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Advances in understanding the Kombolgie Subgroup and unconformity-related uranium deposits in the Alligator Rivers Uranium Field and how to explore for them using lithogeochemical principles

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The Alligator Rivers Uranium Field (ARUF) includes the mined and unmined Jabiluka, Ranger, Koongarra and Nabarlek unconformity-related uranium deposits and several small prospects including the newly discovered King River prospect. Uranium mineralisation is hosted by a variety of metamorphosed Nimbawah Domain lithologies that are unconformably overlain by the Kombolgie Subgroup, a basin package of unmetamorphosed arenites and mafic volcanics. All of the uranium deposits and prospects preserve an identical alteration assemblage that is subdivided into a distal and proximal alteration zone. The distal alteration zone comprises an assemblage of sericite and chlorite that replace albite and amphibole. In some cases, this alteration can be traced >1000 m from the proximal alteration zone that is dominated by uraninite, hematite, chlorite and sericite. Uranium precipitated in the basement as uraninite at 1680 Ma at around 200°C from a fluid having $\delta^{18}\text{O}_{\text{fluid}}$ values of $3.0 \pm 2.8\text{‰}$ and $\delta\text{D}_{\text{fluid}}$ values of $-28 \pm 13\text{‰}$ VSMOW reflecting an evolved marine source. These geochemical properties are indistinguishable from those recorded by diagenetic illite and chlorite that were collected from the Kombolgie Subgroup sandstones across the ARUF. The illite and chlorite formed in diagenetic aquifers, and where these aquifers intersected favourable basement rocks, such as those containing graphite or other reductants, U was precipitated as uraninite. Therefore, it is proposed that the Kombolgie Subgroup is the source for fluids that formed the deposits. A post-ore alteration assemblage dominated by chlorite, but also comprising quartz $\pm$ dolomite $\pm$ sulfide veins cut the uranium mineralisation at all deposits and has historically been recorded as part of the syn-ore mineralisation event. However, these minerals formed anywhere between 1500 to 630 Ma from fluids that have distinctly lower $\delta^{18}\text{O}_{\text{fluid}}$ values around 1.5‰ and lower $\delta\text{D}_{\text{fluid}}$ values around -45‰ reflecting a meteoric water origin. Despite unconformity-related uranium deposits having a large alteration halo, they remain difficult to find. The subtle alteration of albite to sericite several hundred metres from mineralisation occurs in isolation of any increase in trace elements such as U and radiogenic Pb and can be difficult or impossible to identify in hand specimen. Whole rock geochemical data indicate that Pearce Element Ratio (PER) analysis and General Element Ratio (GER) analysis may vector into this subtle alteration because it does not rely on an increase in trace elements to identify proximity to ore. PER and GER plots, Al/Ti vs (2Ca + Na + K)/Ti, Na/Al vs (Na + K)/Al, K/Al vs (Na + K)/Al and (Fe + Mg)/Al vs (Na + K)/Al provide a visual guide that readily distinguish unaltered from altered samples. A plot of (Na + K)/Al and (Fe + Mg)/Al on the x-axis against the concentration of trace elements on the y-axis reveals that U, Pb, Mo, Cu, B, Br, Ce, Y, Li, Ni, V and Nd are associated with the most intensely altered samples. The lithogeochemical vectors should aid explorers searching for uranium mineralisation in a prospective basin environment, but exploration must first focus on the characteristics of the basin to assess its mineralisation potential. A holistic model that describes the evolution of the Kombolgie Subgroup from deposition through diagenesis to formation of the uranium deposits in the underlying basement rocks is presented and has application to other basins that are considered prospective for unconformity-related uranium deposits. The model outlines that explorers will need to consider the thickness of the sedimentary pile, its lithological composition relative to depositional setting, the depth to which the sediments were buried during diagenesis and the degree of diagenesis achieved, which may be time dependant, before deciding on the prospectivity of the basin.

KEY WORDS: Uranium, unconformity, Jabiluka, Ranger, King River, Nabarlek, lithogeochemistry, Pearce Element Ratios, Kombolgie Subgroup, Australia.

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INTRODUCTION

The Alligator Rivers Uranium Field (ARUF) in the Northern Territory, Australia, hosts mined and unmined unconformity-related U deposits, including Jabiluka, Ranger, Koongarra, Nabarlek (mined out) and the lesser known King River prospect. Mineralisation is hosted in a range of lithologies belonging to the Nimbuwah Domain, including the Cahill Formation, the Kukalak Gneiss and the Nourlangie Schist. Most of the ARUF deposits are well described with the most recent papers focussing on petrology and geochemistry (Wilde & Wall 1987; Needham 1988; Wilde *et al.* 1989; Hancock *et al.* 1990; Wilde & Noakes 1990; Komninou & Sverjensky 1996; Polito *et al.* 2004, 2005). Whereas this information helps to better formulate deposit models and stimulate debate on the origin of the ore fluids, few of these papers have attempted to address how to geochemically explore for unconformity-related uranium mineralisation.

