemical modelling programs.

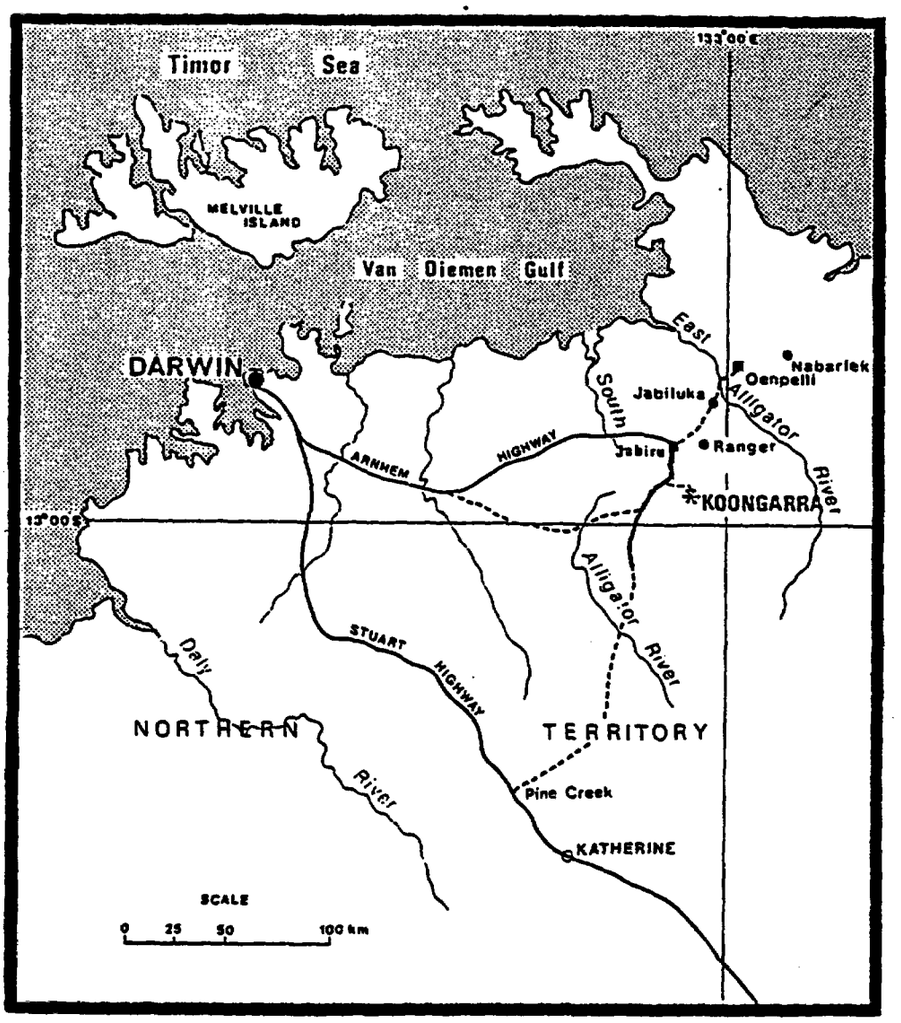

FIGURE 1 Location of the Koongarra uranium deposit.

2. BACKGROUND INFORMATION

(Extracts from 'A Geochemical Study of the Koongarra Uranium Deposit, NT, Australia, A. A. Snelling, PhD Thesis)

A. The Koongarra Uranium Ore Deposit

The Koongarra Mineralisation

Uranium mineralisation at Koongarra occurs in two distinct but clearly related orebodies, separated by about 100 m of barren schists (Figure 2). Both orebodies strike and dip broadly parallel to the Koongarra Reverse Fault, the footwall to the ore zone (Figure 3). Primary mineralisation is largely confined to quartz-chlorite schists immediately above the fault zone and a graphitic quartz-chlorite schist forms a distinctive hanging wall unit. At the northern end of the No. 1 orebody, and in the No. 2 orebody, mineralisation persists into the overlying garnetiferous quartz-muscovite-chlorite schists. The geology of the deposit has been described by Foy and Pedersen (1976) and Pedersen (1978).

The more south-westerly of the two orebodies, the No. 1 orebody (Figure 2), has a strike length of 450 m and persists to a depth of about 100 m. Secondary uranium mineralisation is present from the surface down to the base of weathering some 25 m down and forms a tongue-like body of ore grade material dispersing downslope for about 80 m to the south-east (Figure 3). There is also some dispersion of secondary uranium mineralisation within the main fault. In cross-section, the primary ore zone consists of a series of partially coalescing lenses, that have the appearance of being stratabound (see Figure 3). The width of the primary ore zone averages 30 m at the top of the unweathered schists, tapering out along strike, and down dip to about 100 m below surface. The strongest mineralisation, with most assay values in excess of 1% U_3U_8 (Foy & Pedersen, 1976) is over a thickness of several metres just below the graphitic hanging

wall schist unit. This high grade ore is persistent both along strike and down dip. Mineralisation of varying grades, however, occurs down through the schist sequence and along minor fractures. Closer to the footwall fault breccia the ore is of lower grade, is more sporadic, and tends to fade more rapidly with depth. Some lenses of ore are present within and just above the main brecciated zone, but they die out completely in areas of brecciated arenites of the Kombolgie Formation.

In the No 2 orebody, mineralisation occurs over a strike length of about 100 m and persists down dip to at least 250 m (Figure 4). There is little indication of secondary uranium mineralisation in the weathered schist zone as the zone of weathering has not yet intersected the top of this orebody to produce a zone of secondary uranium mineralisation like that in the No. 1 orebody. In cross-section the ore zone tends to be oval in shape, and is made up of a series of partially coalescing lenses. Ore grades are generally lower than in the No. 1 orebody, but the mineralisation is present over greater widths.

One of the most important features of uranium mineralisation at Koongarra is the occurrence of abundant secondary minerals, principally within the secondary dispersion fan above the No. 1 orebody but also to a lesser extent in the primary ore zones.

These secondary uranium minerals are noted for their variety of brillant colours, so their presence is easily recognised both in surface samples and in drill core.

Even cursory examination of drill-core shows that much of the secondary mineralisation is derived from in situ oxidation and alteration of pitchblendes, and by remobilisation of uranium. These processes therefore represent the latest stage in the genesis of the deposit as we know it today,

processes which may still be in operation. Uranyl phosphates are found in the tail of the secondary dispersion fan, while uranyl silicates, with only one exception, are either in the primary ore zone below, or at the interface between the two.

In the No. 2 orebody, which has no secondary dispersion fan, only uranyl silicates have been found.

The differences in distribution and style between uranyl silicates and phosphates strongly suggests a two stage process for their development under different physico-chemical conditions. The first stage was perhaps the in situ alteration of pitchblendes to form uranyl silicates associated with chlorite veining at depth within the primary ore zones of both the No. 1 and No. 2 orebodies. Subsequent intersection of the zone of surface weathering with the top of the primary ore zone of the No. 1 orebody only, has resulted in the partial alteration of both pitchblendes and uranyl silicates to form uranyl phosphates within the secondary dispersion fan.

In primary ore zones at Koongarra, where apatite (the source of phosphorous) is stable, the soluble uranyl silicates precipitated under slightly reducing and weakly acidic conditions with negligible to zero concentrations of phosphate and vandate in the groundwaters. In the weathered zone, however, the precipitation of relatively insoluble uranyl phosphates has been favoured by oxidising conditions and much higher phosphate concentrations (due to weathering of apatite) in the groundwaters. It is also possible to conclude that vanadium concentrations in the circulating fluids at Koongarra were very low compared to phosphorous because carnotite is so sparse in its occurrence in the weathered zone.

The Role of Groundwater Movements

Groundwater movements at Koongarra have been investigated by Australian Groundwater Consultants Pty Ltd and McMahon, Burgess and Yeates (1978), and their conclusions are shown schematically in Figure 5. The depicted groundwater paths indicate the source and direction of the groundwaters responsible for the oxidation, hydration and silicification of the pitchblendes and for the contemporaneous chlorite production, and of the groundwaters responsible for the weathering and dispersion of the primary ore zone. The distribution of secondary uranium minerals thus produced is also shown schematically in Figure 5.

Recharge of groundwaters to the weathered Cahill schists occurs via downflow parallel to, and in close proximity to, the reverse fault in both the underlying Kombolgie sandstone and the schists. Although the fault zone breccia was found to be practically impermeable, some water flows from the Kombolgie sandstone into the schists via cross fractures which offset the fault. Once in the schists groundwater flow is towards the south-east, away from the Mount Brockman escarpment. The weathered zone essentially acts as an aquitard or capping that prevents downwards percolation of surficial waters. Nevertheless there is a separate flow of surficial groundwaters within the weathered zone, again flowing downslope towards the south-east away from the Mount Brockman escarpment. Tritium dating of waters from below the weathered zone indicates that flow rates are very low (Australian Groundwater Consultants Pty Ltd & McMahon et al., 1978).

The weathered zone groundwaters are by nature, being essentially surficial, oxidising and this is supported by oxidation mineralogy in the weathered zone, including the occurrence of carnotite, and the non-occurrence of pitchblendes. Below the base of weathering, conditions are slightly reducing, particuarly in the presence of graphite

where pitchblende and sulphides are stable. Consequently, any dissolved uranium still present in the groundwaters may well have been reprecipitated as supergene pitchblende, thus further upgrading the high grade ore zone.

These movements of groundwaters responsible for oxidation, hydration and silicification of pitchblendes at depth and for contemporaneous chlorite production have apparently been active for over 350,000 years as this is the approximate age of the leading edge of the secondary dispersion fan. However, the alteration processes probably began at depth well before this. Removal of cover rocks, particularly the Kombolgie sandstone, is estimated to have taken place well in excess of a million years BP, ample time for alteration observed at depth to be produced. The reverse fault and its off-setting fractures must have developed even earlier still, as they have facilitated erosion of the cover rocks and later provided access for the penetrating groundwaters responsible for the pitchblende alteration. The present state of the No. 2 orebody provides a clear picture of what the No. 1 orebody would have been like prior to surficial weathering and attendant destruction of primary ore. Clearly once surficial weathering processes commenced to destroy pitchblendes and uranyl silicates producing uranyl phosphates in their place, lateral groundwater flow dissolved and dispersed the uranium down-slope in the direction away from the Mount Brockman escarpment (Figure 5). This process, which continues at the present time, has thus been responsible for producing the prominent dispersion fan above the No. 1 orebody.

Summary and Conclusions

(1) Chlorite-producing, low temperature groundwaters brought oxidising-hydrating conditions to the primary ore veins. In situ alteration of pitchblendes stabilised uranium-lead oxides and uranyl silicates following minor migration of dissolved uranium. The circulating groundwaters produced hematitic alteration as they moved across the reverse fault zone and into the host schists

below the orebodies. These processes occurred in a low pressure environment, probably after the earliest removal of cover rocks.

(2) Subsequent penetration of surficial weathering processes to the top of the No. 1 orebody produced prolific uranyl phosphates from the weathered and leached former pitchblendes and uranyl silicates under oxidising conditions.

(3) Contemporaneous and subsequent lateral groundwater movement through the weathered zone has dispersed uranium downslope to form the prominent secondary dispersion fan above the No. 1 orebody.

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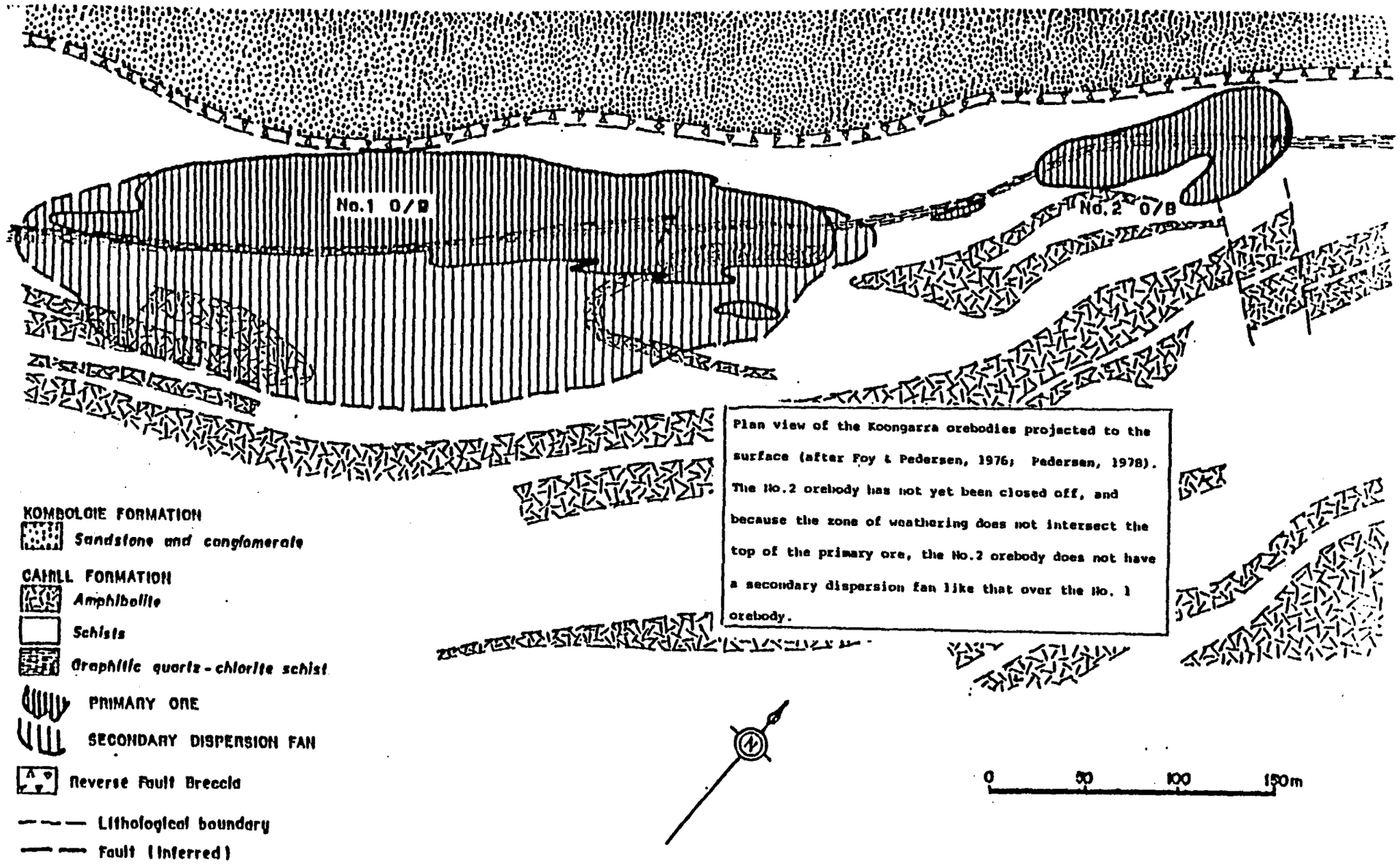
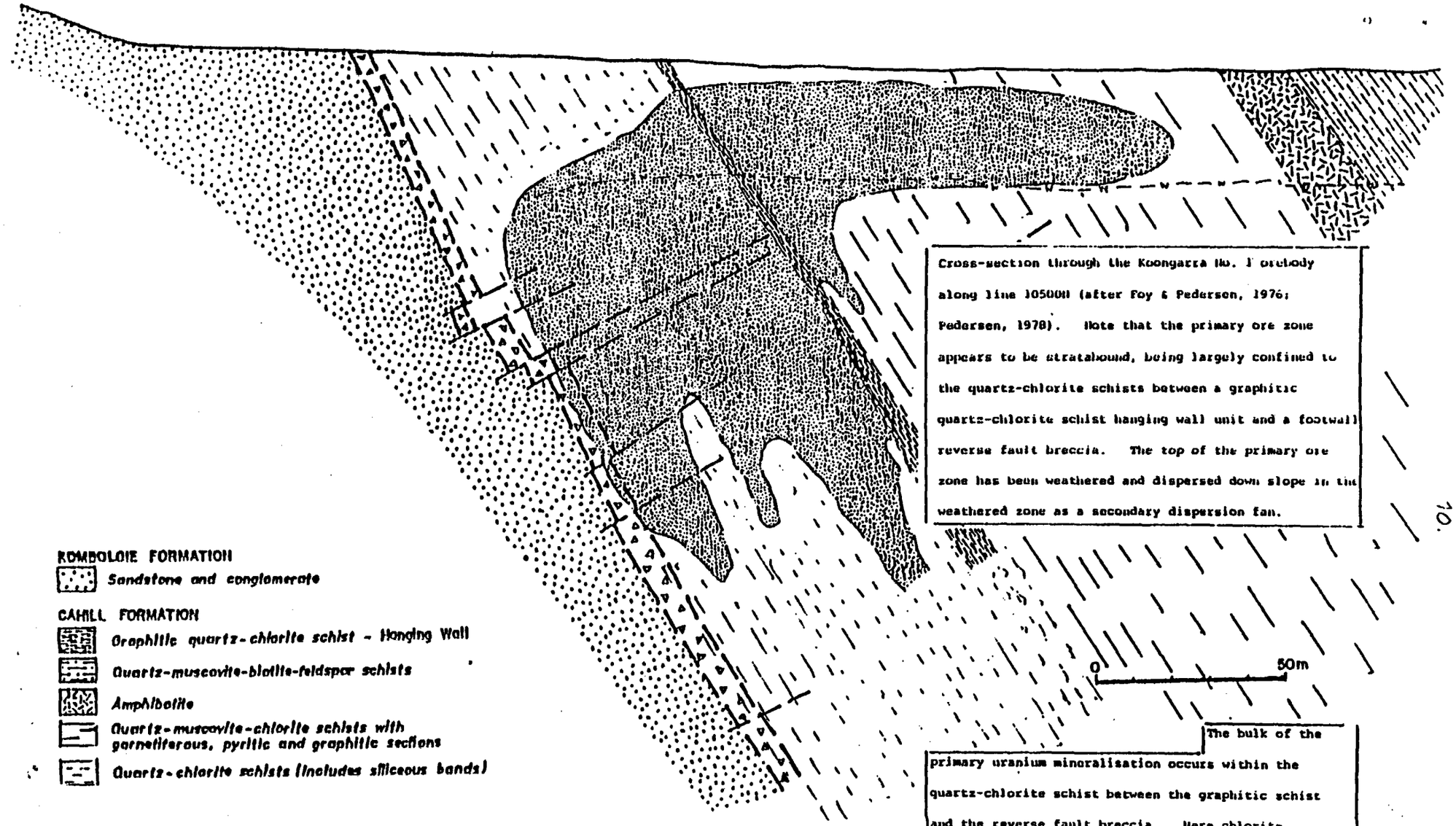


