A R A P

THE INTERNATIONAL ALLIGATOR RIVERS ANALOGUE PROJECT

Background and Results

An OECD/NEA International Project Managed by:
Australian Nuclear Science and Technology Organisation (ANSTO)
Environmental Science Program

NUCLEAR ENERGY AGENCY
ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

NEA ANSTO

ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

Pursuant to Article 1 of the Convention signed in Paris on 14th December 1960, and which came into force on 30th September 1961, the Organisation for Economic Co-operation and Development (OECD) shall promote policies designed:

- to achieve the highest sustainable economic growth and employment and a rising standard of living in Member countries, while maintaining financial stability, and thus to contribute to the development of the world economy;
- to contribute to sound economic expansion in Member as well as non-member countries in the process of economic development; and
- to contribute to the expansion of world trade on a multilateral, non-discriminatory basis in accordance with international obligations.

The original Member countries of the OECD are Austria, Belgium,Canada, Denmark, France, Germany, Greece, Iceland, Ireland, Italy, Luxembourg, the Netherlands, Norway, Portugal, Spain, Sweden, Switzerland, Turkey, the United Kingdom and the United States. The following countries became Members subsequently through accession at the dates indicated hereafter: Japan (28th April 1964), Finland (28th January 1969), Australia (7th June 1971), New Zealand (29th May 1973) and Mexico (18th May 1994). The Commission of the European Communities takes part in the work of the OECD (Article 13 of the OECD Convention).

NUCLEAR ENERGY AGENCY

The OECD Nuclear Energy Agency (NEA) was established on 1st February 1958 under the name of the OEEC European Nuclear Energy Agency. It received its present designation on 20th April 1972, when Japan became its first non-European full Member. NEA membership today consists of all European Member countries of OECD as well as Australia, Canada, Japan, Republic of Korea, Mexico and the United States. The Commission of the European Communities takes part in the work of the Agency.

The primary objective of NEA is to promote co-operation among the governments of its participating countries in furthering the development of nuclear power as a safe, environmentally acceptable and economic energy source.

This is achieved by:

- *encouraging harmonization of national regulatory policies and practices, with particular reference to the safety of nuclear installations, protection of man against ionising radiation and preservation of the environment, radioactive waste management, and nuclear third party liability and insurance;*
- *assessing the contribution of nuclear power to the overall energy supply by keeping under review the technical and economic aspects of nuclear power growth and forecasting demand and supply for the different phases of the nuclear fuel cycle;*
- *developing exchanges of scientific and technical information particularly through participation in common services;*
- *setting up international research and development programmes and joint undertakings.*

In these and related tasks, NEA works in close collaboration with the International Atomic Energy Agency in Vienna, with which it has concluded a Co-operation Agreement, as well as with other international organisations in the nuclear field.

PREFACE

The Koongarra uranium ore deposit is located in the Alligator Rivers Region of the Northern Territory of Australia. Many of the processes that have controlled the development of this natural system are relevant to the performance assessment of radioactive waste repositories. An Agreement was reached in 1987 by a number of agencies concerned with radioactive waste disposal, to set up the International Alligator Rivers Analogue Project (ARAP) to study relevant aspects of the hydrological and geochemical evolution of the site. The Project ran for five years. The Project Manager was Mr P Duerden (ANSTO) and the Steering Committee Chairs, Dr C J Hardy (1986-1991) and Dr W M Zuk (1991-1992).

The work was undertaken by ARAP through an Agreement sponsored by the OECD Nuclear Energy Agency (NEA). The Agreement was signed by the following organisations: the Australian Nuclear Science and Technology Organisation (ANSTO); the Japan Atomic Energy Research Institute (JAERI); the Power Reactor and Nuclear Fuel Development Corporation of Japan (PNC); the Swedish Nuclear Power Inspectorate (SKI); the UK Department of the Environment (UKDoE); and the US Nuclear Regulatory Commission (USNRC). ANSTO was the managing participant.

The Final Report will consist of a series of 16 volumes describing the work of the Project; these are listed below:

No.	Title	Lead Author(s)
1	Summary of Findings	P Duerden, D A Lever, D A Sverjensky and L R Townley
2	Geologic Setting	A A Snelling
3	Geomorphology and Paleoclimatic History	K-H Wyrwoll
4	Geophysics, Petrophysics and Structure	D W Emerson
5	Hydrogeological Field Studies	S N Davis
6	Hydrogeological Modelling	L R Townley
7	Groundwater Chemistry	T E Payne
8	Chemistry and Mineralogy of Rocks and Soils	R Edis

9	Weathering and its Effects on Uranium Redistribution	T Murakami
10	Geochemical Data Bases	D G Bennett and D Read
11	Geochemical Modelling of Secondary Uranium Ore Formation	D A Sverjensky
12	Geochemical Modelling of Present-day Groundwaters	D A Sverjensky
13	Uranium Sorption	T D Waite
14	Radionuclide Transport	C Golian and D A Lever
15	Geochemistry of ^{239}Pu, ^{129}I, ^{99}Tc and ^{36}Cl	J T Fabryka-Martin
16	Scenarios	K Skagius and S Wingefors

This Summary Report was compiled from the above information by Peter Duerden and Peter Airey.

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EXECUTIVE SUMMARY

The Koongarra uranium deposit in the Alligator Rivers Region of the Northern Territory of Australia was studied as a natural analogue of the far field behaviour of high level waste repositories following groundwater ingress.

The Koongarra analogue incorporates the following elements: the evolution of the structural geology and the primary ore; the weathering processes; the evolution of the uranium dispersion fan; the formation and transport of transuranic elements and fission products; the distribution of uranium series nuclides between the mineral phases of the host rock; phase transformations; flow path characteristics and their variability; and transport of uranium series nuclides by groundwater and colloids.

Since accessible timescales for relevant naturally occurring processes range from less than 1 to about 3 M years, laboratory investigations supporting radionuclide transport modelling can be set within timeframes which span by at least an order of magnitude those of interest to performance assessment.

Fifteen mathematical modelling approaches were developed for processes as diverse as groundwater transport, host rock weathering, radionuclide sorption, evolution of the uranium dispersion fan and the distribution of uranium series nuclides between mineral assemblages in weathered host rock. Some of these models are relevant to performance assessment at the level of individual processes and subsystem performance. The modelling of the evolution of the secondary mineralisation at Koongarra involves a level of complexity of a similar order to the modelling of the performance of an overall repository system. A better understanding of the scope and limitations of different concepts and models was obtained using a methodology similar to that in a joint SKI/SKB scenario development project. The ARAP was accepted as one of the test cases within Phase I of the NEA International INTRAVAL Project.

Through the ARAP, new insights into the application of the natural analogue approach to the assessment of potential waste repository sites were obtained.

ALLIGATOR RIVERS ANALOGUE PROJECT (ARAP)

1. INTRODUCTION

It is currently accepted that extrapolations of laboratory derived data to performance assessment studies of nuclear waste repositories over long times and distances are not entirely satisfactory. Consequently, a concept based on modelling the results of laboratory studies, field experiments and natural analogue studies has been developed and is being used in many countries.

Chapman et al (1984) discussed in detail the application of natural analogue studies to a range of potential problems at all stages of a waste disposal strategy. Birchard (1989) suggested that although analogues may provide only a first order validation of radionuclide transport over the thousand to million year timescale, this level of validation may be sufficient for high-level waste performance assessment. The major uncertainties of natural systems are the initial and boundary conditions which need to be applied in modelling. However, the difficulties presented to the modeller when trying to establish the behaviour of the system over the past hundreds of thousands of years is only marginally more complex than the problem presented to the designer of a nuclear waste repository who has to predict its long term behaviour into the uncertain future.

Petit (1992) warns that reasoning by analogy should be used with caution but stresses that its role "should not be underestimated, notably for the safety assessment of a disposal concept in a geological formation and for the communication of scientific results to the general public, who are not trained to the scientific method".

2. URANIUM ORE-DEPOSITS AS NATURAL ANALOGUES

Uranium orebodies contain a wide range of radionuclides and heavy metals of direct interest to waste repository design, including actinides, radium isotopes, lead isotopes and rare earth elements, as well as ultra-low levels of significant transuranic elements (^{239}Pu, ^{237}Np) and fission products (^{99}Tc, ^{129}I). Hence, they provide the opportunity for direct measurement of the migration of relevant radionuclides that has occurred over geological time and allow a reduction of uncertainties in the prediction of long-term transport of these radionuclides. The accessible timescales range from <1 year to about 500,000 or more years, depending on the process being studied. As uranium series elements are ubiquitous and can be measured at very low levels, there is also the possibility of transferring information from the analogue to any proposed repository site.

A number of uranium ore deposits have been considered as the basis of such studies, including the Oklo deposits in Gabon, ore deposits near Pocos de Caldas in Brazil, the Tono Mine in Japan, the Alligator Rivers region of the Northern Territory of Australia and numerous smaller uranium deposits including those at Palmottu, Finland; South Terras, England and Needle's Eye and Broubster in Scotland. All these studies have been carried out with the aim of improving the basis for understanding natural rock-water interactions and their effect on radionuclide transport; however, their focus has varied as a result of the different nature of the sites.