Exploration for unconformity-related uranium deposits in the ARUF is commonly hampered by the overlying Kombolgie Subgroup, although some prospective basement is known to crop out in the Kakadu National Park, which is off-limits to explorers. Despite the alteration halos around some of the uranium deposits visibly extending beyond 1000 m from mineralisation (e.g. Nabarlek), unconformity-related deposits are difficult to find. One reason for this is that path-finder elements associated with mineralisation are generally at background levels until well within the proximal alteration zone. For example, Ferguson & Winer (1980) found that U correlates poorly with other trace elements when its concentration in a sample is <40 ppm, but a good correlation with As, Nb, Mo, Pb, Sc, Co and other path-finder elements is achieved when the U concentration in a sample is in excess of 500 ppm. Data collected from the Jabiluka deposit for this study indicates that by the time a sample contains 40 ppm U, chlorite and sericite alteration is clearly visible and the need for a trace element vector to indicate alteration and mineralisation becomes more academic in nature.

Explorers for unconformity-related uranium deposits are regularly confronted with having to drill several hundred metres through the overlying sandstone packages before reaching the unconformity and underlying basement. The exercise is costly and therefore the explorer must know whether or not the drill hole has pierced the edge of a mineralised system before deciding to expand or abandon the exploration program. The early stages of distal alteration around an unconformity-related uranium deposit can be difficult or impossible to see in hand specimens, and the explorer may have to send samples to a laboratory for a petrological study to assess if any alteration exists. Pearce Element Ratios and General Element Ratios may be one way to maximise the exploration effort because whole rock geochemistry is commonly obtained by explorers during exploration and can be used to model petrology and alteration when the correct ratios are employed.

This paper is a culmination of data collected by the authors over eight years in the ARUF (Kyser *et al.* 2000; Hiatt & Kyser 2002; Polito *et al.* 2004, 2005, 2006a; Hiatt

et al. 2007; Kyser 2007). Previously published information is combined with never-before-published descriptions from the recently discovered King River prospect (Beckett 2008) and geochemistry from this and other deposits, including the Ranger uranium deposit, is presented. The data from several deposits, prospects and a wide area of Kombolgie Subgroup sandstone facies is then geologically modelled in a way that holistically looks at the ARUF as one system. A Pearce Element Ratio and General Element Ratio study complements the petrological observations made by the authors and may prove to be a useful tool for explorers searching for unconformity-related uranium deposits in this and other basins.

REGIONAL GEOLOGY

The ARUF lies within the Pine Creek Orogen, approximately 230 km east of Darwin (Figure 1). The Pine Creek Orogen forms the northern margin of the North Australian Craton (Plumb 1979) and comprises sequences of carbonaceous, clastic and volcanogenic sedimentary rocks deposited upon rifted Neoproterozoic crystalline basement, which were subsequently deformed, metamorphosed, and intruded by syn- to post-orogenic granitoids and mafic bodies (Neumann & Fraser 2007; Hollis *et al.* 2009a). The Pine Creek Orogen is divided into three domains, which from west to east are the Litchfield Province, the Central Domain, and the Nimbuwah Domain. The principal U deposits in the ARUF, Jabiluka, Ranger, Nabarlek and Koongarra are located within the Nimbuwah Domain.

The Nimbuwah Domain comprises the 2520±3 Ma Nanambu Complex (Hollis *et al.* 2009a), the Kakadu Group, the Cahill Formation and the Nourlangie Schist (Neumann & Fraser 2007). The 1870±6 Ma Zamu Dolerite intrudes the Cahill Formation and Nourlangie Schist placing a younger age on these formations. The Nimbuwah Domain underwent metamorphism up to granulite facies between ca 1867 and 1860 Ma, coincident with the emplacement of syn-orogenic granodioritic plutons belonging to the Nimbuwah Complex (Worden 2006; Worden *et al.* 2008a; Hollis *et al.* 2009b), reaching lower to middle amphibolite facies in the area near the Jabiluka deposit. Deformation associated with this event is constrained by the 1847±1 Ma age of a post-orogenic pegmatite at the Ranger uranium mine (Annesley *et al.* 2002) and the post-orogenic Nabarlek and Tin Camp Granites (1818±7 Ma and 1846±13 Ma respectively; Worden 2006; Worden *et al.* 2008b).

The Nimbuwah Domain is unconformably overlain by unmetamorphosed arenites and mafic volcanic rocks belonging to the Kombolgie Subgroup (Sweet *et al.* 1999a), which is the basal succession of the Proterozoic Katherine River Group (Needham 1988; Sweet *et al.* 1999a). The Katherine River Group has a maximum and minimum depositional age between 1822±5 Ma and 1720±7 Ma constrained by U–Pb ages on the underlying Plum Tree Creek Volcanics (Sweet *et al.* 1999a) and intrusion by the Jimbu Microgranite at 1720±7 Ma (Rawlings 1999) and the Oenpelli Dolerite at 1723±6 Ma (Edgecombe *et al.* 2002).