Plan view of the Koongarra orebodies projected to the surface (after Foy & Pedersen, 1976; Pedersen, 1978). The No.2 orebody has not yet been closed off, and because the zone of weathering does not intersect the top of the primary ore, the No.2 orebody does not have a secondary dispersion fan like that over the No. 1 orebody.

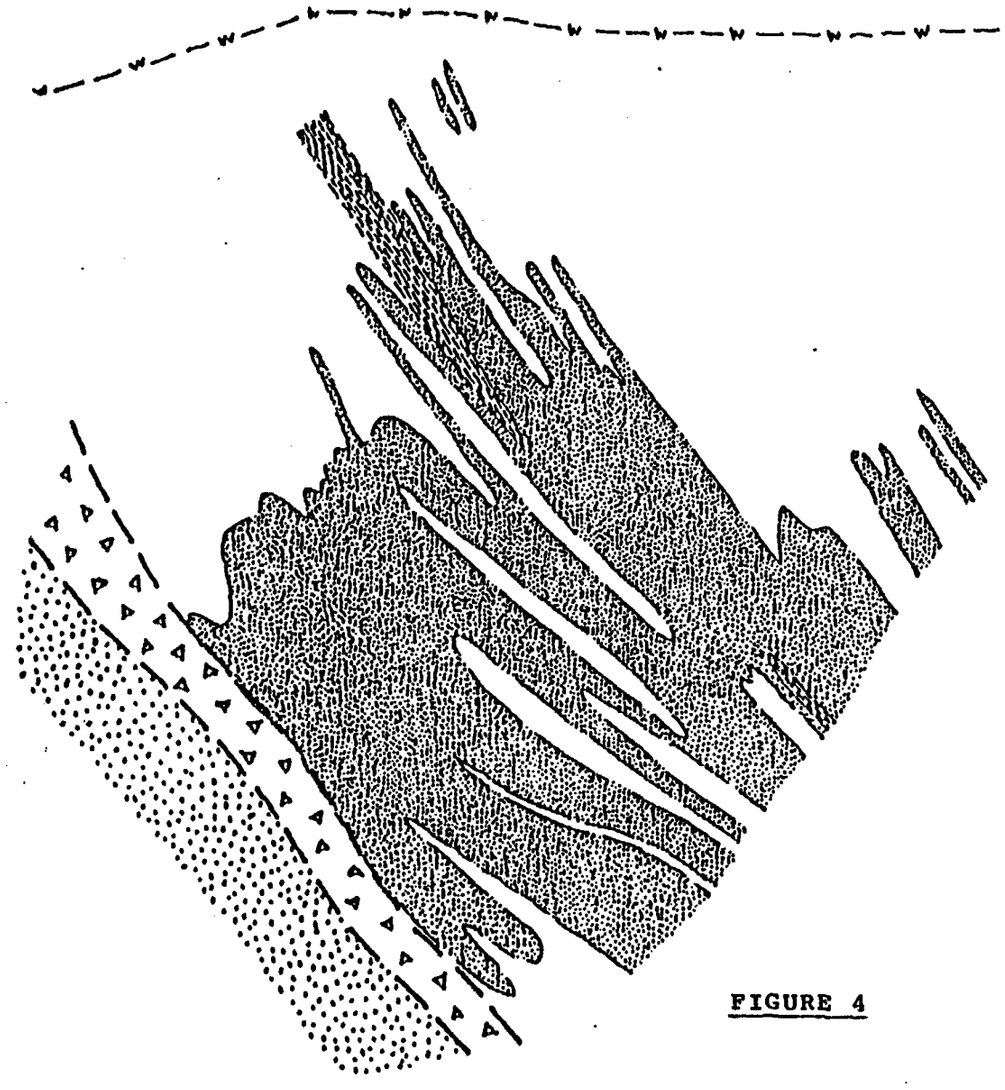
FIGURE 2



Cross-section through the Koongarra No. 1 orebody along line 10500N (after Foy & Pedersen, 1976; Pedersen, 1978). Note that the primary ore zone appears to be stratabound, being largely confined to the quartz-chlorite schists between a graphitic quartz-chlorite schist hanging wall unit and a footwall reverse fault breccia. The top of the primary ore zone has been weathered and dispersed down slope in the weathered zone as a secondary dispersion fan.

The bulk of the primary uranium mineralisation occurs within the quartz-chlorite schist between the graphitic schist and the reverse fault breccia. Here chlorite alteration is severest and no contained amphibolite units are recognisable. Siliceous bands are believed to represent former dolomite lenses.

FIGURE 3



Cross-section through the Koongarra No. 2 orebody along line 118000N (after Noranda Australia Limited, 1974). Note that the primary ore zone is deeper so that its top has not yet been intersected by the zone of weathering. Furthermore the ore zone extends above the graphitic hanging wall unit, and has not yet been closed off at depth.

0 50m

KOMBOLOIE FORMATION

Sandstone and conglomerate

CAHILL FORMATION

Schists and amphibolite

Graphitic quartz-chlorite schist - Hanging Wall

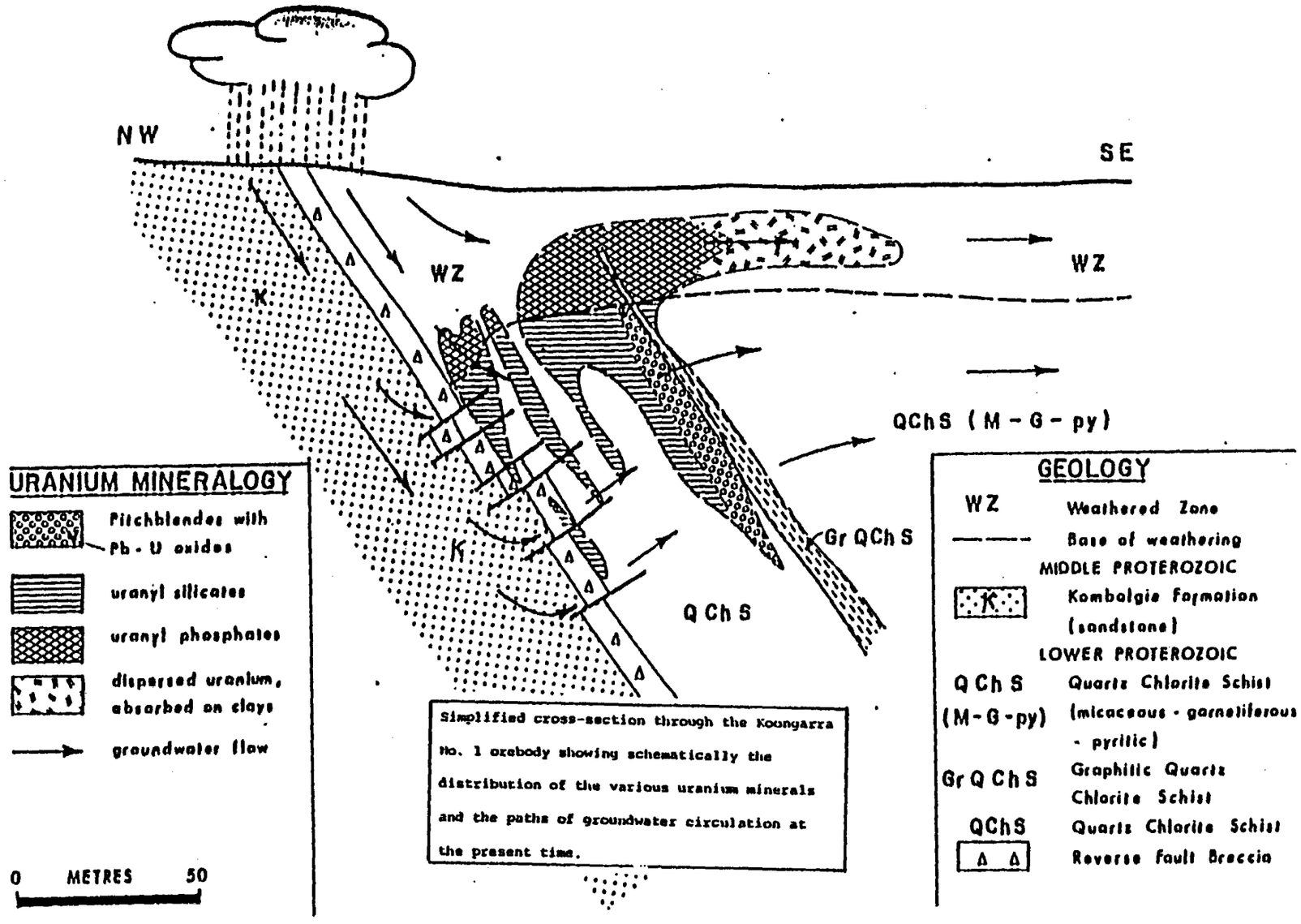
OREBODY

Reverse Fault Breccia - Footwall

Fault (Inferred)

Base of weathered zone

FIGURE 4



Simplified cross-section through the Koongarra No. 1 orebody showing schematically the distribution of the various uranium minerals and the paths of groundwater circulation at the present time.

FIGURE 5

B. Hydrologic Information

The study region, in common with much of northern Australia, has a monsoonal climate with almost all the rainfall occurring in a wet season between November and March.

General migration of groundwater is from the north, at the foot of a prominent south facing, escarpment, towards the south through the ore deposit with natural groundwater discharge taking place by evapotranspiration and direct discharge into a stream in the southern part of the site during the wet season. Rainfall records have been kept for the last 17 years and water levels in 61 wells have been monitored for various periods up to 7 years in duration since 1971.

Hydrogeologic characterization of the bedrock has been obtained from 24 drawdown and recovery tests and 50 water pressure tests. Five aquifer tests have also been made to help characterize the flow system in the bedrock.

However, further hydrogeologic work is planned in the early stages of the international Alligator Rivers Analogue Project to help determine possible trajectories and travel times. This work will be aimed to investigate whether the groundwater flow is primarily along fractures even in the highly weathered zone, and what is the general depth of transition between matrix flow and fracture flow. Connections, if any, between groundwater in the bedrock and groundwater in the surficial deposits will also be investigated. It is planned that approximately 10 further wells will be drilled for additional groundwater sampling and aquifer tests.

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C. Geologic Data

The geological data available is based on the mineralogical and uranium assay logs of 140 percussion holes and 107 drill cores in the immediate vicinity of the uranium deposit (Figure 6), together with data on over 300 backhoe pits and auger holes, geophysical surveys and geologic maps of surface exposures. The drill core material and pulp from the percussion holes is stored at the site and is available for examination. This material has been the subject of an extensive measurement program which studied the distribution of uranium and thorium series radionuclides in the region of the ore body.