3. REVIEW OF PREVIOUS WORK IN THE ALLIGATOR RIVERS REGION

3.1 Early Investigations

There was little scientific study of the Alligator Rivers region of the Northern Territory (Figure 1), prior to the discovery of uranium. Although the first European explorers came to the region in 1845, early efforts to develop a pastoral industry in the Northern Territory had little impact. The area around Oenpelli, where recording of climatic data commenced in 1910-11, was included in an Aboriginal reserve in 1920 with a mission station being opened in 1925. After World War II, efforts were made to obtain more information about the region. A land resource survey was carried out in 1946 by the Commonwealth Scientific and Industrial Research Organisation (CSIRO) and a 1948 American-Australian scientific expedition to Arnhem Land reported on the existence of Aboriginal paintings and provided information about vegetation and wildlife.

Following the 1953 uranium discovery at El Sherana in the Alligator Rivers Region there were extensive searches for further deposits, although lack of accessibility meant that at that time the region was still preserved in a near natural state. Airborne spectrometer surveys in 1969, followed by ground exploration of anomalies, led to the discovery of the Koongarra, Nabarlek, Ranger and Jabiluka uranium deposits. A systematic geological and geophysical survey was then started by the Bureau of Mineral Resources (now the Australian Geological Survey Office) and in 1972 the Australian Government and the mining companies initiated a series of fact-finding surveys and investigations as part of an overall base-line study of the Alligator Rivers Region. A major focus of this study was to investigate the sensitivity of the region to various kinds of development, a direction which eventually led to Government policy that mining should only proceed at two of the deposits (Ranger and Nabarlek).

3.2 USNRC Analogue Project 1981-87

The USNRC funded study *Radionuclide Migration around Uranium Ore Bodies - Analogue of Radioactive Waste Repositories* was directed towards identifying processes of potential significance over long timescales to the far field behaviour of a high-level waste repository (Airey et al., 1986). Initially the work encompassed the four major ore deposits in the region; in the final stages of the project all work was focussed on Koongarra.

Four main themes were developed:

1. *Stationary phase:* Particular emphasis was placed on the development of sequential extraction schemes for the study of the distribution of uranium series on identified mineral phases within the host rock;

2. *Groundwater and Colloids:* Previous groundwater investigations were extended to include D/H, ^{14}C, $^{234}U/^{238}U$ and $^{230}Th/^{234}U$ as well as trace elements and tritium. In addition, an Amicon ultrafiltration system was adapted to concentrate the colloid fraction in pumped groundwater from the orebodies out of contact with air. It was found that negligible uranium isotopes were associated with the colloids, though significant amounts of thorium were present.

   A separate UKDoE funded Koongarra colloid study, undertaken in 1986, extended earlier work and developed equipment for in-field concentration of colloids and fractionation into size ranges while ensuring that the groundwater chemistry and dissolved oxygen levels were unchanged (Ivanovich et al 1988).

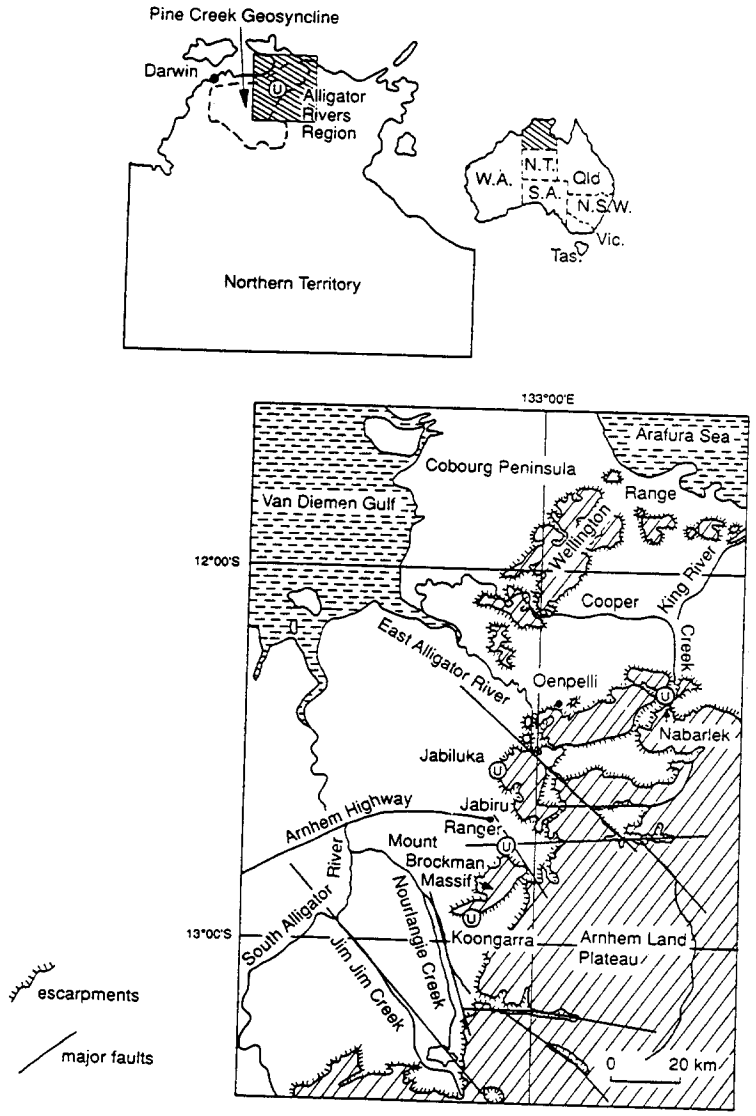

FIGURE 1 Location map showing the four uranium deposits in the Alligator Rivers Region, the sandstone escarpment and major faults (Snelling, ARAP Vol. 2, adapted from Needham and Stuart Smith, 1976).

3. *Fission Products (Iodine-129), Plutonium-239 and Chlorine-36:* Preliminary work was carried out to extend the geochemical natural analogue to fission products and transuranic elements (^{239}Pu). Groundwater samples were collected throughout the project for ^{129}I and ^{36}Cl analysis, and chemical procedures were also developed to separate environmental levels of ^{36}Cl and ^{129}I from host rock. The isotopic analyses were made by accelerator mass spectrometry at the University of Rochester. The aim of the measurements was to compare the measurements of ^{129}I in the host rock and groundwater with production rates from neutron induced and spontaneous fission. However, the task was not completed during this phase of the work.

4. *Modelling:* An open system uranium model was developed to describe the gross features of the ore deposits. Initially it was used to model the rate of advance of the weathering front (on the basis of this approximately balancing the rate of surface erosion). It was then extended to the multiphase system, which had been defined on the basis of the results of the sequential extraction measurements. The open system model was then used to quantify the rate of redistribution of uranium at Koongarra, with the model being calibrated against uranium series data. The work suggested that the time period for the secondary mineralisation was in the range 1-3 million years.

4. THE ALLIGATOR RIVERS ANALOGUE PROJECT (ARAP)

Extension of the analogue work through OECD Nuclear Energy Agency (NEA) sponsored ARAP project was agreed in 1987 (Duerden 1992). Funding was provided by the organisations listed in Table 1 to support activities of an extensive research team (Table 2). The Government of the Northern Territory of Australia provided separate funding for drilling new wells at the site.

The overall aim of the study was:

- To contribute to the production of reliable and realistic models for radionuclide migration within geological environments relevant to the assessment of the safety of radioactive waste repositories, and
- To develop methods of validation of models using a combination of laboratory and field data associated with the Koongarra uranium deposit.

The study was directed towards understanding the evolution of the secondary uranium mineralisation and the distribution of radionuclides within the dispersion fan. The work was focused on investigating the original weathering of the Koongarra region, the groundwater flow, the alteration of the host rock and primary uranium, the rock/groundwater interactions including adsorption desorption, radionuclide transport, the development of the dispersion fan, and the in-situ production and mobility of long lived fission products such as ^{99}Tc and ^{129}I and transuranic nuclides such as ^{239}Pu.

The experimental programs also resulted in the development of substantial drillcore and groundwater databases for all the distinctive geologic units in the vicinity of Koongarra. It has therefore been possible to test various aspects of radionuclide solubility/speciation calculations, of surface complexation models and of theoretical models for the formation of the various zones of specific secondary uranium minerals.