GEOLOGICAL OVERVIEW OF THE KOMBOLGIE SUBGROUP

The Kombolgie Subgroup is formally divided into six formations based on geological contacts, from oldest to youngest: the Mamadawere Sandstone, the Nungalarri Volcanics, the Gumarrirnbang Sandstone, the Gilruth Volcanics, the Marlgowa Sandstone and the McKay Sandstone (Figure 2). However, it is more accurately divided into six sequence stratigraphic

packages comprising alluvial fan to braided fluvial facies, proximal to distal braided fluvial facies, upper shoreface and eolian facies and distal braided fluvial to upper shoreface facies (Hiatt & Kyser 2002; Kyser 2007; Figure 2). The Kombolgie Subgroup records a complex fluid history related to diagenesis, mineralisation, and later tectonic events. The principal diagenetic events are recorded as diagenetic aquitards and diagenetic aquifers (Polito *et al.* 2005, 2006a, b).

Figure 1 Regional geology of the east Alligator Rivers uranium field showing the location of the deposits and prospects referred to in the area. Modified after Needham (1988). Also shown is the location of the N–S cross-section shown in Figure 2.

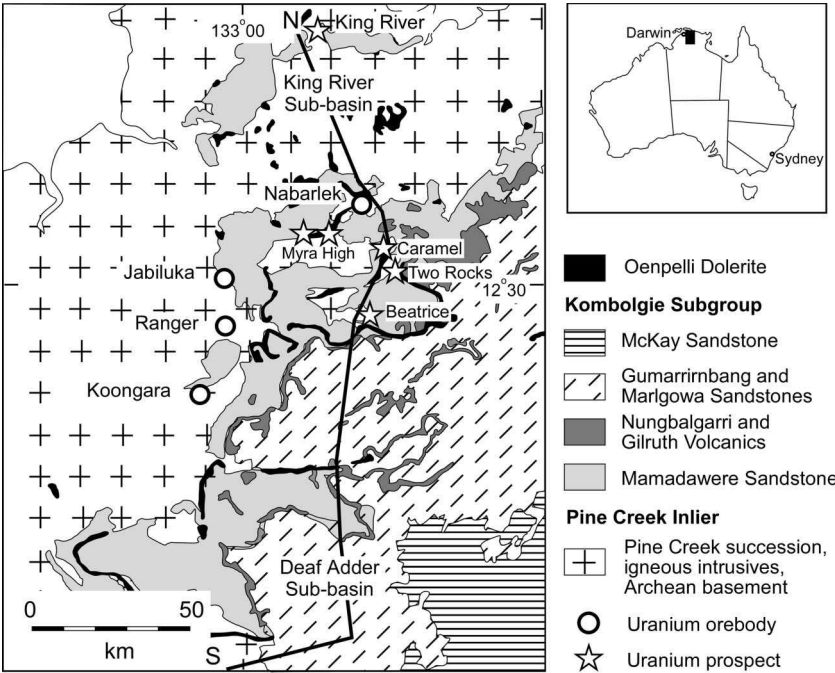
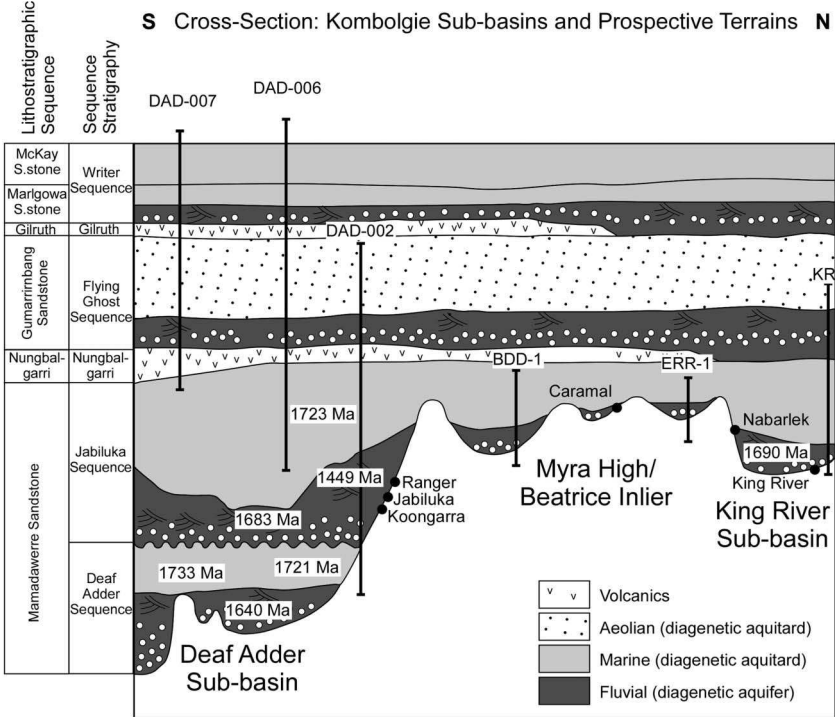