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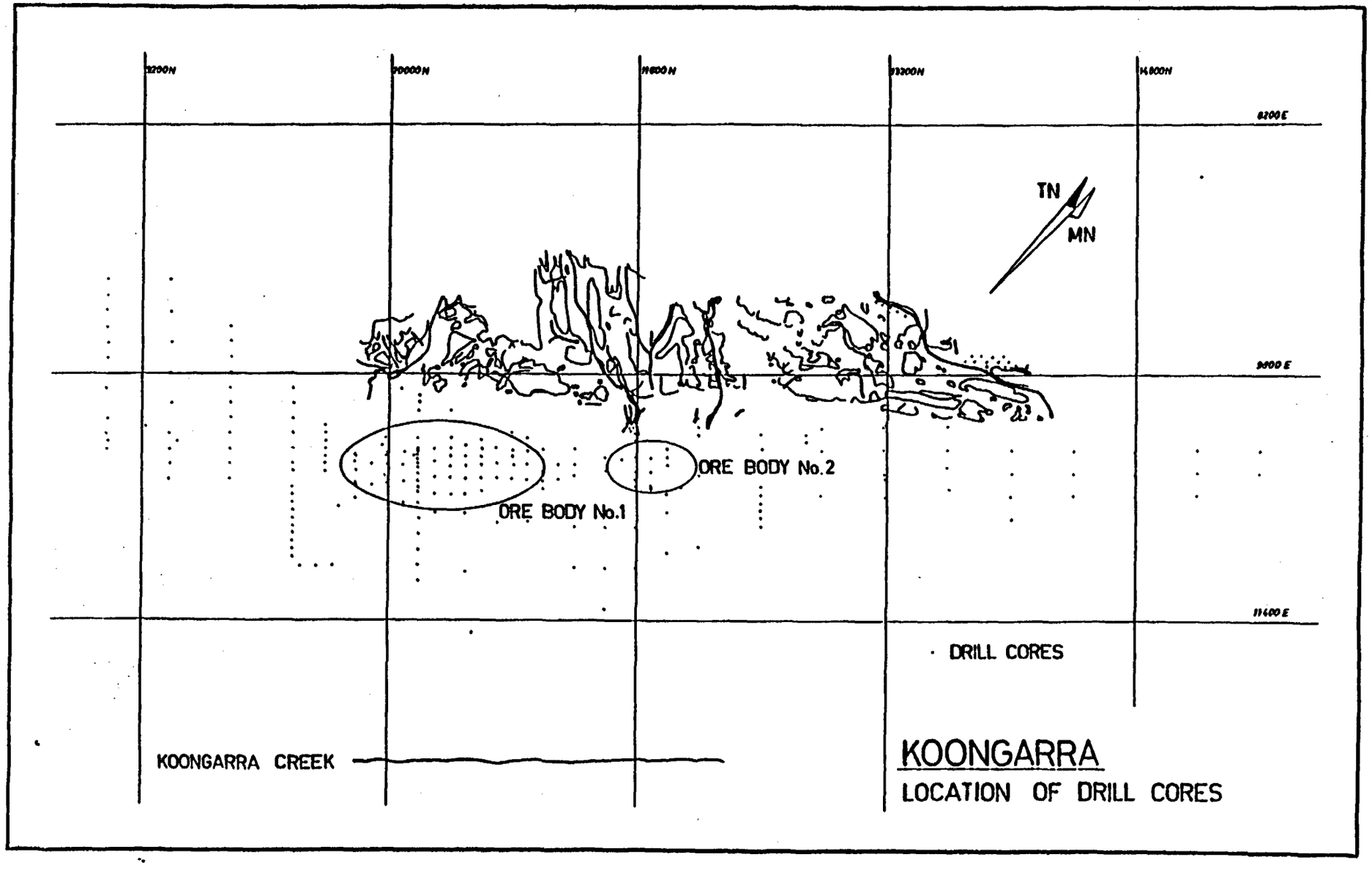
TN
MN
ORE BODY No.2
ORE BODY No.1
DRILL CORES
KOONGARRA CREEK
KOONGARRA
LOCATION OF DRILL CORES

FIGURE 6

D. Groundwater Chemistry

An extensive set of groundwater chemical data has been accumulated from mining investigations and analyses by a number of researchers. As a result, data is available for more than 70 boreholes in the vicinity of Koongarra. This data is of uneven quality and in some cases inconsistent or incomplete, however it is more than adequate to give a general understanding of the groundwater chemistry.

Data for groundwater from several boreholes near the No. 1 oebody is given in Table 1. These groundwaters are characterised by relatively high levels of Mg^{2+} (approximately 20 mg/L) due to the abundance of magnesium in the chloritised mine series rocks which accompany uranium mineralisation. The dominance of Mg^{2+} amongst the cations is reflected by the parameter NMg [where NMg = Mg/[Mg + K + Na + Ca], all being expressed in mg/L). The value of NMg is approximately 0.9 for these groundwaters. The main anion is bicarbonate (HCO_3^-), and the speciation of uranium is probably dominated by uranyl carbonate complexes, which are known to be mobile in natural environments. Fluorine complexes are important in uranium speciation only in acidic waters below the pH of Koongarra groundwaters. Phosphate is a strong uranium complexant and may be significant even at the low levels reported in Table 1.

Redox and pH measurements indicate that dissolved uranium in the groundwater is present in its oxidised U(VI) state which is much more mobile than uranium (IV). The pH of Koongarra waters (mostly between 6.5 and 7.5) is within the range of maximum uranium adsorption and minimum solubility of uranyl minerals. Sorption processes can limit dissolved uranium concentrations to much lower levels than would be expected on the basis of solubility constraints alone.

Dissolved thorium in the groundwater is present at extremely low levels, despite the presence of large amounts of ^{230}Th produced by radioactive decay of ^{234}U. Thorium is very insoluble and tends to adsorb almost immediately onto mineral surfaces when produced by radioactive decay of ^{234}U dissolved in the groundwater. The $^{230}Th/^{234}U$ ratio in groundwaters near the centre of the orebody is about 0.00002, whereas in the solid phase this ratio is near unity.

The pH of Koongarra groundwaters rises from around 5.5 in the Kombolgie sandstone to approximately 8.0 in the dolomites which outcrop to the south-west of the deposit. There is a general increase in pH as the water moves through the flow system. The pH/Eh conditions of groundwaters from KD1 (in the Kombolgie sandstone up-gradient of the deposit) and PH 49 (near the centre of the orebody) are shown in Figure 7. The redox potential is clearly too high for reduced uranium species to be present in the groundwater. On the basis of these measurements it would be predicted that any iron should occur as the highly insoluble Fe^{3+} species. However, measured iron levels are well above the solubility Fe^{3+}, indicating the presence of iron in its reduced (Fe^{2+}) state. This suggests possible mixing of reduced groundwaters carrying Fe^{2+} with more oxidising near-surface groundwaters during sampling.

On the basis of the groundwater chemistry, the area may be divided into several distinct zones. Figure 8 shows their approximate locations, and Table 2 lists the respective data.

<u>Zone I</u>

The Kombolgie sandstone escarpment is, along with direct infiltration, a source of groundwater entering the flow system of the deposits. This flow occurs both at depth across the fault and also closer to the surface in the weathered zone. The pH of Kombolgie groundwater is

slightly acidic (approximately 5.5-6.0), and the waters are relatively oxidising. Kombolgie groundwaters moving across the fault zone below the base of weathering have caused in situ alteration of uraninite veins to secondary minerals. Uraninites furthest from the fault in the high grade zone below the graphitic hanging wall remain unaltered.

Zone II

The groundwaters from boreholes in the orebody are markedly different. The pH is higher, reflecting the general rise in groundwater pH as it moves through the system. There is a much higher level of magnesium, which dominates the cations and the conductivity, greater than 200 µs/cm is over twice that of Kombolgie waters. The level of bicarbonates is highest within this zone (HCO_3^- is the major anion in all sampled groundwaters). The high levels of Mg^{2+} in the groundwaters have been attributed to the abundance of magnesium in the chloritised mine series rocks which accompany uranium mineralisation. Magnesium, expressed as a percentage of total major soluble cations (Ca, Mg, Na, K) comprises 87% in chloritised mine series rocks, but only 25% in unchloritised mine series rocks (Giblin and Snelling, 1983).

Zone III

Little information is available about groundwaters from the No. 2 orebody. This orebody is much deeper than the No. 1 orebody and no dispersion fan is present. It may influence the chemistry of down-gradient groundwaters.

Zone IV(E)

Groundwaters moving through the Cahill formation along strike and to the north-east of the deposit closely

resemble those in the ore zone, but Mg tends to be slightly less dominant (NMg is also decreased by the presence of significant levels of Ca in some groundwaters from this zone). Conductivity remains high but is lower than within the orebody, and uranium is present at lower levels.

Zone IV(W)

The groundwater along strike south-west of the deposit is quite different to the mineralised zone. These waters have very low Mg levels, and low pH and HCO_3^-. This suggests groundwaters in this region may not be related to the orebody and may be fast-moving groundwaters originating from the Kombolgie (which has a similar chemistry) or possibly recent recharge from infiltration.

Two zones adjacent to the orebody might be considered as being down-gradient to the mineralisation.

Zone V

In the first of these zones to the south of the orebody the groundwater chemistry is similar to the ore zone. Magnesium levels remain high possibly reduced slightly by dilution from infiltration, and NMg is almost as high as for the orebody. Bicarbonate levels are fairly high, as in the ore zone, and the pH is somewhat higher following the usual flow trends.

Zone VI

The second zone adjacent to and east of the orebody is not well represented in the data set. However the data is sufficient to show that the groundwaters are quite different to the mineralised zone. Magnesium levels are relatively low, as are the total cations and

conductivity. The pH is higher than in the orebody and the characteristics of the water suggest a component of recent recharge, although it is conceivable that a direct flow path from the Kombolgie exists. This could well be a zone of mixing and may also include a component from hydrogeochemical dispersion down-gradient of the No. 2 orebody.

Zone VII

This is a zone of dolomites which probably has little influence on the immediate vicinity of the orebody, being some distance away. Both Mg and Ca levels are high, and approximately equal as expected in this rock type.

In summary, it appears that the sequence of groundwater chemistry relevant to the deposit initially involves a Kombolgie type groundwater moving into the Cahill formation. This leads to the distinct groundwater type found within the orebody and also along strike to the east. Groundwater flow away from the deposit appears most likely to occur in a southerly direction where groundwater character closely resembles the groundwater of the mineralised zones.

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TABLE 1

GROUNDWATER CHEMISTRY - KOONGARRA

	pH	Eh (mv)	D.O (ppm)	Cond (µs/cm)	K (mg/L)	Na (mg/L)	Mg (mg/L)	Ca (mg/L)	Si (mg/L)	Fe (mg/L)	Mn (mg/L)	HCO_3^- (mg/L)	Cl^- (mg/L)	SO_4^{2-} (mg/L)	F^-* (mg/L)	PO_4^-* (mg/L)	U (µg/L)	NMg
KD 2	6.95	+175	0.6	165	0.65	1.69	15.5	0.92	12.5	1.14	0.04	103	3.0	<0.5	0.3	0.025	0.08	0.90
PH 55	6.8	+140	0.5	175	1.07	1.58	17.1	1.28	12.5	0.77	0.07	116	2.8	<0.5	0.3	0.015	0.40	0.90
PH 14	6.8	+190	0.3	250	0.68	1.42	20.8	1.90	9.4	0.29	0.28	128	5.6	0.8	0.4	0.015	150	0.90
PH 49	6.7	+175	0.4	240	0.58	1.12	23.5	1.6	8.3	0.40	0.13	132	3.9	0.7	0.4	0.010	181	0.93
PH 56	6.8	+170	0.5	185	0.39	0.75	18.0	1.9	10.5	0.31	0.13	109	5.0*	0.5	0.3	0.005	11	0.91

* Cl^- for PH 56 and all PO_4^- and F^- levels from Giblin and Snelling (1983).

All other results from 1986 sampling program.

TABLE 2

GROUNDWATER - CHEMICAL ZONES

Zone Number	Zone	Mg mg/L	NMg %	HCO_3^- mg/L	$NHCO_3^-$ mg/L	Median pH	Total Cations meq/L	Conductivity µs/cm
	Surficial	1	50	2	33	5.5	0.1	20
I	Kombolgie	6	66	40	88	6.2	0.7	90
II	No 1 Orebody	25	93	130	95	6.6	2.2	220
IV(E)	North east along strike of orebody	18	78	114	94	6.7	1.9	150
IV(W)	South west along strike of orebody	3	64	20	71	6.0	0.4	60
V	Down-gradient, south	17	89	85	92	7.0	1.6	130
VI	Down-gradient, east	7	67	42	85	7.1	0.9	70
VII	Dolomite	16	59	140	95	7.5	2.2	230

NOTES: (1) The parameters NMg and $NHCO_3$ are defined as follows:

$$NMg = \frac{Mg}{Mg + Ca + Na + K} \quad \text{(expressed in meg/L)}$$

$$NHCO = \frac{HCO_3}{HCO_3 + SO_4 + Cl} \quad \text{(expressed in meg/L)}$$

(2) Boreholes used in this compilation:

Zone I - KD 1; Zone II - PH 49; Zone IV(E) - PH 118, PH 39, PH 43; Zone IV(W) - PH 162, PH 147, PH 156, KOA; Zone V - PH 73, PH 80, PH 84, PH 88, PH 93, KD 2; Zone VI - PH 94, PH 95, PH 96; Zone VII - KIO 2.

The surficial sample is from the "pilot well point" (approximately 200 m south of KD 3).

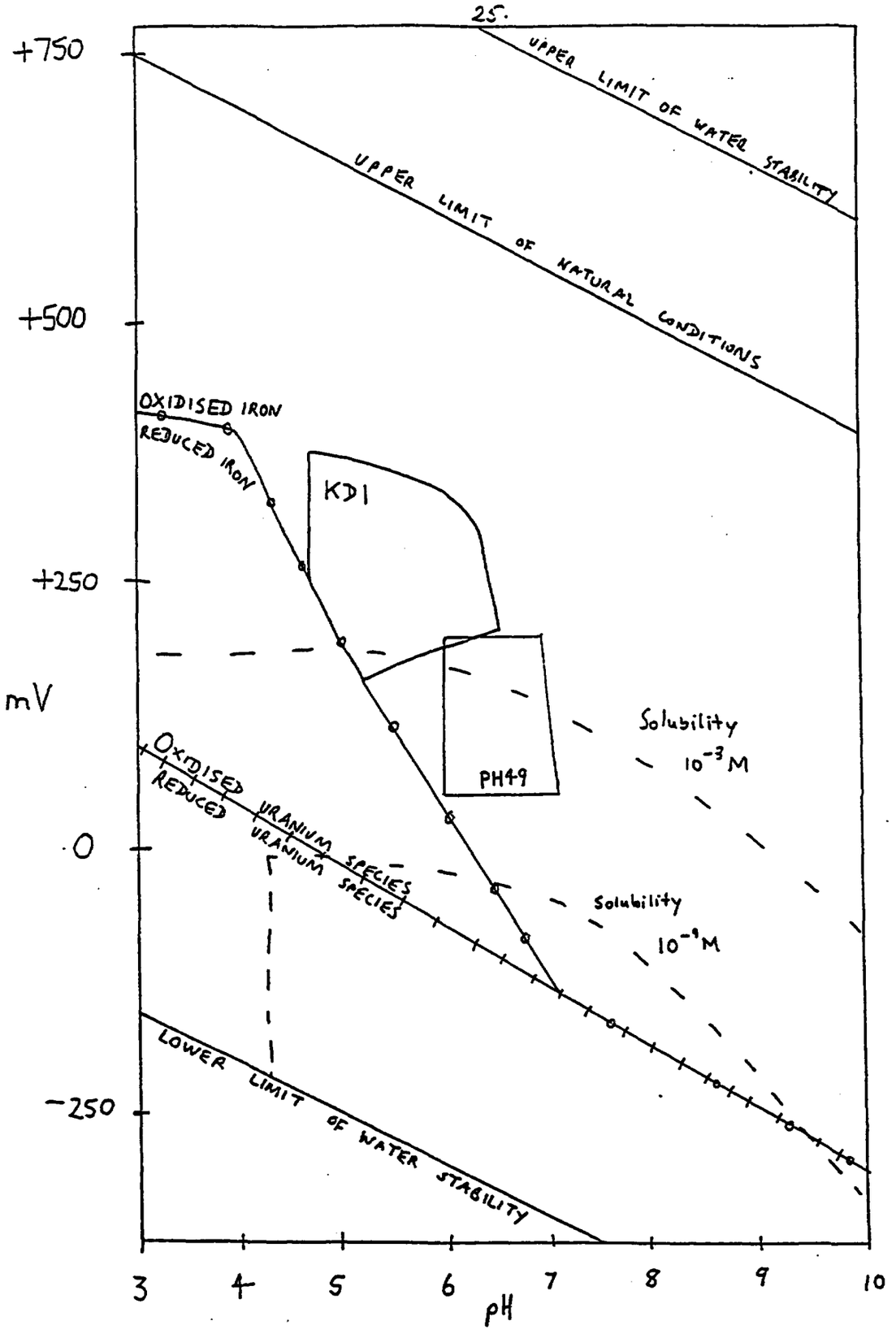

FIGURE 7 Eh/pH (Pourbaix) diagram showing KD 1 and PH 49 groundwaters.
Contours of uranium solubility and approximate boundaries between oxidised and reduced species of iron and uranium are shown.