Table 1. ARAP Agreement Funding Organisations

ARAP Agreement Signatories	
Participants	
Australian Nuclear Science and Technology Organisation	(ANSTO)
Japan Atomic Energy Research Institute	(JAERI)
Swedish Nuclear Power Inspectorate	(SKI)
UK Department of the Environment	(UKDoE)
US Nuclear Regulatory Commission	(USNRC)
ARAP Associate Participant	
Power Reactor and Nuclear Fuel Development Corporation of Japan	(PNC)
Additional Support	
US Department of Energy	(USDOE)
Central Research Institute for Electric Power Industry, Japan	(CRIEPI)

Table 2. Collaborating Laboratories and Principal Investigators

AUSTRALIA		
ANSTO	Mineralogy/Colloids/Rock U Series Transport Modelling Groundwater Chemistry/Sorption Sorption Modelling PIXE/PIGME Uranium Analysis - DNAA Tritium Analysis ^{14}C Measurements	R Edis C Golian T Payne D Waite D Cohen T Wall R Stokes L Smith
ANSTO	AMS ^{36}Cl	J R Bird D Fink C Tuniz A V Jenkinson
ANU	AMS ^{36}Cl	K Fifield G Allen
Consultant	Geology	A Snelling

CSIRO Energy Chemistry	ICP-AES/ICP-MS Anions/Cu TOC	L Dale G Batley R Matthews
CSIRO Water Resources South Australia	Hydrogeochemistry Stable Isotope Measurements	A Herczeg F Leaney
CSIRO Water Resources Western Australia	Hydrology Modelling	L Townley M Trefry R Salaama A D Barr
NT Mines & Energy Dept	Groundwater Sampling/Hydrology	B Treloar
NT Power & Water Authority	Geomorphology/Groundwater/ Geophysics	P Jolly D Pidsley K Martin
Sydney University Geophysics	Petrofabric Analysis and Field Geophysics Measurements	D Emerson K Mills L Q Cao M Hallett
AUSTRALIA		
Sydney University Soil Science	Pedology/Thin Section Analysis	T Koppi D Klessa
University of Western Australia	Geomorphology and Palaeoclimates	K-H Wywroll
JAPAN		
Akita University	Uranium Redistribution	S Nakashima
CRIEPI	Borehole TV Investigations	K Miyakawa
JAERI	Alteration ^{234}Th Transport Modelling Sorption/U Series data Rock U Series/Groundwater, Ra Spectroscopy	T Murakami T Ohnuki K Sekine N Yanase T Nagano
PNC	Groundwater and Rocks REE	T Seo
SWEDEN		
KEMAKTA	Transport Modelling Hydrochemistry	K Skagius K Pers F Brandberg M Lindgren
SKI	Scenario Development/Validation	S Wingefors

UNITED KINGDOM		
W S Atkins	Geochemical Modelling	D Read D Bennett
UKAEA, Harwell Laboratory	Transport Modelling/ Hydrology Modelling	D A Lever A J Baker M Ivanovich G Longworth
USA		
Johns Hopkins University	Geochemical Modelling	D Sverjensky J Raffensperger
LANL	^{36}Cl, ^{99}Tc, ^{129}I, ^{239}Pu Analysis and Production Rate Modelling	D Curtis J Fabryka-Martin M Attrep F Roensch
PNL	Hydrology (Fracture) Modelling	J Smoot
United States Geological Survey	Sorption Modelling	J Davis
University of Arizona	Hydrogeology	S N Davis B Marley S Braumiller J Norris
University of New Mexico	Alteration of Uranium Minerals	R C Ewing H Isobe*
USNRC	Geochemical Modelling	G Birchard
* On attachment from JAERI		

Three independent methods of describing the evolution of the hydrogeochemistry of the system have been compared to give confidence that an adequate understanding of the radionuclide transport has been gained. The first was a study of the geomorphology of the site, so bounds could be set on the time frames within which the radionuclide processes occurred. The second was an improved understanding of the mechanism of the groundwater induced transport of key elements now and in the past. The third method made use of the distinctive information from uranium series disequilibria and provided data for the calibration of radionuclide transport and performance assessment models.

5. SITE CHARACTERISATION

5.1 Geology

The Koongarra uranium deposit lies about 25 km south of the small town of Jabiru in the Northern Territory of Australia. Koongarra is one of four major uranium deposits, which are situated either in valleys between eroded outliers of the Arnhem Land Plateau or on the edges of the lowland floodplains in the lower reaches of the major river systems. All of them are adjacent to prominent sandstone escarpments, Koongarra being in the valley between the main Arnhem Land Plateau and the Mt Brockman Massif outlier adjacent to the escarpment (Figure 2).

The uranium mineralisation at Koongarra occurs in a layered Lower Proterozoic sequence which was metamorphosed to schists and dolomite between 1,870 and 1,800 My, accompanied by severe folding. This was followed by a ~150 My period of uplift, weathering and erosion which produced a new land surface on which thick layers of sandstone (the Kombolgie Formation) were then deposited (probably between 1,690 and 1,600 My).

The Koongarra deposit today lies beneath a gently sloping ground surface below the Kombolgie sandstone escarpment of the Mount Brockman Massif. The uranium occurs in two distinct orebodies (Figure 3), separated by about 100 m of barren schists, and consists of partially coalescing lenses that are elongated and dip at 55° broadly parallel to the reverse fault breccia which forms the footwall to the ore zone (Figure 4). The movement along the fault has reversed the normal sequence of strata by bringing the overlying younger sandstone down underneath the older tilted schist layers. This fault zone forms the lower boundary (or footwall) to the uranium ore zone. The strongest mineralisation with grades in excess of 1%U over several metres are just below a distinctive sheared graphite-quartz-chlorite schist unit that forms the hanging wall to the ore. Immediately above, and up-dip of, this primary ore in both the transitional zone and the weathered schist is ore that was obviously once primary but now is weathered. Consequently it is here designated as weathered primary ore. Adjacent to it and downslope of it, again within the weathered schist and transitional zone, is ore that represents uranium which has been mobilised from the weathered primary ore and dispersed downslope to form what has been designated as the dispersion fan. The weathered primary ore and the dispersion fan together are here called the secondary ore.

The No. 1 orebody (Figure 3) is elongated over a distance of 450 m and persists to a depth of about 100 m. Secondary uranium mineralisation, derived from leaching of the primary mineralised zone is characterised by uranyl phosphates. It is present from the surface down to the base of weathering over some 25-30 m and forms a tongue-like body of ore-grade material dispersed down-slope for about 80 m to the south-east. Away from the tail of the dispersion fan the uranium appears to be dispersed in the weathered schists and is apparently adsorbed onto the surfaces of clay and iron oxide minerals. Uranyl phosphates have not yet been identified in this area, although they have been the focus of an extensive mineralogical study.

In the No. 2 orebody, the mineralisation is elongated over a distance of about 100 m and persists down dip at 55° to at least 250 m. The primary ore consists of uraninite veins that crosscut the foliation of the brecciated and hydrothermally altered host schist.

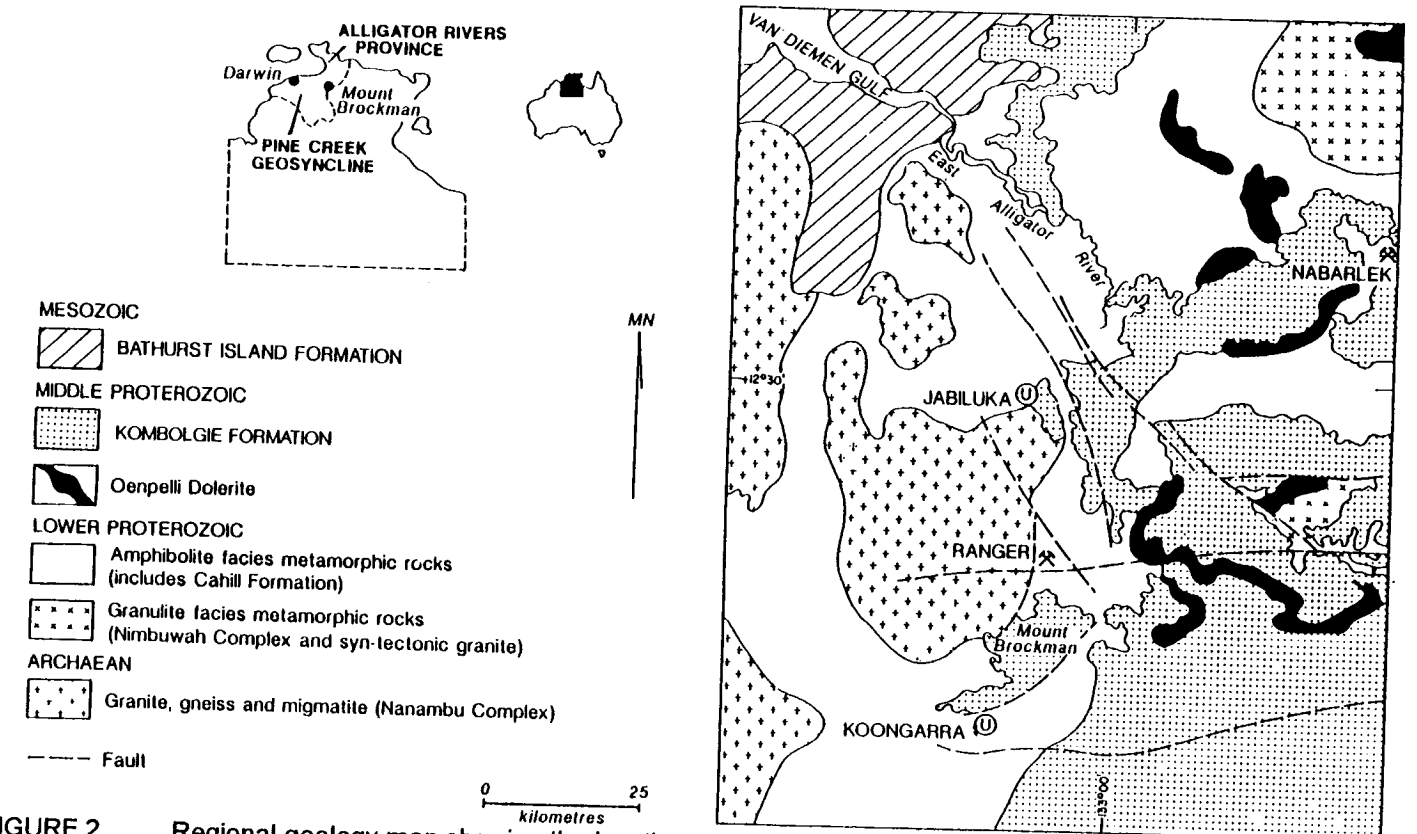

FIGURE 2 Regional geology map showing the location of the Koongarra uranium deposit (Snelling, ARAP Vol. 2, after Needham, 1984).