Figure 2 Cross-section of the Kombolgie Subgroup overlying undifferentiated basement rock showing the mapped sandstone and volcanic lithologies, the corresponding sequence stratigraphy identified from outcrop and various drill-core (Hiatt & Kyser 2002; Hiatt *et al.* 2007; Kyser 2007) and the location of some of the U deposits in the ARUF. Also shown are the ^{40}Ar – ^{39}Ar ages of diagenetic illite from the diagenetic aquifers and aquitards.



Diagenetic aquitards typically form in compositionally mature (i.e. 100% quartz) sandstones that lack detrital clay or feldspar (Kyser 2007). These sandstone facies were originally porous when deposited. However, quartz cement derived from paleoweathering, shallow (<1 km) dissolution of interbedded volcanic clasts and glass or compaction-driven, grain-to-grain pressure solution, precipitated as quartz overgrowths in these facies and cemented the detrital grains together (Polito *et al.* 2006b; Hiatt *et al.* 2007). The quartz cements played a major role in controlling the paleohydraulic properties of the Kombolgie Subgroup by occluding porosity and decreasing permeability. Trough cross-bedded quartzites, distal fluvial and coastal sandstones, and fine-grained sandstones are commonly well cemented by these quartz overgrowths. The quartz cements record $\delta^{18}\text{O}_{\text{fluid}}$ values of $0 \pm 4\%$ VSMOW, indicative of marine fluids and preserve fluid inclusions that homogenise below 130°C (Hiatt *et al.* 2007).

In contrast, diagenetic aquifers commonly form in the poorly sorted pebbly and conglomeratic sandstones of proximal fluvial facies (Hiatt & Kyser 2000; Kyser *et al.* 2000; Polito *et al.* 2006b). These facies were originally poor transmitters of fluid, but during deep burial, silicate solubility increased and led to framework dissolution of detrital feldspar, smectitic clays, quartz overgrowths and detrital quartz grains and the creation of secondary porosity (Hiatt & Kyser 2000; Polito *et al.* 2006a, b; Kyser 2007). Illite is the primary diagenetic phase that formed and filled the secondary pore spaces in the poorly sorted clastic facies of the Kombolgie Subgroup, but chlorite and kaolinite are locally common (Hiatt & Kyser 2000; Kyser *et al.* 2000). Illite crystallinity data indicate that the basinal brines in the Kombolgie Subgroup diagenetic aquifers had temperatures ranging between 150 and 230°C (Polito *et al.* 2006a, b; Kyser 2007), corresponding to burial depths of 4 to 8 km assuming a thermal gradient of 30 to 35°C (Pagel *et al.* 1980; Derome *et al.* 2003; Hiatt *et al.* 2007). The diagenetic illite and chlorite preserve $\delta^{18}\text{O}_{\text{fluid}}$ values of $4.5 \pm 2\%$ and $\delta\text{D}_{\text{fluid}}$ values of $-25 \pm 15\%$, indicative of basinal brine that evolved from a marine-dominated fluid with a minor meteoric component (Longstaffe 1987, 2000).

Contact metamorphism caused by the intrusion of the Oenpelli Dolerite into the Kombolgie Subgroup at ca 1723 Ma records the diagenetic state of the clastic sediments at that time in the basin's history. Where the Oenpelli Dolerite came in contact with fluid-bearing diagenetic aquifers, quartz dissolution of the surrounding sediments occurred, and doubly terminated quartz crystals precipitated. Conversely, where the Oenpelli Dolerite intruded the diagenetic aquitards, no physical change to the sandstones is observed (Kyser *et al.* 2000). The occurrence of pyrophyllite and muscovite near the Oenpelli Dolerite in Kombolgie Subgroup sediments in the King River Sub-basin (Figure 1, 2) indicates that diagenetic illite and kaolinite had already formed in some diagenetic aquifers as a result of fluid flow prior to intrusion by the Oenpelli Dolerite.

The compartmentalisation of the Kombolgie Subgroup by the diagenetic aquitards appears to have had a significant impact on the location of the unconformity-

related U deposits. Indeed, there appears to be a spatial relationship between the diagenetic aquifers and the location of the unconformity-related U deposits and showings that lie to the southwest of the Myra/Beatrice inlier (Figure 2). The U deposits all lie beneath the unconformity in basement rocks beneath diagenetic aquifers (Hiatt & Kyser 2002; Kyser 2007). These aquifers are capped by diagenetic aquitards, suggesting that flow of mineralising fluids within the basin was in confined diagenetic aquifers. Where these diagenetic aquifers intersected favourable basement rocks, such as those containing graphite or other reductants, U was precipitated as uraninite (Polito *et al.* 2004, 2005).