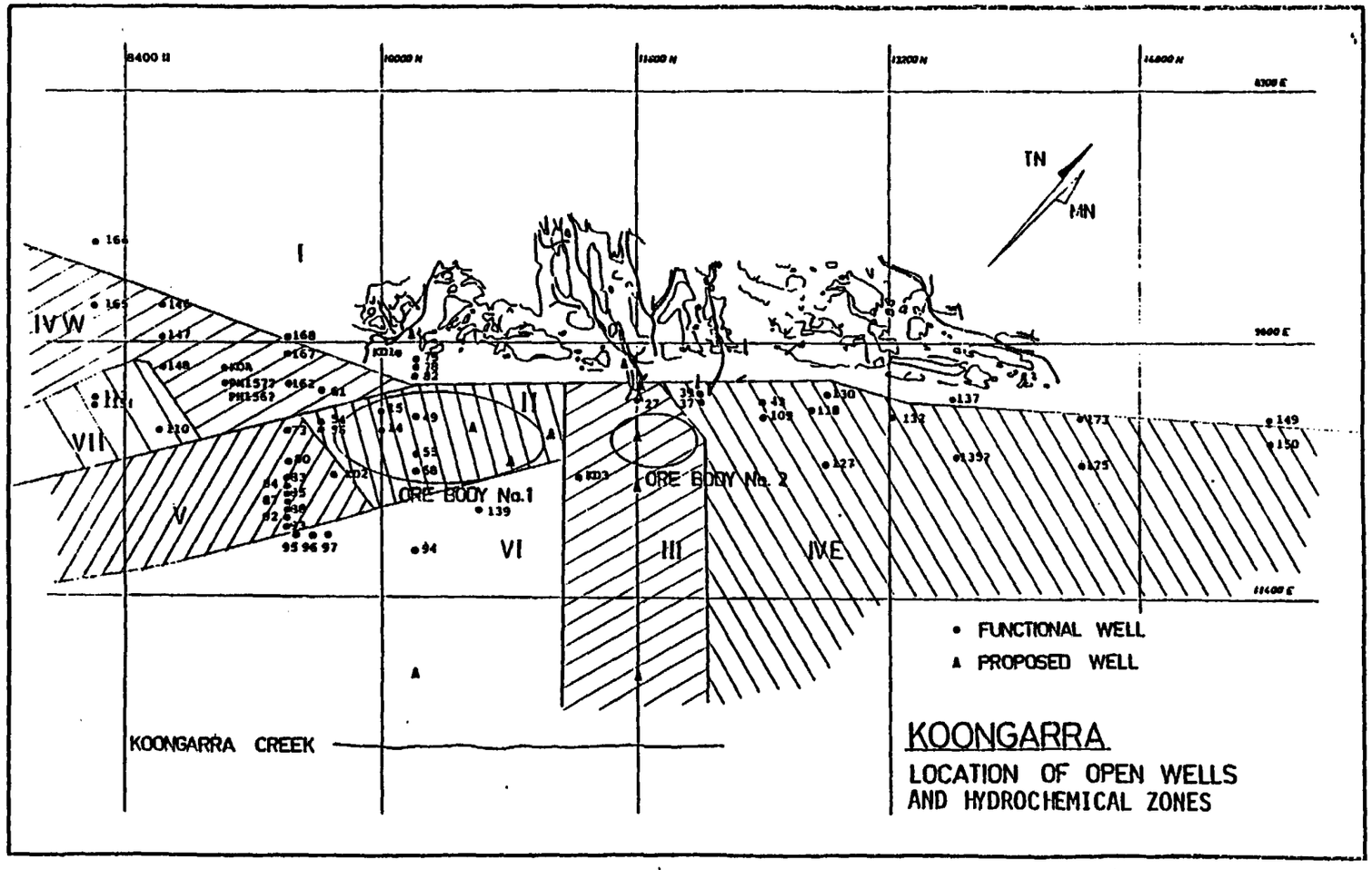
TN
MN
I
IVW
VII
V
VI
II
III
IVE
ORE BODY No.1
ORE BODY No. 2
KOONGARRA CREEK
• FUNCTIONAL WELL
▲ PROPOSED WELL
KOONGARRA
LOCATION OF OPEN WELLS
AND HYDROCHEMICAL ZONES

FIGURE 8

E. Summary of Soil Horizons

The mineralogical properties of the soil horizons are illustrated in Figure 9 and briefly described below.

D Horizon

D_R The major constituents of the consolidated rock are

i) muscovite 2M, or more rarely, 1M,
ii) di-trioctahedral and/or trioctahedral chlorite IIb,
iii) quartz.

Anatase and rutile are often found.

The chlorites are penninites and clinochores and are rich in Si, with medium Mg and low Fe. Tetrahedral Al is high and the chlorites weather to vermiculite/beidellite.

In addition to the D_R horizon, which comprises the major part of the D horizon, four non-continuous zones of alteration are also observed in the upper part of the D horizon.

D_B Biotite is remnant in the quartz, muscovite, chlorite schist. Biotite weathers more readily than the other minerals and along with uraninite is an initially weathered mineral. Frequently seen to be releasing FeIII as it weathers.

D_V Vermiculite and related interstratified minerals have formed from biotite.

D_O FeII in chlorite has oxidised and the chloride X-ray diffraction ratios have altered.

D_A Argillised rock.

D Horizon

Chlorite has weathered to smectite, kaolinite and goethite.

B Horizon

FeIII reduced to FeII and leached in the B_2 horizon, leading to development of indurated laterite horizon (B_1) and nodules around quartz grains. Pedological chlorites develop and smectites weather.

A Horizon

Quartz, kaolinite, pedalogical chlorite, hematite, goethite and maghemite.

An important aspect of Umobilisation seems to be oxidation of UO_2IV to UVI before oxidation of FeII to FeIII. This means that the UVI can move without being adsorbed by the FeIII oxides and oxyhydroxides.

Figure 10 shows a representation of the weathering and groundwater flow where the transect from the unconformity is divided into three zones:

i) the kaolinitic zone (A_1 and B_1 horizons),
ii) the intermediate zone (B_2, B_3, C horizons),
iii) unweathered rock.

The deepened kaolinitic zone close to the unconformity can be clearly observed.

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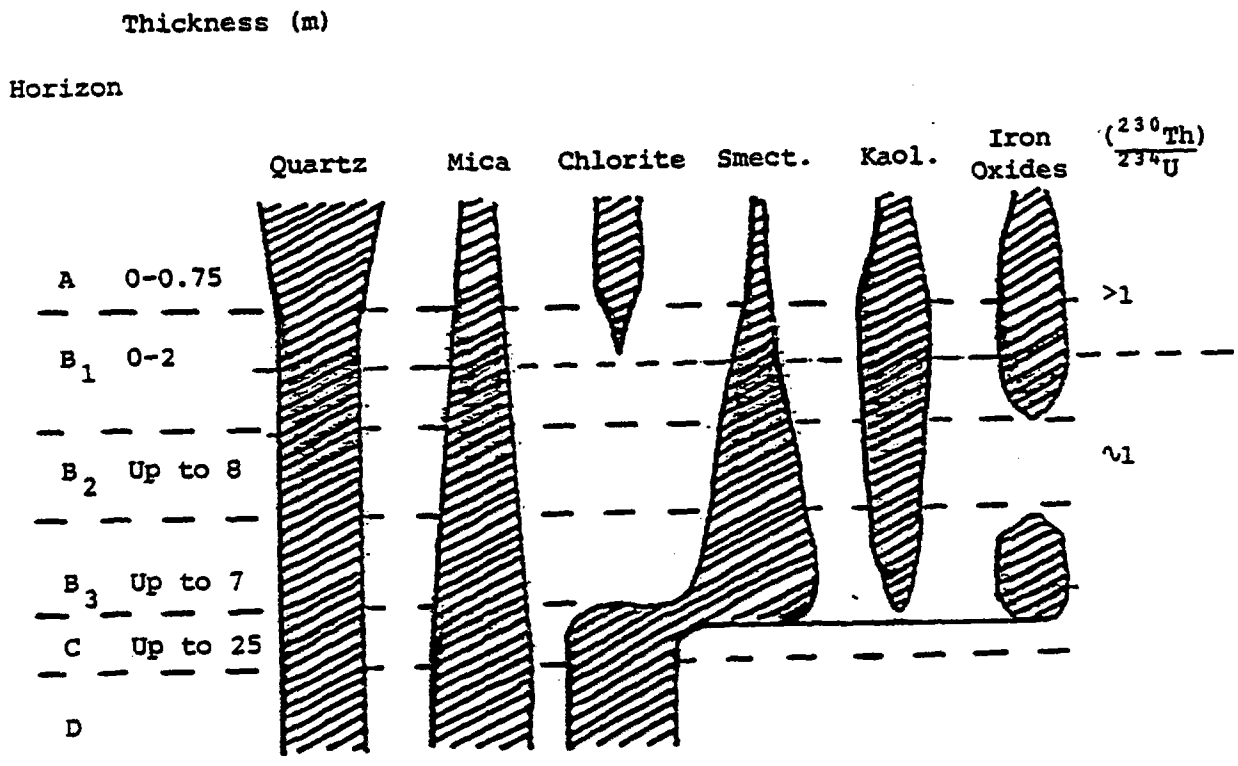

FIGURE 9 Weathering Pattern at Koongarra.

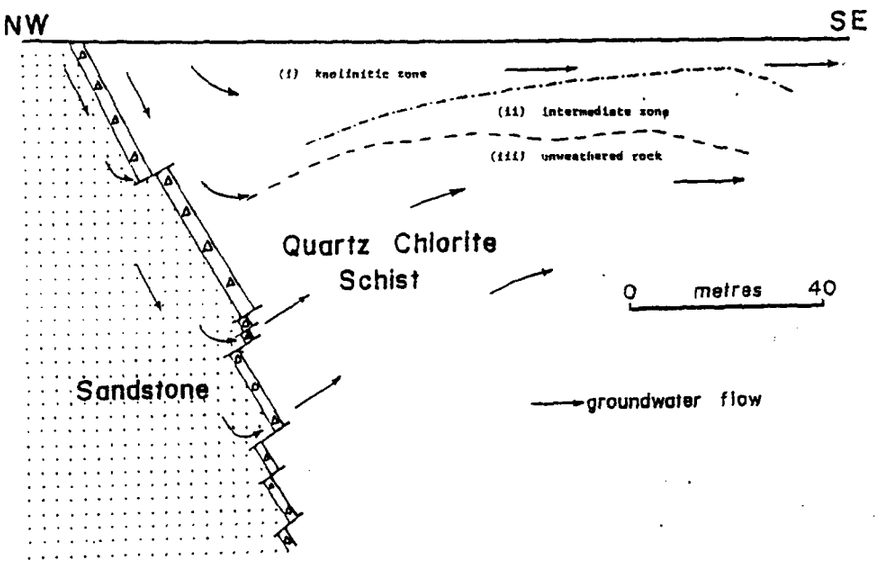

FIGURE 10 Diagramatic representation of Koongarra showing weathering and groundwater flow.

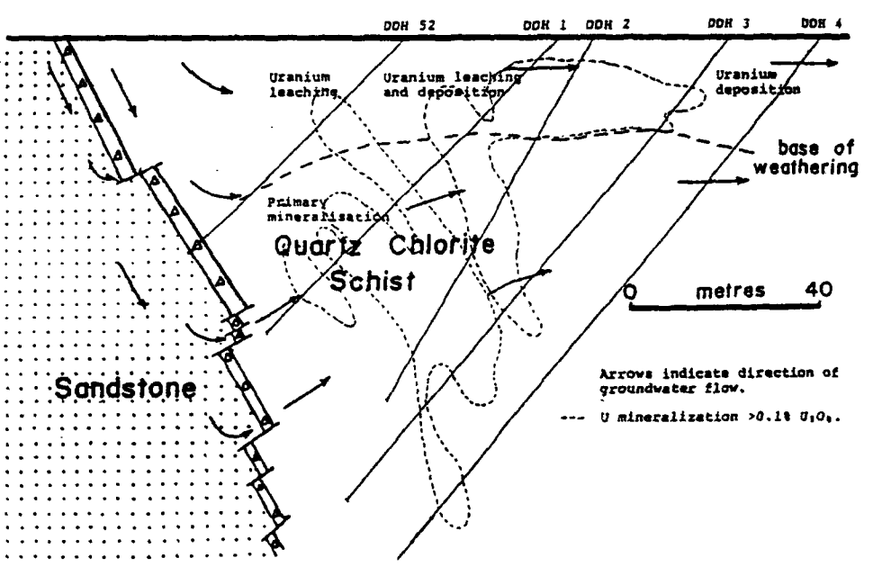

FIGURE 11 Representation of Koongarra showing areas of leaching and accumulation

F. Uranium Series Disequilibria

An extensive study of the distribution of uranium. thorium and radium isotopes at the Koongarra uranium deposit has identified well-defined areas of uranium leaching and accumulation within the 30 m deep weathered zone (Figure 11). The data accumulated so far has the potential to enable calculation of the extent and timescale of uranim migration by the appliction of mathematical modelling techniques. A more extensive series of measurements would clearly be of benefit.

In estimating the extent and direction of uranium migation, the assumation is made that the geochemical mobility of thorium and protactinium are negligible compared to that of uranium. This is a commonly used assumption and there is every indication that it is valid in the present context. (The activity of thorium in Koongarra groundwaters was found to be a few orders of magnitude below that of uranium.) The deviation in $^{230}Th/^{234}U$ and $^{231}Pa/^{235}U$ activity ratios from unity can therefore be used to ascertain the recent leaching or depositional behaviour of uranium in a particular location.

An extract from the results of a series of measurements of $^{234}U/^{238}U$, $^{230}Th/^{234}U$ and $^{226}Ra/^{230}Th$ activity ratios is shown in Table 3. The following trends were evident:

(a) Measurable disequilibria were largely confined to the weathered zone.

(b) The region of high $^{230}Th/^{234}U$ ratios correspond to what Snelling (1980) described as a "previous upward extension of the primary orebody".

(c) The region of low $^{230}Th/^{234}U$ ratios corresponded to the so-called 'secondary dispersion fan', described by Snelling (1980) as comprising secondary uranium mineralisation, mainly uranyl phosphates, and uranium dispersed onto clays.

(d) The $^{230}Th/^{234}U$ and $^{234}U/^{238}U$ activity ratios tended to deviate on the same side of unity in individual samples, i.e. either they were both greater than or both less than unity. This indicated either that ^{238}U was more mobile than ^{234}U in this system, or that a multi-stage uranium mobilisation had occurred. There is overwhelming evidence for the former, despite the fact that in most geochemical systems, ^{234}U is in fact the more mobile.