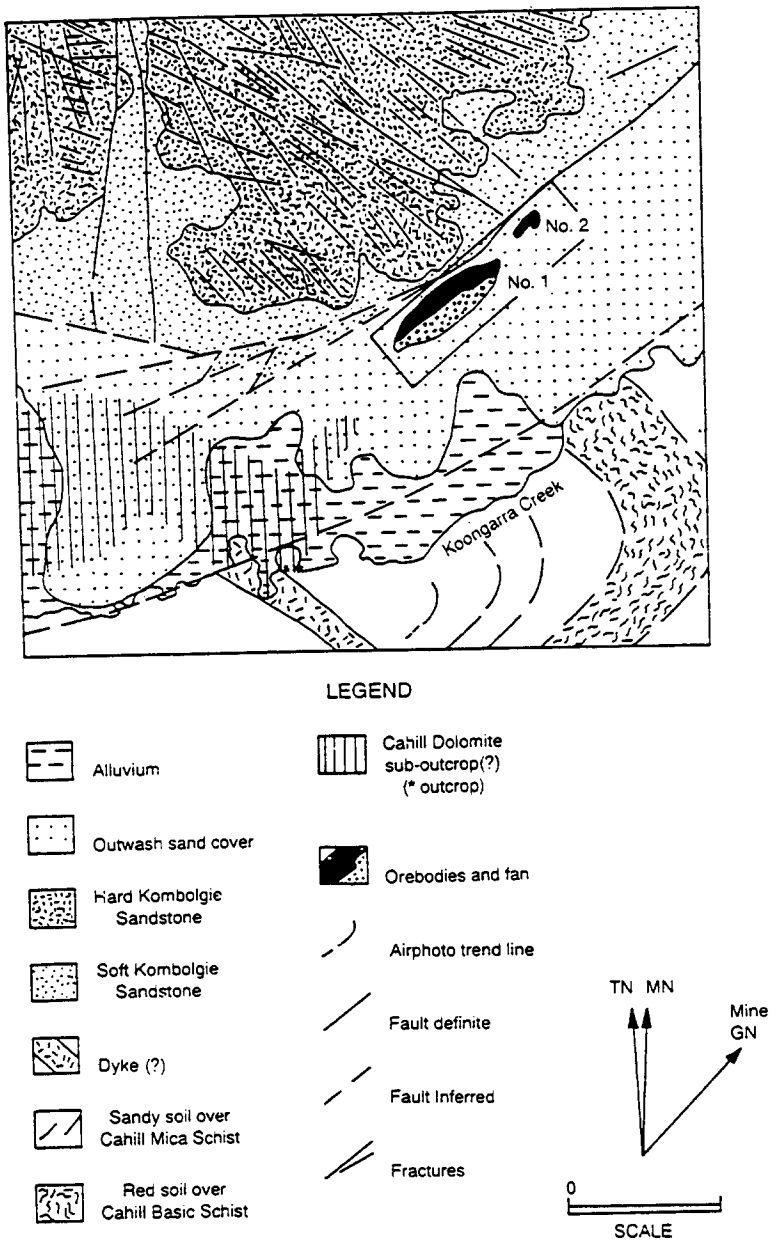

FIGURE 3 Koongarra area geology (Snelling, ARAP Vol. 2, Emerson et al., ARAP VOL. 4).

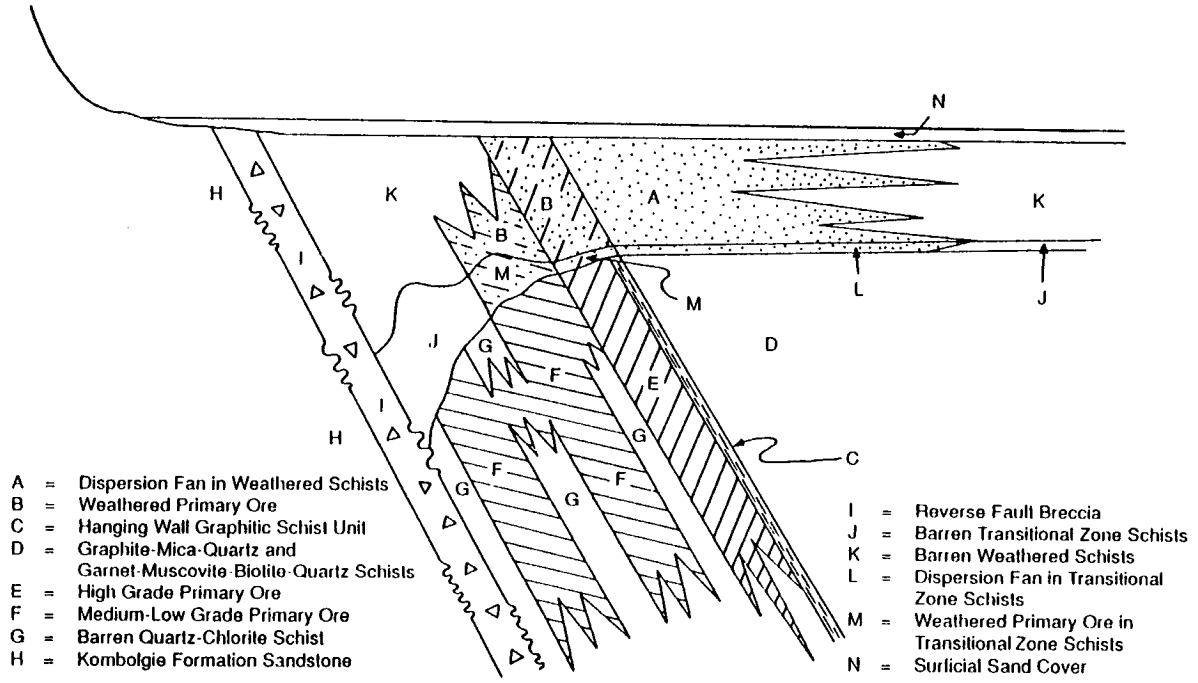

FIGURE 4 Schematic cross-section through the Koongarra No. 1 Orebody showing the rock units of the mine sequence, the weathered and transition zones, and the definitions of the ore zones (Snelling, ARAP Vol. 2).

5.2 Geomorphology and Hydrogeology

On the basis of an evaluation of the structural geology of the site it now appears that the retreat of the escarpment has played no part in the development of the dispersion fan associated with the uranium deposit. The conclusions of this work point to the fact that the dispersion fan could only have come into existence fairly recently, i.e. during the last 1-2 My. The assumption is made that the present dispersion fan was not preceded by a previous dispersion fan(s) which has been lost in the surface lowering. The overall lowering of the Koolpinyah surface in the Koongarra region may have been of the order of 100 m since the Kombolgie cover was lost.

A number of different approaches have been made to try to understand the dynamics of groundwater flow through the orezone region; whether fluid flow is through fractured or porous media or both; and in particular, how this has influenced the transport of uranium and the formation of the dispersion fan (see insert, 'Geophysics, Petrophysics and Structure'). There are two major potential inputs of groundwater. The first these is flow from the vicinity of the Koongarra fault into the Cahill formation which hosts the uranium mineralisation. A second major source is infiltrating waters which permeate downwards from the surface, and cause a gradual mixing and dilution of the characteristics of groundwaters from the mineralised zone. As a result, the chemistry of shallow groundwaters is dependent on depth. Further details are provided in the insert, 'Hydrogeology of Koongarra'.

During the last two million years the area would have experienced significant climate changes though the nature of these is unclear. However, by using the events of the Late Quaternary as a guide, indications are that climate states since the Middle (Early?) Pleistocene consisted of long periods (100 Ka) of drier conditions interrupted by shorter periods (10 Ka) of conditions similar to those prevailing in the area today.

Understanding the hydrogeology of the site was an important aspect of the project with a major effort being made to use existing hydrogeological flow models to interpret available data and to predict rates and directions of groundwater flow. The Koongarra site has proved to be complex and difficult to model because sub-surface flow is a minuscule component of the overall water balance of the region. Significant flow occurs as fracture flow rather than as porous media flow, the region is extremely heterogeneous and many of the available hydrogeological measurements were collected from exploration holes constructed to determine the extent of the uranium ore, not to understand groundwater flow.

All but one of the models used were porous media models, which assumed that the fractured medium acts as a continuum and that flow is governed by Darcy's Law with some effective average hydraulic continuity; the other was a fracture flow model, based on generating a finite number of discrete fractures with random orientations. The objective of determining rates and directions of groundwater movement was not achieved with any degree of confidence. However, each type of model was able to describe some individual features of the site and the hierarchy of all of the models used by the number of groups working in various locations worldwide was able to provide a better understanding of the Koongarra hydrogeology.