GEOLOGICAL OVERVIEW OF THE ARUF DEPOSITS

The geological details of the ARUF deposits are well described (Anthony 1975; Eupene *et al.* 1975; Foy & Pedersen 1975; Rowntree & Mosher 1975; Hegge 1977; Binns *et al.* 1980a, b; Ferguson 1980; Needham & Stuart-Smith 1980; Page *et al.* 1980; Snelling 1980, 1990; Ypma & Fuzikawa 1980; Gustafson & Curtis 1983; Ewers *et al.* 1984; Johnston 1984; Wilde & Wall 1987; Needham 1988; Wilde *et al.* 1989; Wilde & Noakes 1990; Hancock *et al.* 1990; Komninou & Sverjensky 1996; Polito *et al.* 2004, 2005; Hollis *et al.* 2009a). In this paper, only specific aspects of the Jabiluka, Ranger and Nabarlek deposits and the King River prospect are described and discussed. Reference is made to the Koongarra deposit, but no formal study of this deposit was carried out by the authors.

The Jabiluka, Ranger and Koongarra deposits are hosted by a dipping sequence of predominantly chlorite–biotite–muscovite–feldspar schists that form the lower member of the Cahill Formation. At Jabiluka and Ranger, the Cahill Formation has been divided into a series of mine sequences that are either carbonaceous/graphitic-bearing members that host the majority of the identified mineralisation or schist-dominated members that are non-carbonaceous quartz–muscovite–chlorite schists containing limited mineralisation (Eupene *et al.* 1975; Hegge 1977; Hancock *et al.* 1990).

The Nabarlek deposit was once thought to be hosted by the Myra Falls Metamorphics (Anthony 1975; Wilde & Wall 1987), but recent work by Hollis *et al.* (2009a, b) suggests that the deposit is probably hosted by the Nourlangie Schist (Hollis pers. com.). The Nourlangie Schist at Nabarlek comprises (1) the Lower Schist Unit, a ca 200 m-thick sequence of muscovite–quartz–biotite schist, interlayered with thin intervals of hornblende–plagioclase–biotite–clinopyroxene amphibolite; (2) the Footwall Amphibolite, a ca 100 m package of interlayered amphibolite and schist that hosts most of the ore; and (3) the Hanging-Wall schist, an alternating package of quartz-rich psammitic schists, muscovite-rich pelitic to semi-pelitic schists and thin quartzite and amphibolite layers that is the only other host to known mineralisation.

The lesser-known King River prospect in the Wellington Range, Cobourg Peninsula region is currently being evaluated by Cameco Corporation (Beckitt 2008). The project comprises three prospect areas called

Aurari North, Kuroikin and Laterite (Figure 3). These prospects are located approximately 250 km east of Darwin are situated within the Nimbuwah Domain and occur adjacent to the Aurari Fault. The basement geology comprises the prospective Nimbuwah Complex and the recently named Kukalak Gneiss (Hollis *et al.* 2009a). The Oenpelli Dolerite is prevalent at King River ranging from 1 to 300 m thick. It intrudes the basement lithologies and lies at the unconformity contact of the overlying the Kombolgie Subgroup sandstone (Beckitt 2008). The Aurari Fault is a major structure in the project area that trends 330°, is approximately 100 m wide, and marks the western edge of the outcropping Kombolgie Subgroup sandstone (Figure 3; Beckitt 2008). Mineralisation is hosted in (1) silicified microbrecciated gneisses that occur in the hanging wall of the Aurari Fault zone at the Aurari North prospect, (2) silicified microbrecciated gneisses and variably altered feldspar-biotite gneisses immediately below the Oenpelli Dolerite at the Laterite Area, and (3) foliated biotite granodiorite and tonalite in the footwall block of the northeast-dipping Aurari Fault zone at Kuroikin.

SYN-ORE PROCESSES IN THE LARGE BASEMENT-HOSTED URANIUM DEPOSITS

Alteration

Basement-hosted alteration is extensive at all of the large deposits. It can be traced up to 1000 m away from mineralisation at Nabarlek (Wilde & Wall, 1987) and is open in all directions around the Jabiluka deposit where

drilling exists up to 200 m beyond the known mineralisation. At the smaller King River prospect, alteration is restricted to a few tens of metres around veinlets and disseminations of uraninite indicating restricted fluid access. In all examples, the alteration assemblage varies only slightly in detail with the main differences between each deposit being due to variations in the host-rock assemblage. At the Jabiluka, Ranger, Koongarra and Nabarlek deposits, alteration associated with uranium mineralisation is loosely divided into an outer (distal) and inner (proximal) halo.