Figure 12 shows a plot of $^{234}U/^{238}U$ vs $^{230}Th/^{238}U$ activity ratios ($^{230}Th/^{238}U$ is the product of $^{234}U\backslash^{238}U$ and $^{230}Th/^{234}U$ activity ratios). A similar diagram was used by Osmond et al. (1981) to represent activity ratio patterns in frontal systems. It can be seen that the points plot in two main regions:

1) $^{238}U < {}^{234}U < {}^{230}Th$, corresponding to leaching of uranium with preferential leaching of ^{238}U.

2) $^{238}U > {}^{234}U > {}^{230}Th$, corresponding to accumulation of uranium that is depleted in ^{234}U.

An additional parameter of great value is the $^{234}Pa/^{235}U$ activity ratio, which can be derived with reasonable confidence from the $^{227}Ac/^{235}U$ ratio, measurable by alpha spectrometry. Table 4 compares the measurements of various activity ratios in individual samples. Bearing in mind that the half-life of ^{231}Pa is about half that of ^{230}Th, it can be seen that there is reasonable concordancy between the measured $^{230}Th/^{234}U$, $^{227}Ac/^{235}U$ and $^{23}Pa/^{235}U$ activity ratios. In future work, simultaneous measurement of the $^{230}Th/^{234}U$ and $^{231}Pa/^{235}U$activity ratios would be potentially valuable in predicting timescales of uranium mobilisation.

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Table 3

Uranium, thorium concentrations and activity ratios in core and pulp samples from the Koongarra Uranium Deposit.

Drill Hole	Length (m)	Depth (m)	Label	[U] (µg/g)		$^{234}U/^{238}U$	$^{230}Th/^{234}U$	[Th] (µg/g)		$^{226}Ra/^{230}Th$
DDH 52	0-1.5	0-1.1	a	637	(12)	1.358 (.010)	1.59 (.05)	11	(4)	0.68
	1.5-3	1.1-2.2	b	383	(12)	1.220 (.021)	1.63 (.06)	11	(4)	0.70
	2.74	1.9	-	414	(9)	1.158 (.016)	1.43 (.04)	13	(4)	---
	3.12	2.2	-	66	(2)	1.169 (.016)	1.23 (.04)	2	(1)	---
	3-4.6	2.2-3.2	c	89	(2)	1.170 (.017)	1.25 (.04)	2	(1)	0.75
	8.22	5.8	-	33.4	(.7)	1.140 (.020)	1.97 (.08)	22	(2)	1.21
	9.6	6.8	-	965	(20)	1.048 (.015)	1.12 (.04)	155	(9)	---
	9.75	6.9	-	168	(4)	1.108 (.008)	1.46 (.03)	31	(2)	0.98
	11.0	7.8	-	66.0	(1.2)	1.242 (.009)	---	---		---
	13.7-15	9.7-10.8	d	392	(8)	1.167 (.012)	1.14 (.03)	19	(3)	1.01
	16.8-18	11.9-12.9	e	692	(16)	1.165 (.020)	1.18 (.03)	35	(4)	---
	20-21.3	14.0-15.1	f	1970	(40)	1.029 (.009)	1.01 (.04)	< 20		0.87
	25.9	18.3	-	4100	(90)	0.994 (.009)	1.00 (.02)	21	(4)	0.76
	35.4	25.0	-	219	(5)	0.999 (.009)	0.99 (.02)	16	(1)	0.92
	39.6	28.0	-	319	(6)	1.066 (.008)	1.00 (.02)	26	(2)	---
	42.0	29.7	-	133	(3)	1.002 (.012)	---	---		---
	*49-50.3	34.5-35.6	g	213	(3)	1.004 (.016)	1.03 (.03)	24	(3)	1.12
DDH 1	3.0	2.2	-	641	(12)	1.120 (.013)	1.17 (.03)	15	(3)	---
	20-21.3	14.4-15.1	1a	815	(20)	1.034 (.008)	1.02 (.02)	< 20		---
	*41.5	29.3	-	29.7	(.5)	1.015 (.010)	0.94 (.04)	29	(1)	---
	*49-50.3	34.5-35.6	1b	214	(5)	1.001 (.018)	1.04 (.04)	18	(2)	1.14

Table 4

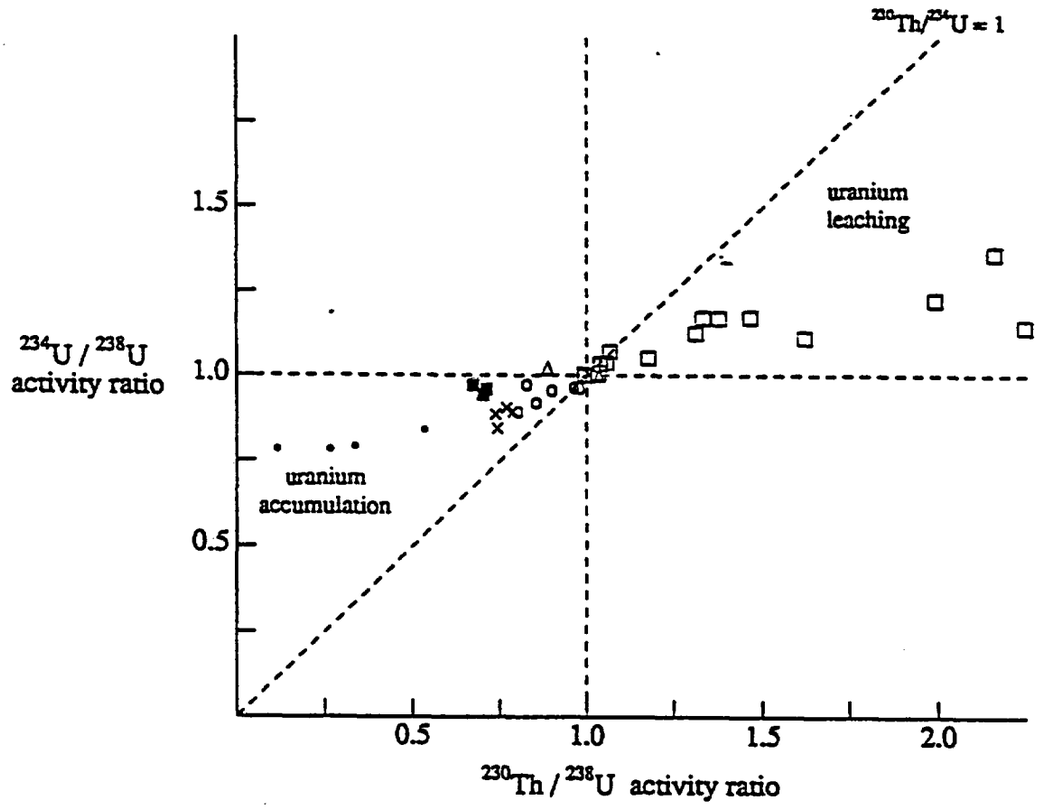
Comparison of $^{234}U/^{238}U$, $^{230}Th/^{234}U$, $^{227}Ac/^{235}U$ and $^{231}Pa/^{235}U$ activity ratios in selected samples from the Ranger and Koongarra Uranium Deposits.

Drillhole/depth		[U] (mg/g)	$^{234}U/^{238}U$	$^{230}Th/^{234}U$	$^{227}Ac/^{235}U$	$^{231}Pa/^{235}U$
S1/149	0.0m	1.24	1.136 (.018)	1.36 (.04)	1.22 (.05)	1.35
S1/149	3.0m	0.20	1.053 (.014)	4.43 (.12)	2.22 (.09)	1.98
DDH 52	0-1.5m	0.64	1.358 (.010)	1.59 (.05)	1.29 (.06)	—
DDH 4	27.4-29m	0.72	0.843 (.007)	0.88 (.03)	0.95 (.05)	—
Saleeite (a)		480	0.785 (.008)	0.334 (.009)	0.517 (.036)	—
Saleeite (b)		520	0.795 (.009)	0.416 (.012)	0.635 (.021)	—
Saleeite (c)		480	0.844 (.010)	0.632 (.015)	0.79 (.04)	—

Numbers in brackets indicate 1σ uncertainties. $^{231}Pa/^{235}U$ activity ratios were measured at the UKAERE laboratories, Harwell, UK. Drillhole S1/149 was located at the Ranger uranium deposit. Other samples were tken from the Koongarra uranium deposit.

Figure 12

$^{234}U / ^{238}U$ versus $^{230}Th / ^{238}U$ in koongarra core samples



□	DDH 52, DDH 1 samples	×	DDH 4, PH 55 samples
Δ	DDH 2 samples	■	PH 58, PH 60 and PH 89 samples
o	DDH 3 samples	•	saleeite crystals from DDH 64

Dashed lines intersect at point of secular equilibrium

Diagonal line represents locus of points at which $^{230}Th/^{234}U = 1$

G. Uranium Series Fractionation in Mineral Phases

An additional study has focussed on the phase distribution of uranium and thorium in the weathered zone. Apart from the 'accessible' uranium a high proportion (normally over 50%) appears to become incorporated into iron oxides where it is shielded from the effects of groundwater. A smaller portion (10-20%) associates with more weathering-resistant minerals. It appears that 'inaccessible' phases become enriched in ^{234}U and ^{230}Th at the expense of accessible phases. It would appear that

i) the uranium of the amorphous, but not of the crystalline iron, is directly accessible to groundwater;

ii) on the geological timescale, exchange occurs between the amorphous and crystalline iron;

iii) the bulk of the uranium and thorium is associated with the iron phases, whereas an excess of radium accumulates in the clay/quartz phase;

iv) the recoil effect leads to an accumulation of the daughter element in the clay/quartz; and

v) the greater mobility of ^{238}U at Koongara is a result of α recoil emplacement into less accessible sites. This should not be overlooked in any modelling study.

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H. Uranium Sorption

Laboratory sorption experiments have been carried out, using samples from Koongarra core DDH 52, and a simulated groundwater containing ^{236}U as the aqueous phase. Uranium adsorption was pH-dependent, with a maximum measured K_d of about 6 x 10^3 mL/g near pH 8.0 (Figure 13). During these experiments the desorption of ^{238}U into the groundwater was also measured.

The sample used for the adsorption experiments were subjected to a sequential extraction procedure to determine the partitioning of ^{238}U and ^{236}U amongst mineral phases. The results showed that some of the natural ^{238}U was found in the relatively mild extractants with most of the newly adsorbed ^{236}U. The presence of ^{238}U in both resistant and weakly bound phases, and the transfer processes occurring between the two, should clearly be taken into account when modelling uranium migration.

Distribution coefficients have also been measured for uranium and thorium adsorbed on natural particles in Koongarra groundwater. Uranium distribution coefficients were between 2.6 x 10^4 mL/g and 6.5 x 10^4 mL/g. Distribution coefficients for ^{230}Th were much higher and variable, ranging from 2.6 x 10^6 mL/g to 1.1 x 10^9 mL/g. The relatively constant distribution coefficients found for ^{238}U provide evidence that the distribution of uranium is governed by sorption equilibria. The difference between laboratory and field K_d values for uranium was tentatively attributed to the presence of highly sorptive coating minerals on the natural particles due to their prolonged exposure to groundwater. The much higher distribution coefficients and the extremely low groundwater levels measured for thorium confirm its immobility compared to uranium. The variability in thorium K_ds suggests that simple sorption models will be inadequate to model thorium movement.

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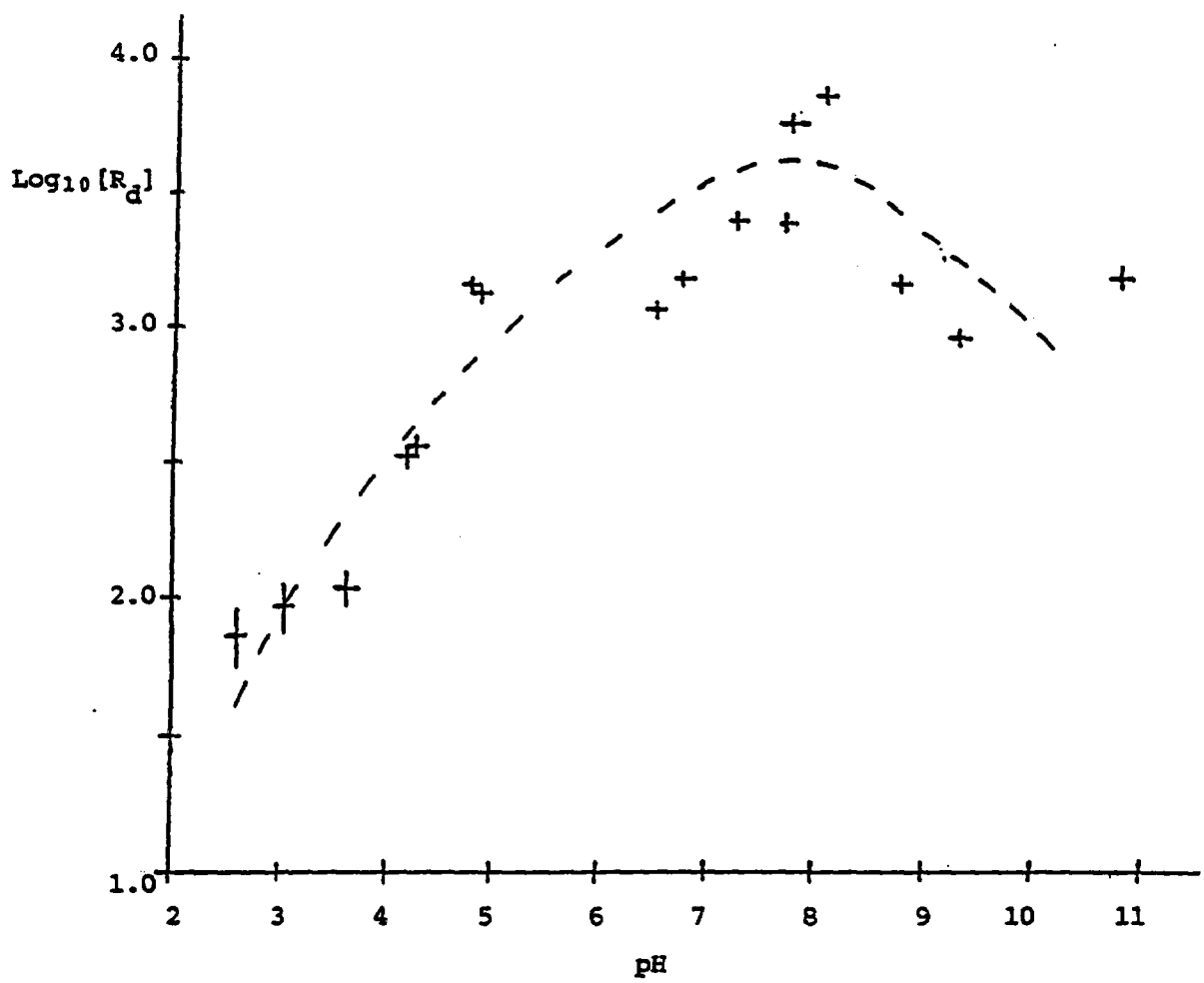

FIGURE 13 Distribution ratios for ^{236}U adsorption on Koongarra DDH 52 (15 m) sample.