HYDROGEOLOGY OF KOONGARRA

From a hydraulic viewpoint the geologic materials can be divided into five groups.

1 *Kombolgie Formation*, a late Precambrian sandstone with low to moderate porosity and low hydraulic conductivity (Figure 4H).

2 *Surficial sands, silts, and clays* which cover most of the site and extend from the present-day land surface to depths of 0.5 to 9 m (Figure 4N). The sands of the surficial sediments are probably the most permeable of the geologic materials. During the dry part of the year, most of the surficial materials are not saturated.

3 *Residual clay grading downward into moderately weathered Cahill schists* (Figure 4A-K). The clay has a thickness which varies from about 2 to 20 m. It is porous but has, in general, a very low hydraulic conductivity.

4 *Partly weathered and fractured Cahill schist.* Under present climatic conditions, this zone is saturated throughout the year and constitutes an aquifer which is about 10 to more than 30 m thick. The aquifer is extremely heterogeneous and in many locations is dominated by fracture flow which imparts an apparent anisotropy to the flow field.

5 *Unweathered Cahill schist* in which water is generally semistatic owing to very low porosities and permeabilities (Figure 4D). Although some fracture flow undoubtedly exists at depths in excess of 1.0 km, the bulk of the groundwater flow in the Cahill schist is probably at depths of less than 100 m.

Seasonal variations of water levels in about 60 wells at the site have been monitored for periods exceeding 7 years. The levels show groundwater recharge in the wet season and discharge in the rest of the year and have defined southerly hydraulic gradients. Long-term aquifer tests were completed at various times during 1980, 1987, 1988, 1989. All drawdown cones were elongated in general northeast-southwest to north-south directions. Three of the aquifer tests showed highly localised drawdowns in a narrow band suggesting strongly that most of the groundwater flowing into the pumped well was transmitted through well-defined fractures. Transmissivities varied from 0.22 to 66.8 m^2/day with a median value of 26 m^2/day. Most water was pumped from the partly weathered Cahill schist. If the thickness of the water-bearing zone is assumed to be 40 m, then an average hydraulic conductivity of 0.65 m/day is suggested.

5.3 Chemistry and Mineralogy of Rocks and Solids

The chemistry and mineralogy of rock and soil at Koongarra have been investigated to gain an understanding of the mechanism of retardation of uranium series radionuclides over long timescales.

The main mineral constituents of the host schists are quartz, chlorite and muscovite. Small amounts of graphite, sulfides, garnets, feldspars and remnant biotite also occur, particularly outside of the primary ore zone. The forms and abundances of the mineral constituents greatly affect weathering processes, final products, groundwater chemistry and radionuclide-mineral interactions. Within the primary ore zone, the nature of the chlorite is mainly magnesium-rich. Outside of this inner halo of hydrothermal alteration, iron-rich chlorite tends to dominate. In the weathered zone above the primary ore body, kaolinite is the only significant clay mineral. Near the base of weathering, there is a thin layer of vermiculite. Down-gradient, where the degree of hydrothermal alteration of the rock is less, smectite also becomes a significant component of the weathered zone.

Goethite, hematite and ferrihydrite are the major iron oxides present at Koongarra. They occur as fissure and remnant schistosity coatings, dispersed clay coatings, and nodules. The different forms of iron oxide probably play different roles in the capture and storage of otherwise mobile species. The amount of iron oxides is very variable, with occasional bands of bleached clay juxtaposed with iron rich bands. The greatest heterogeneity in the distribution of iron appears to be with the crystalline phases. The concentration of ferrihydrite, estimated by chemical extraction, was between 7 and 29 mg/g, making up an average of 10% of the total iron. Lithiophorite is the only manganese mineral identified in the weathered zone, and mainly occurs coating fissures and remnant schistosity. Coatings of iron and manganese oxy-hydroxides on pore and fissure surfaces are well placed for contact with migrating species (Figure 5).

Associations between various elements, and between elements and minerals were examined. In the unweathered rock, uranium appears to be associated with a few trace elements, but not with any major rock forming elements. There has been very little penetration of uranium into the quartz chlorite matrix from fissures; an apparent diffusivity of 10^{-24} m^2s^{-1} was estimated.

In the secondary ore body (weathered), uranium is correlated with phosphorus, and the uranyl phosphate mineral saleeite has been detected, particularly near the base of weathering. In the weathered zone, uranium was found to be associated with secondary oxides of iron and manganese. Phase selective extraction schemes showed that the uranium in the weathered zone is mainly accessible to the groundwater (adsorbed or associated with amorphous phases, 10-73%), or entrapped within crystalline iron oxides (<27-82%). The relationship with iron oxy-hydroxide appears to be of a diffuse nature, indicative of sorption as the initial mechanism for association. However within manganese phases, uranium is almost exclusively associated with "hot spots" containing a cerium oxide phase.

Uranium series disequilibria have been extensively examined, particularly in the weathered zone of Koongarra. The isotopes of central interest were ^{238}U, ^{234}U, ^{230}Th and ^{226}Ra. In the unweathered zone (E, Figure 4), the ore material and the host rock were at approximate secular equilibrium, except for a small apparent depletion of ^{226}Ra. In the weathered rock, above the primary mineralised zone (B, Figure 4), high $^{230}Th/^{234}U$ activity ratios indicate strong recent leaching of uranium. In these leached areas, the $^{234}U/^{238}U$ activity ratios are high; above unity in the bulk rock and near unity in the accessible phases (extractable with Tamms acid oxalate (TAO)). This indicates a net preferential leaching of ^{238}U relative to ^{234}U. The most intensely leached samples came from near the surface, near the up-flow boundary of the primary ore (M, L, Figure 4). The most intense recent accumulation of uranium appears to have occurred near

the base of weathering, about 0-50 m from the down-flow boundary of primary ore. This highlights the importance of the early stages of weathering to the migration and retardation of uranium (Section 6.1).

In summary, considerable information has been gathered on the migration of uranium at Koongarra, and, inparticular on the importance of early stages of weathering in radionuclide mobility; the behaviour of uranium in weathered environments; the development of isotopic disequilibria under weathering conditions; the behaviour of rare earth elements, and the associations of uranium with mineral phases.

5.4 Groundwater Chemistry

Groundwaters in the mineralised zones at Koongarra exhibit elevated levels of uranium and the radionuclides of its decay series. The pH is slightly acidic or neutral, and the major ion chemistry is dominated by magnesium and bicarbonate. Groundwaters are quite dilute, with total dissolved solids (TDS) below 200 mg/L. In many groundwater samples, partial pressures of CO_2 are substantially elevated relative to those of surface waters.

When groundwaters intersect the deposit, uranium concentrations rise steeply, to levels up to four orders of magnitude above background concentrations. Down-gradient, there is a gradual decrease over approximately 200 metres. The $^{234}U/^{238}U$ isotope ratio is strongly dependent on depth. In groundwaters of the weathered zone, $^{234}U/^{238}U$ ratios are often substantially below unity. These low ratios are correlated to those in accessible (readily leached) uranium of the solid phase. In contrast, the groundwater $^{234}U/^{238}U$ ratios in the unweathered zone generally exceed unity.

Uranium appears to be mobile in the weathered zone as uranyl carbonate complexes. Other inorganic uranium complexants are not present at levels sufficient to influence uranium speciation, with the possible exception of phosphate. Uranyl phosphate minerals occur in the secondary uranium mineralisation and the levels of phosphate in some Koongarra groundwaters are unusually high. Total organic carbon levels are generally low, suggesting that uranium complexation by organic species plays a minor role.

The measured concentrations of thorium isotopes in Koongarra groundwaters were extremely low, with $^{230}Th/^{234}U$ ratios below 0.001. Therefore, ^{230}Th is substantially less mobile than ^{234}U and can be regarded as immobile for modelling purposes. Radium is mobile in deeper groundwaters, and may be precipitated, together with manganese and ferric oxyhydroxides, at the base of the weathered zone. The distributions of ^{238}U and other isotopes, such as ^{222}Rn and ^{210}Pb, suggested dispersion towards the south, as well as to the south-east.

Dissolved CO_2 in Koongarra groundwater plays an important role in weathering of minerals such as chlorite. This process is responsible for the elevated levels of Mg^{2+} and HCO_3^-. Groundwater in the centre of the orebody has a low proportion of ^{14}C. Therefore, the CO_2 is not, in the main, derived from the root zone of plants. Based on ^{13}C data, a possible ultimate source of the CO_2 is bacterial degradation of organic material, such as the dispersed graphite in the Cahill formation. Another significant input of CO_2 into the system may be the CO_2 present in up-gradient (Kombolgie) groundwaters.

Iron seems to play an important role in the system. The state of saturation of groundwaters with respect to amorphous iron minerals is correlated to measured redox values, and some iron-rich colloids are present. Iron minerals adsorb uranium strongly, and adsorption may be a more important mechanism for uranium retardation than mineral precipitation as solubility products for uranium minerals are rarely approached, except in the immediate vicinity of the orebody.