The petrology of the ARUF deposits has been described in detail (see references above), and only a brief review is presented here (Figure 4). Distal alteration is represented at each deposit by the partial to complete replacement of metamorphic biotite, garnet, amphibole, feldspar and sillimanite by chlorite and fine-grained sericite. Petrographic analysis demonstrates that sericite alteration of feldspar occurs first and furthest from the ore zone (Figure 5a), followed by sericite replacement of amphibole and sillimanite and then by increasing amounts of chlorite toward mineralisation (Figure 5b; Polito *et al.* 2004, 2005). Alexandre *et al.* (2005) demonstrated that the formation of crystalline illite/sericite from amphibole and feldspar around the U deposits in the Athabasca Basin created void space due to the minerals' contrasting molar volume. These authors proposed that void space creation promoted additional fluid flow into the basement, with the release of Mg and Fe from minerals such as amphibole, resulting in the precipitation of void-consuming chlorite. This hypothesis adequately explains the observations at all of the deposits studied in the ARUF. At the boundary between the distal and proximal zones, muscovite, relatively unaltered in the distal zone, is replaced by sericite (Figure 5c).

Within the proximal mineralised zone, alteration becomes more intense and complex. Mineralised and unmineralised breccias are common in the inner alteration zone and typically comprise strongly chloritised and sericitised host-rock cemented by uraninite, hematite, sericite/illite, chlorite $\pm$ tourmaline $\pm$ quartz (Kubler crystallinity indices calculated using X-ray diffraction data from phyllosilicate-rich samples in the ARUF deposits indicate that sericite and illite ($\pm$ phengite) may coexist in the proximal alteration zone). This assemblage is often cross-cut by other generations of uraninite, chlorite, sericite/illite and hematite. Porosity creation and fluid flow in the proximal alteration zone is primarily controlled by faulting, shearing and brecciation, which contrasts with the passive porosity creation in the distal alteration zone. At Jabiluka and King River, uraninite is observed with euhedral quartz $\pm$ hematite veins that are surrounded by an illite/phengite/sericite and hematite selvage (Figure 5d). Where mineralisation is most intense the alteration assemblage is typically represented by uraninite, brick red to purple hematite, pale yellow to white sericite or phengite and only minor pale green or white chlorite (Polito *et al.* 2005). This intense hematite-phengite alteration is best developed at Nabarlek where the hematite can comprise up to 20% of the alteration assemblage (Figure 5e); however, it is also observed at

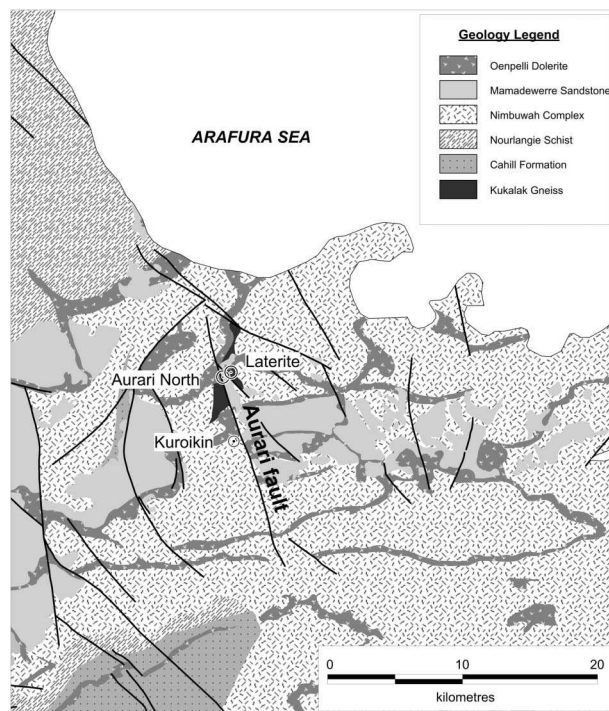


Figure 3 King River prospect comprising the three prospects, Aurari North, Kuroikin and Laterite occurring adjacent to the Aurari Fault.