3. PERFORMED AND PLANNED EXPERIMENTAL INVESTIGATIONS

Work at the Alligator Rivers uranium deposits has been in progress since 1981. A summary of past field and laboratory investigation has been included in Section 2. A number of additional experiments are being carried out in conjunction with them and these are summarised below.

A. Fission Products and Transuranic Elements

A major study has been carried out in the last 4 years during which analytical procedures for ^{129}I, ^{36}Cl, ^{99}Tc and ^{239}Pu in samples of rock and groundwater have been developed.

Iodine-129

Ore samples from the weathered top of the Koongarra ore body were found to have atomic ratios $^{129}I/U$ ranging from 5 to 22% of predicted equilibrium values. Down-gradient (and in oxic zones of dispersed uranium at the Ranger ore deposit) there appears to be enrichment of $^{129}I/I$ ratios considerably higher than estimated for atmospheric background. Samples collected up-gradient had the lowest ratios, with samples from boreholes intersecting the primary ore zones having the highest values. Down-gradient ratios were variably lower, but still well above background. However ^{36}Cl analyses showed no apparent trend with location in the deposit.

The results indicate strong partitioning of ^{129}I into groundwater during weathering of primary ore, which is consistent with the depleted $^{129}I/U$ ratios of ore samples from this zone. Down-gradient of the primary ore deposit the ^{129}I concentrates decrease due to dilution and to removal from solution by sorption. However $^{36}Cl/Cl$ measurements for the water suggest that all the ^{129}I decreases can be attributed to sorption.

Technetium-99

Measurements for one ore and one groundwater sample from Koongarra have been reported by David Curtis and June Fabryka-Martin. When the mobility of technetium in the system was compared to that of uranium, the molar ratio of the groundwater sample 2.8 x 15^{11} was a factor of 2-17 times greater than predicted for equilibrium conditions.

Plutonium-239

Measurements for two ore samples, one of 24% uranium described as quartz chlorite schist containing pitchblende and yellow secondary uranium mineral, the second containing 14% uranium in the form of primary pitchblende, have been reported.

Assuming that $^{239}Pu/^{238}U$ in the primary pitchblende represents nuclear equilibrium, the simplest interpretation suggests that ^{239}Pu and U are also in nuclear equilibrium in the schist sample. Thus plutonium and uranium hve been unfractionated in this sample for the last 10^5 y. Either the conditions that produced the mineralogical alterations did not fractionate the two actinides on the scale of the sample size, or alteration of the sample occurred more than 10^5 y ago.

It is planned that the abundances of ^{239}Pu, ^{129}I and ^{99}Tc in rock samples from Koongarra will be measured and inter-compared so that calculations of their production and models of the dispersion through the secondary mineralization can be evaluated. It is possible that uncertainties resulting from the lack of knowledge of inground neutron fluence in the production models will then be minimized so that the effects of isotope migration in the overall system can be derived.

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B. Colloids

The possible importance of colloids in groundwater transport of such radionuclides as Th and Pu was identified in the early stages of work in the Alligator Rivers Region. If colloidal transport should prove to be significant, one of the potential implications would be that linear K_d factors should no longer be used to describe solute/host rock interactions.

Sampling techniques aimed to concentrate the colloid fraction of groundwater samples have been used so that we were able to characterise the colloid and measure the enhanced fraction of uranium and thorium radionuclides.

The investigations have included particle size analysis by photon correlation spectroscopy and transmission electronmicroscopy and associated EDS and EELS analysis as well as radionuclide, elemental composition, cation, anion and total organic carbon analyses.

Particulate material with sizes in the range 1-5 μm was shown to be the most important carrier of thorium and actinium in 5 μm filtered groundwater; however, it is possible that the material is an artefact of the action of the submersible pump. If this is not the case, this fraction could be very significant for radionuclide transport in fractured material or in systems where channelling is important.

The following conclusions have been drawn, based on the work carried out.

1) Most elements which are present in measurable levels in unfiltered groundwater are contained within or adsorbed on particles above one micron in size, or are dissolved. No element was significantly enriched in the colloidal size range. The majority of uranium is dissolved, whereas almost all the thorium and actinium are associated with particulates above 1 μm in size. Thus for these radionuclides, only a small fraction of the total radionuclide content in the unfiltered groundwater is associated with the colloid phase.

2) About 0.1-1% of the uranium activity and 10-70% of the thorium activity in the sub-1 μm fraction are associated with the colloid phase. There is evidence to suggest that the colloidal uranium is in adsorption/desorption equilibrium with the groundwater.

3) The uranium content of the ultrafiltrate increases with the ore grade over several orders of magnitude, but the ^{230}Th concentration is extremely low and does not increase significantly despite very large variations in ^{230}Th content of the host rock.

4) Measurable levels of uranium, thorium and iron are retained by the Amicon filter and may be either colloidal or sorbed from solution. The presence of actinium in the fraction retained by the Amicon filter may indicate an association with colloidal particles which would be evidence for actinium mobility in groundwater. The level of ^{230}Th in the <1 μm-filtered groundwater and associated with colloidal particles is very low and indicates that it is the least mobile of the radionuclides studied.

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C. Fracture Transport

A study of sections of crystalline rock adjoining a water conducting fracture has revealed that activity ratios can fluctuate sharply within a few centimetres of the fraction surface.

The techniques of electronmicroscopy (in conjunction with an electronmicroprobe), PIXE-PIGME analysis using a 2-D microprobe with a 50 μm diameter proton beam and fission and alpha track mapping are being employed to study the migration of uranium and associated elements in new fracture regions. A series of samples has been selected from a drillcore at depths through the weathered and non-weathered regions and thin sections have been prepared.

D. Planned Investigations

I Hydrogeology

Objectives

- To develop an understanding of the water transport pathways in the region of the Koongarra deposit.

Proposed Work

a) Review and make preliminary interpretation of existing hydrology data in the Koongarra area. Recommend positions of any additional boreholes and data required.

b) Develop a model of groundwater flow in the Koongarra area in conjunction with sub-project I.

c) Drill additional boreholes, clean existing boreholes and establish monitoring points for future samples.

d) Conduct hydrological tests in monitoring boreholes, including drawdown tests, static water levels.

e) Make final interpretation of hydrogeological regime in the region of the Koongarra deposit.

II Uranium/thorium series disequilibria studies

Objectives

- To collect field data and associated laboratory data to enable radionuclide transport and geochemical modelling codes to be developed and evaluated.
- To identify and quantify processes which occur on both short and long geological timescales and which may be significant in the transport or retardation of radionuclides in the geosphere.

Proposed Work

a) To collect and analyse drill core samples from Koongarra for data base preparation from both weathered and unweathered material within the Koongarra uranium ore deposit and extending downstream into the sub-economic zone. The analyses will include uranium and thorium series nuclides, lead isotope ratios and elemental composition data.

b) To develop and utilise sequential extraction techniques and other chemical and physical procedures to measure the fractionation of uranium and thorium series nuclides in iron/clay/quartz phases, etc.

c) To study uranium series disequilibria and other associated indicator elements in the vicinity of water conducting fractures in drill core.

d) To evaluate the influence of phase boundaries on nuclide migration.

e) To undertake laboratory studies of the interaction of uranium and thorium series nuclides, fission products, and transuranic nuclides with synthetic samples and selected core samples from Koongarra.

III Colloids and groundwater

Objectives

- To investigate the physical and chemical composition and characteristics of natural colloids present in groundwater at the Koongarra site.
- To investigate the significance of colloids in the transport of radionuclides through the geosphere.
- To investigate the interaction of groundwater and the host rock in the Koongarra body and their influence on radionuclide transport.

Proposed Work

a) To collect samples of groundwater from Koongarra.

b) To utilise and develop techniques for concentrating the colloidal fraction in groundwater samples.

c) To develop techniques for collection and fractionation of particulate samples in the 5 μm to 0.1 μm range.

d) To measure, using a wide range of techniques, the chemical and physical properties of groundwater colloids and particulates, and compare these data with rock sample data.

e) To provide groundwater analyses for geochemical modelling.

f) To interpret the results in terms of groundwater/rock interactions and of radionuclide transport.

g) To identify the significance of colloid transport of radionuclides in the geosphere.

h) To prepare a program for carrying out colloid tracer experiments in the field using closely spaced drill holes.

i) To carry out colloid tracer experiment and interpret results.

IV Fission products studies

Objectives

- To study the production mechanisms and dispersion via groundwater transport of fission products such as technetium-99 and iodine-129 in the Koongarra ore deposit.

Proposed Work

a) Measure iodine-129 and technetium-99 in rock and water samples collected from the unweathered and weathered zones of the uranium ore deposit at Koongarra.

b) Measure iodine-129 in mineral phases extracted from drill cores and groundwater samples at various depths and distances from the uranium deposit at Koongarra.

c) In conjunction with sub-project IV, sample and measure the chemical constituents of groundwater and rock which are significant in the mobilisation or retardation of the fission products.

d) Study the production mechanisms for iodine-129 and technetium-99.

e) Determine the iodine-129 spontaneous fission product yield for uranium-238.

f) Measure the neutron flux in the lithosphere and establish its importance in the production of fission products (and transuranic nuclides).

g) Verify the initial iodine-129/iodine-127 ratios in surface source materials.

h) Compare measured data with predicted data and incorporate into models of production/dispersion behaviour.

V Transuranic nuclide studies

Objectives

- To study the production mechanisms and dispersion via groundwater transport of transuranic nuclides in the Koongarra ore deposit.
- To develop techniques for measurement of uranium-236/uranium-235 ratios and establish procedures for measurement of integrated neutron flux.

Proposed Work

a) Measure plutonium-239 in samples from the primary and secondary mineralization at Koongarra and extend these measurements if possible to water and concentrated colloid samples from selected drill holes.

b) Select appropriate locations for additional samples on the basis of plutonium-239 results so that sufficient data on plutonium-239 distribution in the direction of the water flow path will be available to describe the production and migration of this nuclide.

c) Develop methods for measurement of uranium-236/uranium-235 and carry out a sampling program to derive integrated neutron flux. This proposal will interact with the neutron flux monitoring program included in sub-project V.

d) Assess the feasibility for measurement of natural levels of neptunium-237 with isotope dilution mass spectrometry.

e) Extend the measurement program to neptunium-237 if this proves feasible.

f) Compare measured data with predicted data and incorporate into models of production/dispersion behaviour.

E. Experimental Schedule

The schedule of current and future work being carried out as a part of the ARAP is given below.

I Hydrogeology

Time Schedule	1 Sept 1987	1 Sept 1988	1 Sept 1989	1 Sept 1990
a) Preliminary study				
b) Model of flow				
c) Additional Boreholes				
d) Hydrological Tests				
e) Hydrogeological Interpretation				

II Uranium/thorium series disequilibria studies

Time Schedule	1 Sept 1987	1 Sept 1988	1 Sept 1989	1 Sept 1990
a) Drill Core Analysis				
b) Uranium/Thorium Fractionation				
c) Fractures Studies				
d) Phase Boundaries				
e) Nuclide – Core Sample Interactions				

III Colloids and Groundwater

Time Schedule	1 Sept 1987	1 Sept 1988	1 Sept 1989	1 Sept 1990
a) Groundwater Sampling				
b) Colloid Concentration/ Fractionation				
c) Particulates				
d) Chemical Physical Properties				
e) Groundwater Analysis				
f) Interpretation				
g) Significance				
h) Tracer Expt. Feasibility Study				
i) Tracer Experiment				

IV Fission Product Studies

Time Schedule	1 Sept 1987	1 Sept 1988	1 Sept 1989	1 Sept 1990
a) Rock and Water				
b) Mineral Phases				
c) Groundwater				
d) Production				
e) Fission Product Yield				
f) Neutron Flux				
g) $^{129}I/^{127}I$ Ratio				
h) Comparison and Modelling				

V Transuranic Nuclide Studies

Time Schedule	1 Sept 1987	1 Sept 1988	1 Sept 1989	1 Sept 1990
a) ^{239}Pu Measurement				
b) Additional Sampling				
c) $^{236}U/^{235}U$				
d) Assess ^{237}Np Measurement				
e) ^{237}Np Measurement				
f) Comparison and Modelling				

4. PERFORMED AND PLANNED MODELLING ITEMS

A number of one-dimensional models of the formation of the secondary deposit have been attempted.

Firstly, the large radiochemical data base available was used in an open system interpretation of the secondary mineralisation process. The Lucas Heights open system approach was modified to conceptually describe the roll front like advancement of the secondary zone of the Koongarra deposit.

The main features of this work are included below.

Secondly, the modelling formalism of hydrodynamic transport was applied to quantitatively follow the ideas developed by Osmond and Cowart. In this work the rock was treated as a single phase in equilibrium with groundwater, hydrodynamic dispersion and transport by colloids were omitted. A small amount of data on the concentration of various lead isotopes in the secondary zone were used for additional estimation of the timescale. A further model (Davidson and Dickson) has considered a porous flow model for steady state transport of Ra in groundwater.

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A. Open System Modelling Approach

1 General Outline

An approach based on natural radioactive decay characteristics of radionuclides of the uranium series, has by definition, a reduced applicability and quantitative evaluation ability. However, it is usually easier to obtain a suitable experimental data base which is limited to a comprehensive set of relevant $^{234}U/^{238}U$, $^{230}Th/^{234}U$ activity ratios (ARs) for individual rock samples from the studied locality. Additional information on radionuclide distribution and on ARs in local groundwater is usually also available.

Mobilization of uranium in a system has been associated with weathering of rock strata where groundwater permeates the region and dissolves the accessible uranium. Within the model described here, it is assumed that at time = 0, following a known or undefined sequence of events the movement of water through the given geological system commenced. Time t = 0 is not necessarily a point value, but may represent a very extended initial period; however, on the relevant timescales it is of negligible duration. The moving water carries along uranium and some of its daughter products while others like thorium-230, are not transported in soluble form. The leaching and eventual desorption of the radionuclides are considered to be first order processes (Airey et al. 1986) and their rates are taken to be proportional to the amount of uranium present in the accessible form.

2 Transport Equations

The generating equations for uranium-238, uranium-234 and thorium-230 are formulated as

$$-\frac{dU_8}{dt} = \lambda_8 U_8 + \xi U_8 \qquad (1)$$

$$-\frac{dU_4}{dt} = \lambda_4 U + R\xi U_4 - \lambda_8 U_8 \qquad (2)$$

$$-\frac{dTh}{dt} = \lambda_0 Th - \lambda_4 U_4 \qquad (3)$$

where U_8, U_4, Th are uranium 238, uranium-234 and thorium-230, respectively, and λ_8, λ_4, λ_0 are the corresponding decay constants, ξ is the leaching/deposition rate and R is the ratio of the ^{234}U to ^{238}U leaching/deposition rate (usually the AR in groundwater adjacent to the deposit).

The assumption that the uranium transport has first order process characteristics is satisfactory for leaching, however it restricts the physical meaning of deposition processes to the situation where there is good correlation between uranium concentration in the given region and its level in the groundwater. It can also be postulated that in the weathered region the relevant property is the number of accessible sites on the amorphous component of the iron or aluminium (Airey et al. 1986).

This problem can be overcome by modifying the model to include additional new variables for the radionuclide concentrations in groundwater. However, this change requires a more comprehensive detailing of the initial conditions.