Colloids are a relatively unimportant mechanism for radionuclide transport in Koongarra groundwaters, and the total number of colloidal particles is very low. Uranium migrates mostly as dissolved species, whereas thorium and actinium are mostly associated with larger, relatively immobile particles (>1.0 μm). However, of the small amount of ^{230}Th that passes through a 1.0 μm filter, a significant proportion is associated with colloidal particles. These colloids mostly consist of iron oxides and clay minerals and carry a small proportion of the uranium in the groundwater. The clay minerals in the colloids reflected the mineralogy of the solid phase from the vicinity of the water sampling point. Actinium appears to be slightly more mobile than thorium and is associated with colloids to a greater extent, although generally present in low concentrations.

In summary, groundwater enters the Cahill formation from the vicinity of the Koongarra fault. The groundwater in the mineralised zone has elevated levels of HCO_3^-, Mg^{2+}, uranium isotopes, and ^{238}U-series radionuclides; and low levels of ^{14}C. Uranium is present in a true dissolved (rather than colloidal) form, probably as uranyl carbonate complexes. As it moves down-gradient, the groundwater is gradually diluted and mixed with local recharge derived from seasonal rainwaters. Dilution and adsorption appear to be the main processes which reduce uranium concentrations once the groundwater is outside the immediate vicinity of the primary ore-zone.

GEOCHEMICAL DATA BASES

During the Project a range of geochemical and hydrogeochemical models were developed to account for measured data from the site and with which to predict site evolution. The majority of these models are based on the premise of thermodynamic chemical equilibrium and employ fundamental thermodynamic data to characterise the chemistry of the system.

The computer codes EQ3/6, PHREEQE, MINTEQ were most commonly used during the Project for geochemical modelling. It is concluded that the degree of uncertainty in geochemical modelling studies arising as a result of using one code rather than another is relatively insignificant when compared to that related to differences in the underlying data bases.

A detailed review of thermodynamic data for uranium and thorium bearing species was also undertaken as these are of particular relevance to Koongarra and its status as a natural analogue from radionuclide transport from a radioactive waste repository. The review considered methods for extending the thermodynamic chemical models to non-standard state conditions such as variations of temperature, pressure and salinity. It is concluded that for the present day Koongarra site, relatively less uncertainty results in making geochemical conditions at standard state conditions (25°C, 1 bar), where the thermodynamic data is best known, than would be the case if the data were extended to cover the slightly higher average temperatures recorded at the site. The data requirements of the two models most often used in the project for sorption, the Triple Layer Model and the Diffuse Double Layer Model have also been described and their availability given.

GEOPHYSICS, PETROPHYSICS AND STRUCTURE

In the geophysical, petrophysical and structural program, the site was studied at regional, field (local), lithological unit and core sample scales by geophysical, geological, and, where appropriate, geotechnical techniques, to determine lithological and structural features relevant to ancient and current groundwater flow. The initial focus of the study was the determination of the physical properties of 306 core samples from 29 drillholes penetrating the various units.

Twelve kilometres of geophysical traverses were completed on the site. Magnetic, gravity, radiometric, spontaneous potential (SP), electrical resistivity and electromagnetic techniques were used to elicit lateral and vertical lithological information, structural trends and discontinuities, and to infer hydrogeological characteristics. Field structural studies were integrated with a borehole television (BTV) mapping program which was carried out over a total length of 469 m in 20 boreholes, to identify geological features, such as the orientations of the schistosities, fracture densities, (*in situ*) aperture widths and orientations. The BTV study showed that the aquifer tests were dominated by fractures in the deeper unweathered zone and identified the dominant schistosity and fracturing that can account for the main trend of the dispersion fan.

The spatial contours of uranium concentration in the weathered zone show that although high grade uranium concentrations can be seen to have migrated in the weathered schist from the primary zone towards the southeast direction a more extensive dispersion fan of uranium (non-oregrade but above background levels) is detected towards the south of the orebody, about 300 m from the primary source. The distribution is clearly similar to that identified from the groundwater chemistry and the geophysical measurements, both of which suggest a present-day southerly direction of flow from the orezone.

6. RADIONUCLIDE TRANSPORT PROCESSES

6.1 Weathering and its Effect on Uranium Redistribution

When we consider radionuclide migration over a geologic timescale, the host rocks around a site should not be thought of as static but as dynamic in nature. The rock-forming minerals will be changing structurally and chemically with time by water-rock interactions, so-called weathering/alteration. Thus, knowledge of the weathering mechanisms is essential along with the sorption and desorption characteristics of the radionuclides if we are to understand migration over periods up to millions of years. In the vicinity of the uranium ore deposit at Koongarra, quartz-chlorite schist, the ore host rock, has been subjected to weathering. Although quartz is resistant to weathering, chlorite has been altered to clays and iron minerals. The chlorite weathering and the uranium association with the weathered minerals have been the main subjects of this study.

The simplified conversion sequence for Fe-rich chlorite weathering was found to be chlorite $\rightarrow$ vermiculite $\rightarrow$ kaolinite. These minerals occur as a function of depth; near the end of the secondary ore deposit, chlorite alone is present at 25 m and deeper, but rapidly decreases in

amount between 25 and 24 m, and disappears at 24 m. Vermiculite appears at 25 m in depth and kaolinite begins to persist at 24 m in depth. Vermiculite disappears at 20 m where kaolinite is predominant as it is in the shallower zone.

Iron and Mg are released in every stage of the weathering, ie during the transformation of chlorite to vermiculite and in the decomposition of vermiculite to kaolinite. Iron, which is closely related to the uranium association, is located between chlorite and vermiculite slabs at first (a few microns in size), and then accumulated between grain boundaries, forming veins occasionally.

Chlorite weathering has affected the uranium redistribution both microscopically and macroscopically. On a metre scale, the abundances of chlorite, vermiculite and kaolinite correspond well to uranium concentrations; the chlorite dominant zone, to the zone of lower uranium concentration; the vermiculite predominant zone, to the zone of intermediate uranium concentration; the kaolinite predominant zone, to the zone of highest uranium concentration. On a millimetre scale, the uranium concentrations are qualitatively to the extent of the weathering; weathered chlorite grains having higher uranium concentration.

Electron microprobe analysis and visible microspectroscopy have revealed that uranium mainly occurs with iron minerals, the weathered products and that sub-micron sized saleeite, a uranyl phosphate, is one of the most probably uranyl phases associated with the iron minerals. The uranium fixation mechanisms are probably saleeite microcrystal coprecipitation and sorption to the iron minerals released during the chlorite weathering.

6.2 Geochemical Modelling of Secondary Ore Formation

The purpose of the study was to establish how the uranyl phosphate zone at the Koongarra site was formed (Figure 5). The overall approach employed theoretical chemical mass transfer calculations and models that permitted investigation and reconstruction of the kinds of waters that could produce the uranyl phosphate zone. These calculations have used the geological and mineralogical data for the Koongarra weathered zone, to constrain the initial compositions and reactions undergone by groundwater during the formation of the uranyl phosphate zone. In carrying out these calculations the present-day analyses of Koongarra waters were used only as a guide to the possible initial composition of the fluids associated with the formation of the phosphate zone. The calculations are, of course, heavily dependent on the thermodynamic data file used for the calculations. Aqueous speciation, saturation state and chemical mass transfer calculations were carried out using the computer programs EQ3NR and EQ6 (Wolery, 1983; Wolery et al., 1984) and a thermodynamic data base generated at the Johns Hopkins University over the last eight years.

The calculations strongly suggest that the uranyl phosphate zone at Koongarra has not formed from present-day groundwaters which appear to be undersaturated with respect to saleeite. Furthermore, as present-day groundwaters descend below the water-table they rapidly lose their atmospheric oxygen imprint, become even more reducing in character, and more undersaturated with respect to saleeite. Because much of the phosphate zone is currently below the water table, under saturated zone conditions, it is suggested in the present study that the uranyl phosphate zone must have formed in the geologic past under unsaturated zone conditions.