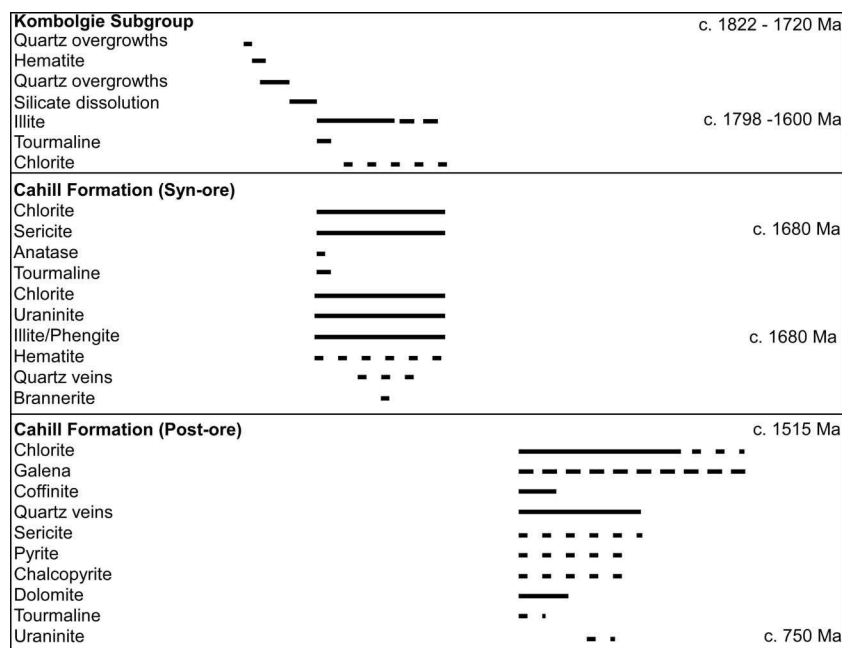


Figure 4 Paragenetic diagram summarising the relative and absolute timing of alteration assemblages in the sandstone and basement lithologies in the ARUF. The ages presented for each event are based on cross-cutting relationships that have been constrained by K/Ar ages (Page *et al.* 1980) and $^{40}\text{Ar}/^{39}\text{Ar}$ and $^{207}\text{Pb}/^{206}\text{Pb}$ ages (Kyser *et al.* 2000; Polito *et al.* 2004, 2005; this study).

Jabiluka where hematite–phengite alteration is restricted to uraninite veins that cut earlier generations of uraninite, chlorite, sericite/illite and hematite (Polito *et al.* 2005). Graphite at Jabiluka and Ranger is obliterated in strongly bleached zones adjacent to massive uraninite veins but is generally preserved, particularly where mineralisation comprises chlorite, sericite and disseminated uraninite (Polito *et al.* 2005).

Fluid characteristics

Temperature estimates, calculated using the methods of Cathelineau & Nieva (1985) and Cathelineau (1988) on electron microprobe data from chlorite phases from Jabiluka, Nabarlek and King River, indicate that porosity-filling, syn-ore chlorite in the outer and inner alteration zones formed at $209 \pm 41^\circ\text{C}$ (Figure 6; Polito *et al.* 2004, 2005). This is in agreement with a temperature estimate of $200 \pm 50^\circ\text{C}$ (Figure 6) calculated from the syn-ore sericite/illite and phengite separates from the same deposits using crystallinity data generated from X-ray diffraction (XRD) patterns (Kubler 1967; Arkai 1991). Illite that formed during burial diagenesis in the Kombolgie Subgroup formed at $180 \pm 50^\circ\text{C}$ (Polito *et al.* 2005).

Fluid inclusions in a syn-ore uraninite-quartz vein from the Jabiluka deposit do not reveal a formation temperature because none of the fluid inclusions analysed homogenised prior to decrepitating, making it impossible to calculate trapping temperatures (Polito *et al.* 2005). Nevertheless, these fluid inclusions have eutectic temperatures of $-35 \pm 5.1^\circ\text{C}$, revealing that the syn-ore fluid was a mixture of CaCl_2 , MgCl and NaCl . The final ice melting temperature in these inclusions ranges between -17.1 and -8.2°C indicating that the syn-ore fluid had moderate to high salinities.

The isotopic characteristics of fluids that transported U to the site of deposition are recorded in the various alteration minerals at each deposit. Syn-ore chlorite and

sericite/illite in the basement preserve $\delta^{18}\text{O}$ values and δD values that indicate formation from fluids having $\delta^{18}\text{O}_{\text{fluid}}$ values of $3.0 \pm 2.8\text{‰}$ and $\delta\text{D}_{\text{fluid}}$ values of $-28 \pm 13\text{‰}$ VSMOW (Figure 7; Table 1; Polito *et al.* 2004, 2005). This assumes their formation at $\sim 200^\circ\text{C}$. Syn-ore chlorite and illite extracted from the alteration halo in the Kombolgie Subgroup sandstones immediately above the deposits formed in equilibrium with a fluid having $\delta^{18}\text{O}_{\text{fluid}}$ values of $4.2 \pm 3.4\text{‰}$ and $\delta\text{D}_{\text{fluid}}$ values of $-28 \pm 18\text{‰}$ VSMOW; indistinguishable from those obtained from the basement-hosted chlorite and illite (Figure 7). The $\delta^{18}\text{O}_{\text{fluid}}$ values are too depleted in ^{18}O to be derived from a metamorphic fluid, whereas the D-enriched $\delta\text{D}_{\text{fluid}}$ values rule out normal magmatic and pure meteoric sources (Taylor 1997). Collectively, this rules out basement-derived fluids for the origin of the uranium. Instead, the alteration minerals precipitated from basinal brines that evolved in sandstone aquifers in the Kombolgie Subgroup from typically marine pore fluids with minor contributions from meteoric sources (Longstaffe 1987, 2000; Kyser *et al.* 2000; Polito *et al.* 2005, 2006b).