In this second approach, the leaching/deposition rate is related to the actual uranium concentration in rock and water, and to the system adsorption ability represented by the adsorption coefficient, K_d. This adsorption coefficient is defined as the ratio of the weight of uranium adsorbed per unit weight of adsorbate to the weight of dissolved uranium per unit volume of solution after adsorption equilibrium has been attained.

The generating equations (1), (2) and (3) were modified to include this representation of the deposition/leaching process:

<u>Uranium-238</u>

$$-\frac{dU_8}{dt} = \lambda_8 U_8 + A_8 \quad , \tag{4}$$

$$= (\lambda_8 + q\Pi\phi)U_8 - \phi U_8^w \quad . \tag{5}$$

Uranium-234

$$-\frac{dU_8}{dt} = \lambda_4 U_4 - \lambda_8 U_8 + A_4 \quad , \tag{6}$$

$$= (\lambda_4 + Rq\bar{\Pi}\phi)U_4 - \lambda_8 U_8 - R\phi U_4^{w} \quad . \tag{7}$$

Thorium-230

$$-\frac{dTh}{dt} = \lambda_0 Th - \lambda_4 U_4 \quad . \tag{8}$$

3 Initial and Boundary Conditions

In many cases, ARs in rock or groundwater can be set up by accepting the general assumption of the model, e.g. that at time t = 0, the primary deposit exists in an unperturbed state and probably is equilibrated, thus $^{234}U/^{238}U$ and $^{230}Th/^{234}U$ are set to unity. In the region of the secondary mineralization, the value of $^{234}U/^{238}U(0)$ is set equal to the ratio in which the water deposited the uranium because the uranium carried by groundwater and deposited in the given region overwhelmed, in mass terms, any uranium initially present in this region during the 'time zero period'. The starting 'point' on such a geological timescale can be of thousands of years duration. Thus when waters flowing through the rich deposit enter the surrounding area where the uranium concentration is many orders of magnitude lower, these waters could during the 'initial time' deposit a substantial amount of uranium and overshadow the original activity ratio. Furthermore, bearing in mind the assumption that thorium does not travel, the initial value of $^{230}Th/^{234}U(0)$ is set to zero.

In the particular case of the secondary mineralization process which has resulted in the formation of the dispersion fan in the Koongarra deposit, the relationship between the timescale of the event and the distance from the original deposit can be used as a common characteristic which constrains and 'interlinks' the model solutions for the individual rock samples. For example, the samples can be chosen along a specific horizontal direction through the secondary deposit although knowledge

of the general direction of the groundwater flow would be desirable in a sample selection such as this. The introduction of a one-dimensional formulation for radionuclide transport which assumes that uranium is transported only along the water flow lines introduces obvious limitations. The increase in uranium is expected only from the regions along such a water flow line and before the receptor region, and any loss from the region is expected to occur only to regions along the direction of flow.

4 Results

The samples were chosen from the centre layer of the secondary mineralization fan, starting from a position close to the primary ore body and extending away from it in a southerly direction. The level chosen is the area 15-20 m below the surface which contains the more uranium rich part of the fan. The area is also characterized by a dominant horizontal water flow pattern.

Figure 14 graphically represents the results of the modelling and shows the relationship between averaged rate of leaching/deposition and the position of the sample in the secondary mineralization region. The general qualitative trend shown by the curve exhibits the previously expected behaviour. The region closer to the primary ore body shows overall domination by the leaching process during the whole formation period, while further away from the original ore body the process shows overall deposition characteristics. This is in agreement with the hypothetical working of the moving front.

The timescale of the process is not unequivocally determined. It can be said however, that the age of the process is at least 700 ky and extends into the region of several My, but self-evidently the sensitivity of any method based on uranium-234 half-life diminishes rapidly at about 1 My.

One additional and significant result of modelling was the determination of the value of R, the $^{234}U/^{238}U$ ratio of the leachate. Although a large variability of the R value was allowed for during computation, the constraints of the modelling approach restricted the derived value of R to the narrow range of 0.80-0.85; a value which is in pleasing agreement with the data for modern groundwater from bore holes in the system.

5 Vertical Dispersion

The advantage of imposing an age-distance relationship is clearly demonstrated when we consider the results obtained from a series of samples which represented the vertical distribution of the uranium in the secondary mineralization fan in a section close to the original ore body. The sample depths ranged from 0-50 m of the DDH 52 drill core.

The solutions, in the form of sometimes large leaching/deposition rate-time domains were the result of incomplete site description and a lack of sufficient constraints.

The leaching process could be seen to be strongest near the surface, due probably to percolating surface water during the wet season, with the rate then decreasing markedly for the samples near the bottom of the weathered zone.

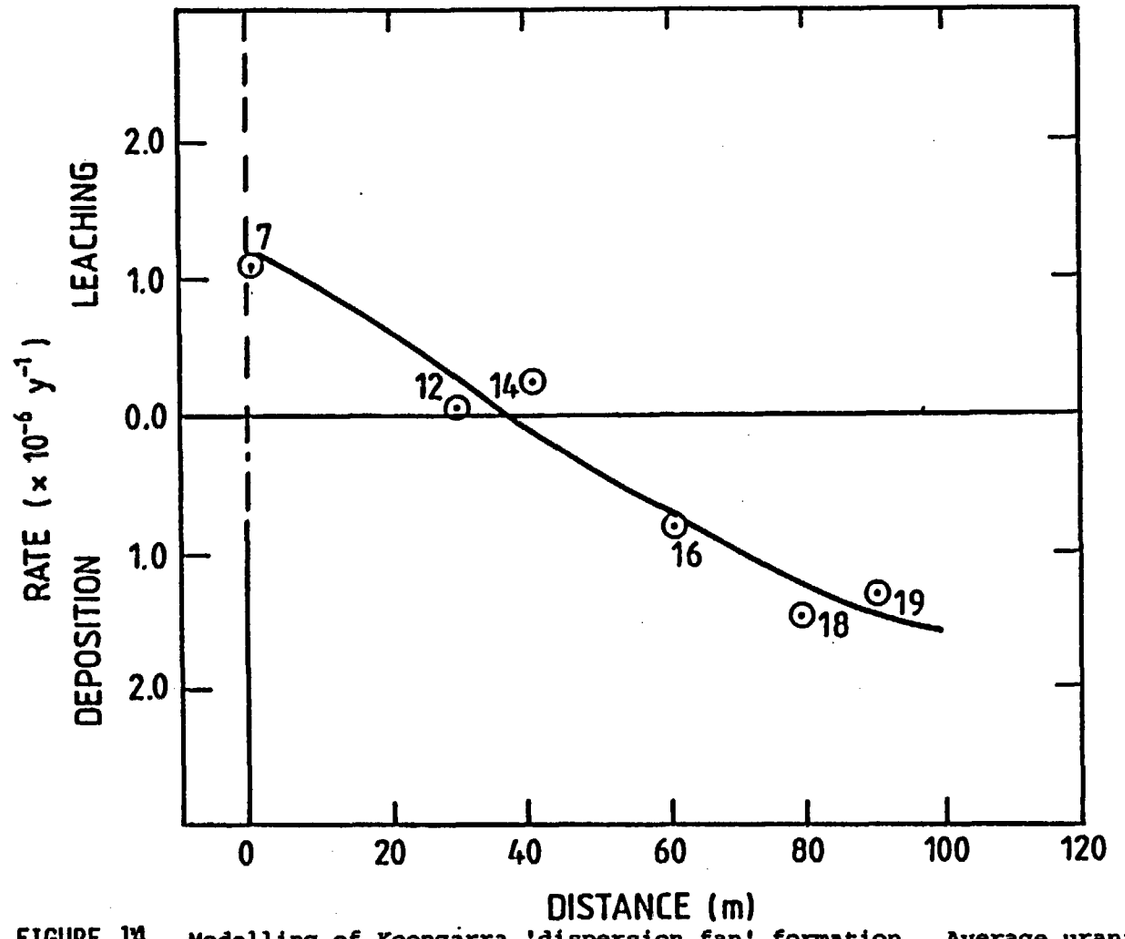

FIGURE 14 Modelling of Koongarra 'dispersion fan' formation. Average uranium transport rates vs distance of the original distance.

B. Transport Modelling

1 A Simple Migration Model

The simplest quantitative one-dimensional model includes the effects of transport by flowing groundwater and linear, equilibrium sorption, described by a constant retardation factor. Different isotopes of the same element can have different distributions between the various rock phases, arising from the effects of α-recoil. This is modelled by assigning to them different retardation factors, although we do not expect the differences to be large. Neither variations vertically nor variations along the migration path are as yet considered. In the chains we examine, it will not be necessary to consider two elements with the same mass number, so we write the concentration in the groundwater of the ith member of the chain with mass number m_i as c_{m_i}, which satisfies the equation

$$R_{m_i} \frac{\partial c_{m_i}}{\partial t} + u \frac{\partial c_{m_i}}{\partial x} + R_{m_i} \lambda_{m_i} c_{m_i} = R_{m_{i-1}} \lambda_{m_{i-1}} c_{m_{i-1}}, \qquad (1)$$

where λ_{m_i} and R_{m_i} are the appropriate decay constant and retardation factor.

A model for the mobilization of the uranium as the rock is weathered is also required. We assume that this takes place comparatively rapidly, leading to a distribution of mobile ^{238}U across the ore body. So we solve an initial value problem with this distribution. Thereafter we assume that linear equilibrium sorption is appropriate. Obviously this treatment of the chemistry is very simplified. The kinetics of leaching and mobilization could be important, as could the kinetics in the depositional region as the distribution in the diferent rock phases develops as a result of α-recoil. In addition, the effects of non-linear sorption and solubility limitation could be significant.

2 General Solutions

With the neglect of dispersion, the initial distribution of ^{238}U, $c^{o}_{238}(x)$, migrates with the retarded groundwater velocity:

$$c_{238}(x,t) = c^{o}_{238}\,(x - ut/R_{238})e^{-\lambda_{238}t}\,. \qquad (2)$$

This leads to the most obvious estimate of the migration time

$$t_u = R_{238}\,L/u = \phi\,R_{238}\,L/q, \qquad (3)$$

Where L is the migration distance, q is the Darcy velocity and ϕ is the porosity.

The general solution for daughter nuclides is

$$c_{m_i}(x,t) = e^{-\lambda_{m_i} t} c^{o}_{m_i}\left(x - \frac{ut}{R_{m_i}}\right) + e^{-\lambda_{m_i} t} \frac{R_{m_{i-1}} \lambda_{m_{i-1}}}{R_{m_i}} ,$$

$$\int_0^t e^{\lambda_{m_i} \bar{t}} c_{m_{i-1}}\left(x - \frac{u(t-\bar{t})}{R_{m_i}}, \bar{t}\right) d\bar{t} . \qquad (4)$$

The solution for the stable daughter m_n is obtained by setting $\lambda_{m_n} = 0$.

From these solutions, the activity a_{m_i} on the rock can be deduced using

$$a_{m_i} = \phi(R_{m_i} - 1)\lambda_{m_i} c_{m_i} . \qquad (5)$$

For most nuclides, $R_{m_i} >> 1$ and so $R_{m_i}-1$ will be replaced by R_{m_i}.

Finally, to obtain solutions that can be used to analyse observed rock ARs, we assumed that $c^{o}_{m_1}$ varies only slowly with distance. Then if we replace $c_{m_{i-1}}$ within the integral in Equation (4) by Taylor series expansions about $x-ut/R_{m_1}$, the integral can be evaluated as power series. The members of the chain are approximately in secular equilibrium, and we investigate the first perturbation to this equilibrium. The AR of successive decaying nuclides is given by

$$\frac{a_{m_i}}{a_{m_{i-1}}} = 1 - \frac{u\left(\frac{R_{m_1}}{R_{m_i}} - 1\right)}{R_{m_1} \lambda_{m_i}} \frac{1}{a_{m_1}} \frac{da_{m_1}}{dx} . \qquad (6)$$

Where

$$u\left(1 - \frac{R_{m_i}}{R_{238}}\right) g'_{m_i} = R_{m_{i-1}} \lambda_{m_{i-1}} g_{m_{i-1}} - R_{m_i} \lambda_{m_i} g_{m_i} . \qquad (7)$$

3 Analysis of Rock ARs

There is a considerable body of data from Koongarra on the ARs on rock of ^{234}U to ^{238}U and ^{230}Th to ^{238}U. A qualitative method of analysis was proposed by Osmond and Cowart , and now we have made it more quantitative.

There are six possible cases, depending on the relative magnitudes of R_{238} R_{234} and R_{230}; however these are really three pairs, with the paired cases having relative rates of migration reversed. The approximate solutions based on Equation (6) are shown as straight lines passing through (1,1).

$$G = \left(1 + \frac{(R_{238}/R_{230}-1)\ \lambda_{234}}{(R_{238}/R_{234}-1)\ \lambda_{230}}\right)^{-1} \tag{8}$$

By examining the value of G, we can see the three different cases. If ^{238}U is more mobile than both ^{234}U and ^{230}Th ($R_{238} < R_{234}$, $R_{238} < R_{230}$) then $0<G<1$ (Figure 8). In the other two cases ^{234}U moves more rapidly than ^{238}U ($R_{234} < R_{238}$). G is ∞ when

$$R_{234} = R_{234}^{\infty} = \left(1 - \left(\frac{R_{238}}{R_{230}} - 1\right) \frac{\lambda_{234}}{\lambda_{230}}\right)^{-1} R_{238} \ . \tag{9}$$

At Koongarra, it is found experimentally that, except in the region where ^{238}U, ^{234}U and ^{230}Th are close to equilibrium, the ARs on drill core sample of ^{234}U to ^{238}U and ^{230}Th to ^{234}U are either both greater than one (on the upstream side of the deposit) or both less than one (on the downstream side). This is consistent with the picture shown in Figure 15 with $R_{238} < R_{234}$, R_{230}. However, it is not the same as the results of Osmond and Cowart, who found that for their case ^{234}U moved more rapidly than ^{238}U. The points obtained from samples from various drill holes are shown in the figure, along with the best linear fit to them. It is thought that ^{230}Th is much less mobile than ^{238}U and ^{234}U, and with the assumption that $R_{230} = \infty$, the retardation factor of ^{234}U is estimated to be 1.2 times that for ^{238}U. The region that is inaccessible if ^{230}Th is immobile is also shown on the figure.

Equation (6) can also be used to estimate the migration time if the gradient of ^{238}U activity is estimated from drill core samples. The estimate is

$$t_u = \frac{R_{238}L}{u} = \frac{\left(\frac{R_{238}}{R_{m_i}} - 1\right)}{\left(1 - \frac{a_{m_i}}{a_{m_{i-1}}}\right)\lambda_{m_i}} \frac{L}{a_{238}} \frac{da_{238}}{dx} . \tag{10}$$

4 Estimates of the Migration Time

In the previous Sub-sections we have developed two methods of estimating the time that the uranium has been migrating. First, one can be found from Equation (3) based on laboratory and field estimates of distribution coefficients and numerical estimates of the groundwater velocities. Alternatively, Equation (10) gives an estimate based on the spatial gradient of ^{238}U activity, and the disequilibrium between ^{234}U and ^{238}U or ^{230}Th and ^{234}U. These are examined in turn.