The results suggest that the uranyl phosphate zone may have formed in a more arid climate that would have been associated with a much lower water table than at present. Such a climate may have prevailed, prior to the relatively recent monsoonal climate about 12,000 years ago. Based

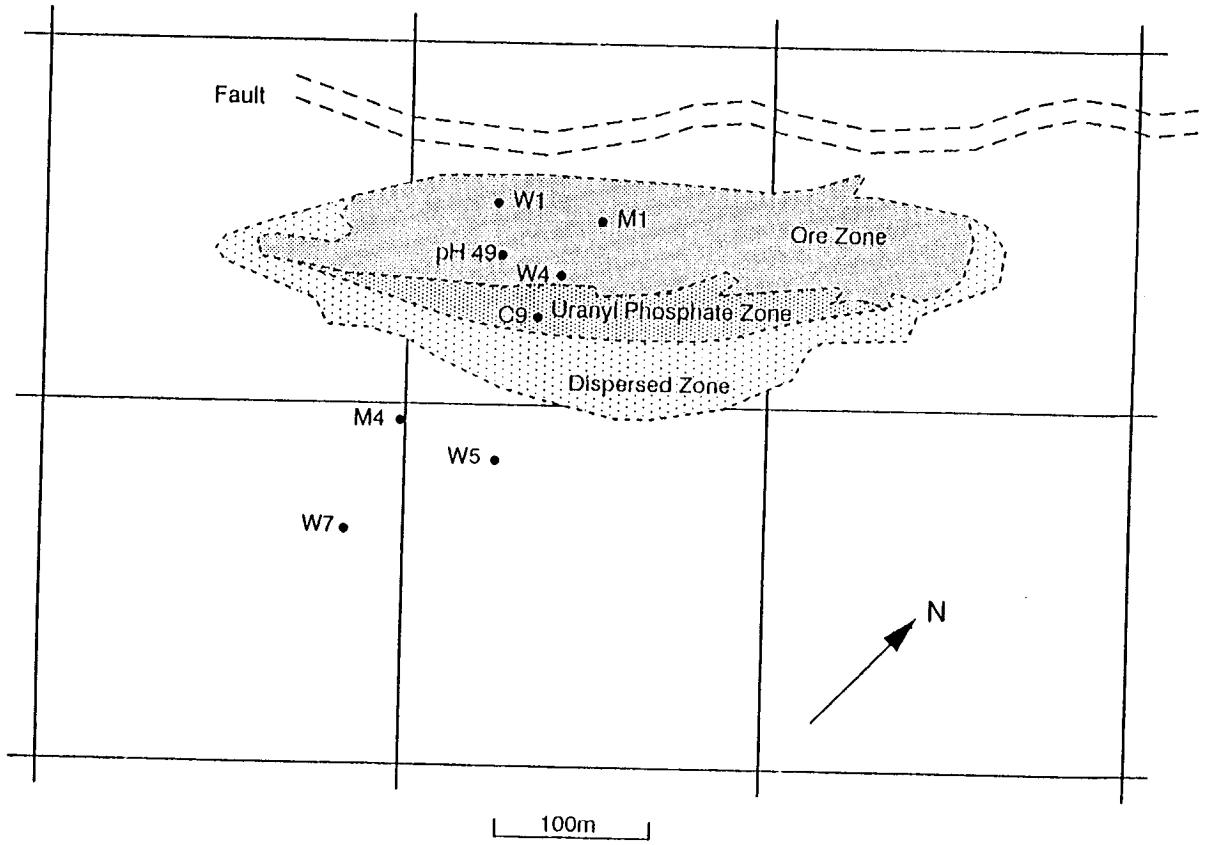

FIGURE 5 Sampling locations considered in the uranyl phosphate zone modelling study (Sverjensky et al., ARAP vol. 11).

on the above results and other studies, the geochemistry and geochemical processes at Koongarra associated with the uranyl phosphate zone can be conveniently divided into two categories according to their relevance to present-day geochemical processes and those that have operated in the geologic past. Information on the present-day geochemistry of soils, weathered rock, groundwaters, and the role of sorption processes forms a basis for a present-day analogue for nuclear waste migration. Information and inferred reconstructions of geochemical processes thought to have operated in the geologic past form a basis for a paleo-analogue for nuclear waste migration.

6.3 Uranium Sorption

While the oxidation of highly insoluble U(IV) to more soluble U(VI) followed by a sequence of U(VI) precipitation-dissolution reactions appear to have contributed to the migration of uranium away from the Koongarra ore deposit in close proximity to the deposit, adsorption-desorption processes become increasingly important in the mitigation of uranium mobility further downfield where the solubility products of U(VI) minerals are no longer exceeded.

Numerous studies of the partitioning of uranium between solid and solution phases have been reported. The distribution coefficients ("K_D values") obtained from such studies are considered to be of little use in predicting the migration of uranium through porous media of variable composition and under solution conditions that exhibit major spatial and temporal variability.

In this study, uranium sorption (adsorption and desorption) to both single, well-defined mineral phases and selected natural substrates has been undertaken. Single phases used include the amorphous iron oxide phase ferrihydrite, crystalline silica and two naturally occurring kaolinites, KGa-1 and Nichika. The surface properties of these materials have been rigorously defined and adsorption studies conducted over a range of solution pH, ionic strength, carbonate content, adsorbent and adsorbate concentrations and in the presence of uranium complexants and (potentially) competing adsorbates (such as phosphate and fluoride).

Results obtained in these adsorption studies to single phases have been modelled using the surface complexation approach with a diffuse double layer description of the electrical double layer. While a single site type was used in early studies, most success has been achieved on assuming the presence of two site types - one site type corresponding to a small set of high affinity cation binding sites and the other type corresponding to the total reactive sites available for sorption of protons, cations and anions.

Fine fractions (nominally < 10 μm) of selected natural substrates have also been prepared and uranyl partitioning between solution and solid phases examined under a variety of suspension conditions. In addition to U(VI) uptake studies using ^{236}U, studies of isotopic exchange between ^{236}U and intrinsic ^{238}U have been undertaken. The exchangeable ^{238}U observed represented "accessible" uranium and generally exhibited a direct proportionality with adsorbed ^{236}U. An estimate of the total accessible U(VI) present in natural substrates by this method was found to be in good agreement with the Tamms acid oxalate (TAO) extractable uranium in these samples. A close correspondence between the $^{234}U/^{238}U$ ratios in this extractable fraction and that in solution (after equilibration with the solid phase) was routinely observed.

A range of ancillary studies have assisted in definition of important sorbing surfaces and in elucidation of the effect of chemical processes on uranium partitioning between solid and solution phase. In particular, examination of U(VI) uptake on preformed and coprecipitated iron oxides indicates very little difference in partitioning and suggests that short term ageing of amorphous iron oxide phases does not dramatically influence the sorbing capacity. The results

of longer term ageing studies are also reported. A dramatic reduction in extent of U(VI) adsorption to natural substrates was observed on treatment of these substrates with dithionite-citrate-bicarbonate (DCB) reagent, confirming that association of U(VI) with iron oxides is critical. DCB leaching of clays alone had very little effect on extent of U(VI) adsorption.

6.4 Radionuclide Transport

Studies of natural geochemical systems can contribute to an understanding of the processes controlling the migration of radionuclides. They complement other experimental studies, such as laboratory and field experiments, in that they give access to the long timescales that are relevant to safety assessments of repositories.

The Koongarra system offers the opportunity for relatively sophisticated modelling, directed at understanding the geochemistry of the system and the processes controlling the extent of uranium transport. The aims of the transport modelling were:

(a) to understand the processes that retard the migration of uranium series radionuclides;

(b) to obtain bounds on the timescale over which the dispersion fan has been formed and to compare these bounds with constraints emerging from geomorphological studies.

Modelling of the system is complicated by a number of uncertainties about the evolution of the system, and in particular, by uncertainties in groundwater flow patterns and flow rates over geological timescales. Climatic variations could have resulted in substantial lowering of the water table so that no horizontal uranium transport occurred. There is also uncertainty in the evolution of hydrogeological parameters with time as a result of the formation of clays, and in the rate at which the weathering front moved downwards.

A large number of models have been employed as part of the ARAP, to interpret the substantial database available for the concentrations of uranium series radionuclides in the groundwater and in the solid phases. Value has been seen in the diversity of modelling approaches. In all 15 modelling approaches are reported, some of which are very similar to those used in safety assessments. The age of the dispersion fan has been estimated to be in the range 0.5 to 3 million years, with a median in the range of 1 to 1.5 million years. These estimates are dependent on assumptions about the variation of the rate of uranium addition to the fan with time. These age estimates are in accord with best estimates of rates of surface erosion over the Koongarra orebody, which suggest initiation of the dispersion fan between one and six million years ago. The ARAP was accepted as one of the test cases within Phase I of the NEA International INTRAVAL Project (see insert).

The models have been used to investigate a number of effects and the values of different parameters. The timescale on which uranium equilibrates between the groundwater and the inaccessible phase has been constrained to be of the same order as the half life of ^{234}U. In this chemically reactive system, partitioning of uranium into a crystalline phase is found to be an important retarding process to uranium transport.

INTRAVAL

INTRAVAL is an international project coordinated by the NEA designed to establish the validity of models used to assess the long term safety of radioactive waste disposal systems.

ARAP has been included as an INTRAVAL Phase I Test Case based on data for migration of uranium in the weathered zone. A number of preliminary transport, hydrology and geochemical modelling reports were prepared, with Koongarra data bases also being used to test a Performance Assessment model. The possible application of the scenario development procedures to the Koongarra site was discussed.

6.5 Geochemistry of ^{239}Pu, ^{129}I, ^{99}Tc and ^{36}Cl

Measured abundances of natural ^{239}Pu, ^{99}Tc and ^{129}I in uranium ores from Koongarra (and other uranium deposits) have been compared to calculated abundances in order to evaluate the degree of retention of these radionuclides by the ore. Measured ^{129}I concentrations in all samples of unweathered primary ore investigated in this study exceeded the minimum concentrations predicted from spontaneous fission of ^{238}U. However, the concentrations were invariably less than the amounts predicted by the source-term model under a wide variety of assumptions. Although the production of ^{129}I by neutron-induced fission of ^{235}U and ^{238}U is difficult to estimate with any accuracy, the measurements suggested that ^{129}I may be depleted to a larger extent in samples from the shallow Koongarra orebody than in samples from the deeper Cigar Lake deposit. Samples from weathered primary ore from Koongarra contained quantities of ^{129}I that are well below the minimum allowable abundance for in-situ production, an unambiguous indicator of loss of this fission product from its site of production, possibly in response to the alteration processes that have occurred in this region of the deposit. The mobility of radioiodine was also apparent from its high concentrations in the groundwaters flowing through the Koongarra deposit.