Migration time based on laboratory sorption value is in the range

$$t_u = 5 \times 10^3 - 3 \times 10^6 \text{ yrs} , \tag{11}$$

whereas the range based on the field estimate is in the range

$$t_u = 1.2 \times 10^5 - 10^6 \text{ yrs} . \tag{12}$$

To estimate the migration time using the disequilibrium data, we need to find $\zeta = L\, a_{238}^{-1}\, da_{238}/dx$.
One from DDH 3 has $\zeta = 0.5$, $^{230}Th/^{234}U$ AR = 0.91 and $^{234}U/^{238}U$ AR = 0.96. If we assume again that ^{230}Th is immobile and that $R_{234}/R_{238} = 1.2$, then these respectively give

$$t_u = 2.4 \times 10^6 \text{ and } 3 \times 10^6 \text{ y} . \tag{13}$$

If thelead and the intermediate daughters are effectively immobile, and the uranium is mobile and migrating as a step function, then the time since the uranium arrived can be calculated from

$$\left(\frac{c_{m_n}}{c_{204}}\right)_{obs} = \left(\frac{c_{m_n}}{c_{204}}\right)_{init} + \left(\frac{c_{m_1}}{c_{204}}\right)_{obs} \left(e^{\lambda_{m_1} t_{m_1}} - 1\right), \tag{14}$$

where c_{204} is the concentration of ^{204}Pb, which does not arise from radioactive decay and so is primordial, and t_{m_1} is the time associated with the decay of parent m_1.

The results for the three samples from the region of primary ore before weathering and the two from outside the ore body yielded the age estimate in 10^8-10^9 range and 2-5.10^7 y respectively.

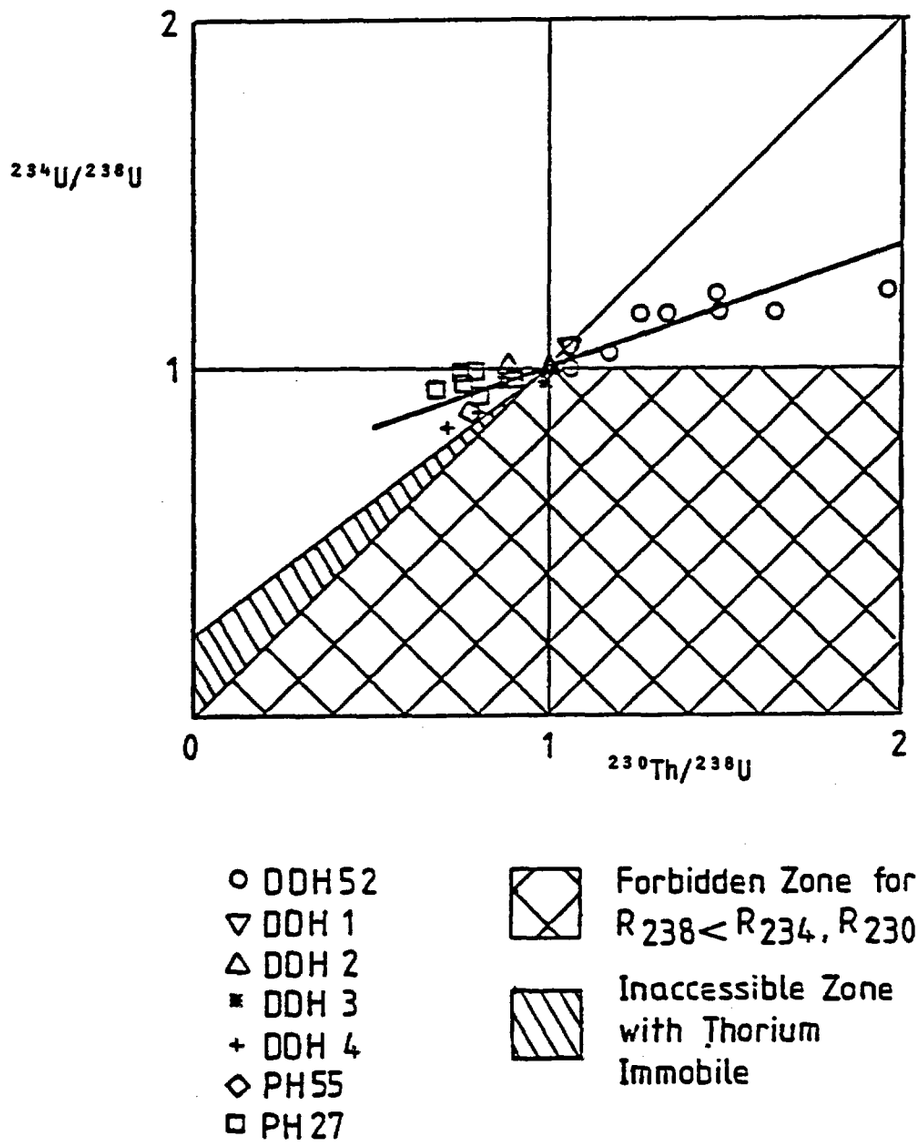

FIGURE 15 Plot of $^{234}U/^{238}U$ and $^{230}Th/^{238}U$ activity ratios found at Koongarra showing best linear fit ($R_{234} = 1.2\ R_{238}$, $R_{230} = \infty$).

C. Planned Modelling within the ARAP

A number of modelling objectives have been drawn up for the ARAP and outlines of the proposed work and a time schedule for its completion were included in the International Agreement. Four tasks were identified at the ARAP Modelling Workshop, 8-12 February 1988, and these are also detailed below.

I ARAP - Proposed work and time schedule

Objectives

- To contribute to the production of reliable and realistic models of radionuclide migration within geological environments relevant to the assessment of safety of radioactive waste repositories.
- To develop methods of validation of models using a combination of laboratory and field data associated particularly with the Koongarra uranium deposit.
- To advise and recommend on the scope of the laboratory and field experiments needed to provide the data.

Proposed Work

a) *Data Base*

Establish a computerised data base containing all available data from Koongarra.

b) *Site Characterisation and Validation of Models*

Develop a regional hydrology model of the Koongarra deposit in conjunction with sub-project II and carry out preliminary predictions of flow and migration pathways in and around the Koongarra deposit.

Apply geochemical modelling techniques for radionuclide source term calculations.

Develop radionuclide transport models and other conceptual models such as the open system model based on the evaluation of the secondary ore zone.

Study the coupling of geochemical and hydrodynamic codes for predicting multi-component chemical-radionuclide transport.

c) *INTRAVAL Case Study*

Develop an outline Koongarra case study specification for consideration by the INTRAVAL group.

Prepare a detailed case study based on comments received from the INTRAVAL group.

d) *Modelling Support Studies*

The modelling program will be closely integrated with the experimental program. Specific support will be provided as described below.

Conduct uranium-thorium disequilibria modelling for macro- and micro-scale studies in support of sub-project III.

Conduct fission product and transuranic element production and migration modelling in support of sub-projects V and VI.

Compare detailed predictions of radionuclide source terms and distribution with field and laboratory observations.

Make recommendations for future field and laboratory studies to support modelling.

Time Schedule	1 Sept 1987	1 Sept 1988	1 Sept 1989	1 Sept 1990
a) Data Base				
b) Site Characterization & Validation				
c) INTRAVAL Case Study				
d) Modelling Support				

II Tasks

A. GEOCHEMICAL MODELLING

AIM

To develop an understanding of uranium mobility in the Koongarra ore deposit as an analogue for the migration of radionuclides from a nuclear waste repository.

OBJECTIVES

I Identification of the controlling reactions for the formation of the uranium phosphate zone.

II Identification of the controlling reactions for the formation of the uranium silicates from pitchblende in the zone just beneath the weathered zone.

PROPOSED WORK

(1) Assemble available mineralogical data and water chemistry data.

(2) Assess the available thermodynamic data for the species of interest (mineral, aqueous, gaseous).

(3) Carry out preliminary calculations of relative mineral stabilities in equilibrium with aqueous fluids and/or use previous available studies.

(4) Carry out a preliminary assessment of the aqueous speciation, solubility and chemical mass transfer during the formation of the uranium phosphate zone and the uranium silicate zone.

(5) Integrate new information on mineralogy and water chemistry into the above tasks as such information becomes available.

B. MIGRATION OF RADIONUCLIDES IN THE WEATHERED ZONE

AIM

To investigate the mobilisation and migration of naturally-occurring radionuclides in the weathered zone of the No 1 ore body at Koongarra.

OBJECTIVES

1) Examine the migration of mobile long-lived uranium isotopes and shorter-lived descendents away from the original region of uranium mineralization in the weathered zone.

2) Examine the mobility of long-lived uranium isotopes in the region of the weathered zone where there was originally primary ore.

PROPOSED WORK

1) Recently, additional data on disequilibria in the uranium series has been acquired. These will be incorporated into the framework of existing models.

2) There are approximations in the existing models associated with (i) the weathering of the ore body, (ii) the time-scale of mobilization, (iii) the groundwater flow, (iv) sorption of radionuclides onto solid material, (v) multi-phase nature of the solid material, (vi) spatial variability, and (vii) temporal variability. The validity of these will be examined.

3) In the light of this examination, additional processes and features will be incorporated into the existing models.

4) The models will be extended to lower members of the decay chains (Ra, Pb isotopes).

5) The position of additional boreholes and additional measurements of activity on rock samples and adjacent water samples will be suggested so that existing data within the ore body and in the region around the ore body will be supplemented.

6) We will interact with other modelling and experimental groups and review models in the light of additional work on groundwater flow, drill core analysis, uranium series disequilibrium analysis, geochemical modelling, colloid transport and geomorphology.

C. OPEN SYSTEM MODELLING

MIGRATION OF RADIONUCLIDES IN THE WEATHERED ZONE

OBJECTIVE

To utilise radiochemical data for selected crystals of saleeite in evaluation of the timescale of advancement of secondary dispersion.

PROPOSED WORK

1) To assess data presently available for saleeite crystals and possibly request new samples.

2) To understand the crystal growth process in the weathered zone environment and try to obtain a time dependent function of this growth including examination of activity ratios of uranium series nuclides.

3) To deduce the local timescale and extrapolate it to the complete weathered zone.

4) To use the obtained time factor in an open system model of the advancement of the dispersion fan (one-dimensional).

5) Extend the model to a two-dimensional system (include vertical sections).

D. WEATHERING HISTORY OF KOONGARRA

OBJECTIVE

To determine the sequence and timing of weathering ovee the Koongarra uranium deposit.

PROPOSED WORK

1) Review previous geomorphological work in the NT and Qld during Tertiary (<10 My) and recent studies in Pine Creek Geosyncline.

2) Consult current researchers in Pine Creek Geosyncline on possible dating of escarpment retreat by mass balance calculations.

3) Investigate age dating of relevant weathering K/Ar (e.g. on jarosites/alunites) or ^{10}Be on accretionary surfaces.

5. VALIDATION ASPECTS

A. Weathered Zone, Formation of the Dispersion Fan

Preliminary estimations of the rate of advancement of the secondary mineralization have been made. Further work should be carried out in association with existing and proposed hydrogeological work which investigates groundwater flow patterns and flow velocities. The effects of transient and spatial changes in flow rates, variation in water chemistry (recharge) and its effect on sorption coefficients will need to be considered. Retardation mechanisms in the secondary mineralization zones should be closely examined.

Comprehensive rock and radiochemical data are already available for specific sections through the ore deposit and extending into the sub-economic zone down-gradient, and are a continuing part of the experimental program being carried out in the Alligator Rivers Analogue Project.

B. Primary Deposit and Transition Zone

A geochemical evaluation of rock/water interactions, of the rate of leaching from the primary uranium ore and the rate of formation of the secondary minerals could be made. Geochemical modelling techniques for radionuclide source term calculations could be developed.

A substantial data base of rock and water chemistry from this region is available, however, further work is in progress.

C. Uranium Fractionation in Mineral Phases

A data base is available for uranium fractionation between amorphous and crystalline iron and clay/quartz phases. Modelling uranium transfer along and between the phases is essential to validate the observation that only the uranium in the amorphous phase is directly accessible to groundwater.

Modelling could include estimation of leaching/deposition rates in the amorphous phase and the exchange rate between the amorphous and crystalline phases. The role of α-recoil in the accumulation of daughter elements in the clay/quartz phase should be considered.

D. Crystalline Rock Zone

The uranium deposit at Koongarra is situatd in both the weathered zone and the unweathered crystalline rock. Heterogeneity in the groundwater flow may have had an important role in the redistribution of the uranium deposit.

The investigation should include the retardation properties of matrix diffusion mechanisms and the development of single and multiple fracture models.

Geochemical modelling combined with isotopic analyses is could also be utilized in the interpretation of the matrix diffusion effects.

Measurements of radionuclide distributions and associated elements in the vicinity of fractures is already underway with a number of fractures from both the crystalline and weathered rock being studied.

E. The Role of Colloid Transport

To design models which will include the effects of colloid and possibly particulate transport in free flowing fracture networks.

A series of measurements aimed to evaluate the significance of colloid transport has already been carried out at Koongarra and is continuing in the Alligator Rivers Analogue Project.

F. Fission Products and Transuranics

A substantial measurement program on ^{129}I, ^{99}Tc and ^{239}Pu is already underway. Preliminary groundwater and dispersion models have been set up to evaluate this data.

The modelling of the weathered zone and the formation of the dispersion fan, will be greatly enhanced by including the distribution of the fission products and transuranics to those of the uranium and thorium series radionuclides.

6. AVAILABLE DATA

A. Hydrogeology

1 Climatological data, including rainfall and temperature.

2 Surface water measurements, including stream flow.

3 Location, elevation, geologic logs, casing and perforation details of all test holes and wells.

4 Map, showing test holes and wells, as well as land-surface contours.

5 Aquifer test results including water-level drawdowns, discharge measurements, and water quality of discharge.

6 Periodic water-level measurements which show seasonal fluctuations and regional gradients.

7 Results of geophysical surveys and back-hoe pits which show thickness of upper deposits.

8 Results of early packer tests in upper part of the bedrock. Resistivity traverses.

B. Hydrochemistry

1 pH, Eh, D.O., conductivity, temperature.

2 Cations - F, Mg, Na, Al, Si, S, K, Ca, Ti, Mn, Fe.

3 Anions - HCO_3^-, SO_4, Cl, PO_4.

4 Trace Metals - Cd, Cr, Cu, Pb, Mn, Mo, Ni, Zn.

5 Isotopic - ^{14}C, $\delta^{13}C$, Ra, Rn, ^{3}H, $^{36}Cl/Cl$, Th, U.

C. Geology and Mineralogy

1 Full geological assay and drill core records.

2 Mineralogical Assay - 140 Percussion holes
107 Drill cores
300 Back-hoe pits and auger holes

3 Geological cross-sections.

4 Uranium assay.

5 X-ray diffraction studies (random powders and size-separated sediment studies).

6 Elemental data for homogenised rock samples.

D. Radiochemical

1 Radionuclide concentrations in rock and groundwater (^{238}U, ^{234}U, ^{232}Th, ^{235}U).

2 Activity ratios of rock and groundwater ($^{234}U/^{238}U$, $^{230}Th/^{234}U$, etc.).

3 Lead isotope ratios - rock samples.

4 Uranium fractionation between mineral phases.

5 α-recoil experiments.

6 Radionuclide levels on colloids.

7 Uranium adsorption, Lab - (K_d) v pH, Field - U and Th K_ds.

8 Uranium and Th concentrations and activity ratios adjacent to water conducting fracture.

9 Iodine-129 in rock and groundwater.

10 Limited ^{99}Tc in rock and groundwater.

11 Limited ^{239}Pu in rock.

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