Predictions of the source-term model indicated that ^{99}Tc production in uranium ores was dominated by spontaneous fission of ^{238}U. In the ore samples studied, production of this fission product by induced fission of ^{235}U was 5-25% of the total, and that by neutron-induced fission of ^{238}U was generally ~4 %. The dominance of the spontaneous fission component should simplify interpretation of measured ^{99}Tc concentrations in uranium minerals. The validity of the source-term model prediction was borne out by two analyses of ^{99}Tc in primary ores, which had ^{99}Tc abundances at the levels predicted by the model, about 10% above that expected for spontaneous fission alone. Release of ^{99}Tc from the ore during weathering was suggested by its presence at measurable concentrations in water from one of the Koongarra boreholes.

Source-term models predicting ^{239}Pu abundances in uranium minerals led to ambiguous conclusions. Several model-dependent evaluations of ^{239}Pu concentrations were compared with measured abundances in primary uranium minerals that were believed to have been closed systems with respect to ^{239}Pu and U. Measured abundances were generally within the range of predicted values. However, the results indicated that, in very heterogeneous media such as that typical of uranium ore deposits, the chemical parameters critical for determination of ^{239}Pu abundances were difficult to characterise with sufficient accuracy to use the results for evaluating the degree of retention or loss of ^{239}Pu from the orebody. The model predictions were

particularly sensitive to spatial variations of U, H and Gd concentrations within a radius of about 20-40 cm of the sample. Consequently, current methods for estimating ^{239}Pu production rates are limited to estimates that are no better than a factor of 2 of the true rate, which is not sufficiently precise to provide unambiguous measures of Pu retention.

Attempts to characterise the aqueous geochemistry of plutonium in Koongarra groundwater indicated the presence of the element in two wells at very low concentrations, on the order of 10^{-18} M. The source of the plutonium in one of the wells is unknown and may be anthropogenic fallout rather than natural plutonium derived from the orebody. The results suggested that mobile plutonium may be in particulate matter and in anionic form. Establishing the validity of these observations, and a less ambiguous interpretation, would require additional, more sophisticated, sampling.

The feasibility of using uranium minerals as analogues for the behaviour of nuclear reaction products (NRP) in spent fuel relies upon a capability to characterise NRP concentrations in source minerals. Extensive modelling work documented in this study emphasises the need to include as much information as possible about sample environs in any NRP production rate model. Even with detailed compositional information, however, uncertainties in model predictions will often be too large to provide reliable estimates of absolute NRP abundances in heterogeneous geologic systems. To address this limitation, one of the most useful conclusions of the modelling study has been to show the extent to which various NRP should be correlated to one another, such that one constrains the production rate of another. For example, ^{36}Cl ($t_{1/2}$, 3.0×10^5 a), another long-lived neutron-capture product found in uranium ores, was shown to be an ideal in-situ monitor of the ^{235}U fission rate, which is the dominant source term for ^{129}I and possibly a significant one for ^{99}Tc. Similarly, $^{239}Pu/U$ ratios can be used to establish limits on the ^{238}U neutron-induced fission rate; the ratios measured in this study suggest that ^{238}U induced fission comprises < 4 % of the total fissions in most of the ore samples studied. These two correlations are only valid if the H/U atom ratio in the sample or in its immediate environs exceeds 6, which roughly corresponds to 6000 ppm H.

7. SCENARIO DEVELOPMENT

The study of natural analogues is recognised as one of the more important methods for the validation of concepts and models for performance assessment of repositories for nuclear waste, and indeed has many features in common with performance assessment. However, if we are to transfer the results of an analogue to better understand repository performance in the long term, there is a need to ensure that the proper questions are being asked and the analogue investigations are focussed on phenomena relevant for validation objectives. In many respects the Koongarra study provided a full spectrum of challenges and opportunities as it was a multi-disciplinary project, ranging from small-scale processes to the evolution of the site and the dispersion of uranium from the primary orebody. Full advantage was made of this in a 'Scenarios Development' task, which had as its objective the application of experience from the development of scenarios for performance assessments to an evaluation of Koongarra.

A major difficulty in trying to model the evolution of a natural analogue, including dispersion of natural radionuclides, is to define the time frame and the initial and boundary conditions. Although information on the history of the site may be obtained from analysis of radioactive disequilibria, independent information is needed for validation of transport models and concepts. It is therefore important to test not one, but several possible alternatives of the site history, ie scenarios. The testing of several scenarios and models should therefore contribute to a better understanding of the ability and limitations of the different concepts and models in the description of processes relevant for performance assessments.

The methodology was similar to that of a joint SKI/SKB scenario development project and followed a technique initially used by Sandia. The initial stages were to identify, document and screen all features, events and processes (242 FEPs) that might have had an influence on the evolution of the Koongarra analogue. The 190 FEPs remaining after the initial screening were classified into 3 subgroups relating to the 'Process System' comprising all deterministic chemical and physical phenomena that might have influenced the formation of the secondary mineralisation and the dispersion in the weathered zone (138 FEPs), 'External Conditions' independent of the process system, such as the rainfall range and intensity (30 FEPs), and 'External Features', which reflect the history of the region but did not influence the evolution of the analogue (13 FEPs). These were then used to describe the analogue in a series of schematic diagrams, which displayed the interconnections and links of causes and effects between different processes. Three major scenarios results, following the rapid weathering of the host rock and eventual oxidation and mobilisation of the uranium ore under unsaturated or saturated conditions, viz uranyl phosphates forming under unsaturated conditions with a periodical evolution of the dispersion fan in conjunction with alternating dry (glacial) and wet periods, uranyl phosphates forming under unsaturated conditions as a single event, and uranyl phosphates forming under saturated conditions in conjunction with periods of higher and lower flow as a result of climatic cycling.

More work is needed to fully explore the approach, however, although the original objectives may have not yet been achieved the work has contributed to a better understanding of the analogue and has provided a basis for further scenario work.

8. CONCLUDING COMMENT

Within the framework of the ARAP, the Koongarra uranium deposit has been developed as an analogue of the behaviour of radioactive waste repositories in the far field on the assumption of post closure breaching by groundwater.

Uranium deposits, which are intersected by groundwater are particularly good analogues because they comprise a spectrum of uranium series nuclides, transuranic elements and fission products similar to many of the nuclear reaction products in spent fuel (albeit in very different concentrations). In addition, selected processes relevant to radionuclide retardation can be studied over time frames ranging from <1 year to millions of years, ie over time frames which span those relevant to the performance assessment of a repository proposal by orders of magnitude. Individual processes studied in the laboratory on the field can be set in the context of an evolving hydrogeological system through the natural analogue. Mathematical models describing the behaviour of systems over the past tens of thousands of years or longer are of a similar level of complexity as those required for long term prediction.

Because of the large amount of systematic work undertaken at Koongarra, a holistic approach to the natural analogue has been possible. A basis exists for outlining not only the interrelationship between the laboratory, field and natural analogue, but also the adaptation of the analogue methodology to the assessment of proposed repository sites. In the following, processes are listed in a broadly decreasing time frame.

1. Evolution of the structural geology of the area and the deposition of the primary ore (1870 to 1600 My).

2. Development of the weathered profile following rapid erosion of the schist overlying the orebody during the Pleistocene (up to 2 My), leading to significant changes of uranium and thorium retardation factors.

3. Evolution of the dispersion fan (the results of transport modelling suggests 0.5 to 3 My with a median in the range 1 to 1.5 My). During this period there were significant paleoclimatic changes with long periods [100 ky] of drier conditions, interrupted by shorter periods [10 ky] similar to those prevailing in the area today.

4. Formation and redistribution of transuranic elements (^{239}Pu) and fission products (^{129}I and ^{99}Tc). The timescales reflect those of the formation of the source uranium, the hydrogeochemistry and the half-life of the isotope (say 0.01 to 3 My).

5. Formation and distribution of uranium series nuclides within the minerals of the weathered host rock; uranium (^{234}U/^{238}U) (0.05-1 My), thorium-230 (0.01-0.3 My), and radium isotopes (and ^{228}Th).

6. Residence time distributions of groundwater which cannot be simply described by grouped parameters because of the non-uniformity of the host rock properties over the region of interest.

About 15 mathematical modelling approaches were developed to describe the above processes. The modelling of the evolution of secondary mineralisation (process 3) involves a level of complexity of a similar order as the modelling of the complexity of an overall repository system. An understanding of the scope and limitations of different concepts and models was obtained using a methodology similar to that in a joint SKI/SKB scenario development project.

Processes 1, 2 and 3 apply specifically to Koongarra, although the insights obtained on the time dependent retardation processes are widely applicable and the major data bases obtained during the work can be applied generally for model testing. Process 4 is specific to any analogue designed around a uranium deposit. Processes 5 and 6 can be studied in any setting and may be used to contribute to repository site evaluation.

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