MINERALISATION AND ALTERATION AT THE KING RIVER PROSPECT

Mineralisation at the King River prospect is characterised by an alteration assemblage that is similar to that observed at the other ARUF deposits. At Aurari North, mineralisation is preceded by pre-ore pervasive silicification associated with chlorite and biotite. Syn-ore alteration comprises disseminated (1–10 μm) and worm-like uraninite, coffinite and sericite (Figures 5f, 8a). Sericite at all of the King River prospects forms the most common alteration mineral proximal to peak radiometric intervals and can be fine-grained, pervasive and pale yellow to brown, or fine- to medium-grained, reddish brown and form anastomosing veinlets

(Figure 5c, d, f, g). Red hematite is developed with the sericite and may be both fracture- or matrix-controlled (Figure 5d, g). A post-ore silicification event characterised by milky white chalcedonic quartz breccia veins and grey-to-white stockwork veinlets is locally developed and may be related to brittle movement along the Aurari Fault Zone. At Kuroikin, mineralisation is characterised by intense chlorite and sericite developed symmetrically around discrete drusy quartz-uraninite veins (Figure 5g). Sericite is more widely developed forming a broader halo around mineralised veins. Several other vein types, seemingly barren of mineralisation, occur at Kuroikin and include yellowish-grey sheeted carbonate veinlets, amorphous quartz-sericite $\pm$ chlorite $\pm$ carbonate veins and pink feldspar-quartz veins. It is uncertain if these veins represent an early barren event or a more distal vein set associated with the mineralised veins. Alteration at Laterite Area exhibits styles that occur at both Aurari North and Kuroikin (e.g. Figure 5d). Early silicification is overprinted by stockwork sericite alteration and chlorite. This alteration is subsequently overprinted by discrete ribboned quartz-sericite $\pm$ chlorite $\pm$ carbonate and drusy quartz-uraninite veins similar to that observed at Kuroikin. Alteration around mineralisation at all three prospects in the King River area is limited in extent and rarely extends more than 10 m beyond visible uraninite (Figure 5g).

POST-ORE PROCESSES AFFECTING THE LARGE BASEMENT-HOSTED URANIUM DEPOSITS

Alteration

Post-ore alteration is common at all of the major deposits and manifests itself as breccias and fractures that are cemented by drusy quartz and chlorite $\pm$ sericite $\pm$ sulfide $\pm$ dolomite. The chlorite ranges from white and pale green to dark green and black, and forms radial fans, massive laths or veins that cut the syn-ore assemblage of uraninite, illite, hematite and chlorite (Polito *et al.* 2004, 2005). In some areas of each of the major deposits, post-ore chlorite is the only alteration mineral present with remnant uraninite. This gives the impression that chlorite is the main, if not the only, alteration mineral associated with U mineralisation. At the Ranger No. 3 deposit, almost all of the samples investigated during this study were dominated by post-ore chlorite and heterogeneous or corroded uraninite (Figure 8b, c).

Mineralisation at all of the major deposits is also cross-cut by several generations of clear to milky-white quartz $\pm$ dolomite veins that are found in the basement and the overlying sandstone (Polito *et al.* 2004, 2005). The quartz veins range from monomineralic comb-textured quartz to composite veins comprising zoned quartz $\pm$ dolomite $\pm$ pyrite $\pm$ chalcopyrite. The cross-cutting nature of these veins clearly indicates that they are late relative to the uranium mineralisation event.

Sulfides are present in all of the deposits and are found disseminated in uraninite, along partings between alteration minerals and encrusted on quartz in

veins. Pyrite, chalcopyrite and galena are the most common sulfides present, but chalcocite, cobaltite, bornite, arsenopyrite and pentlandite have been observed at most deposits, prospects and showings (Figures 5h, 8d). The galena likely formed from radiogenic Pb derived from decaying U. Its cross-cutting nature with the uraninite, plus its coexistence with pyrite and chalcopyrite in the late quartz veins suggests that the sulfides are part of the post-ore assemblage.

Fluid characteristics

Temperature estimates calculated using electron microprobe data from post-ore chlorite from Jabiluka, Nabarlek and Ranger indicate two formation temperatures at $286 \pm 41^\circ\text{C}$ and $200 \pm 15^\circ\text{C}$ (Figure 6; Polito *et al.* 2004, 2005). The former temperature is much higher than temperature estimates from syn-ore chlorite, but support for this temperature range is found in fluid inclusions in the post-ore quartz veins.

Fluid inclusions in post-ore quartz veins contain both aqueous-vapour fluid inclusions and aqueous-vapour fluid inclusions containing one or more daughter minerals (Derome *et al.* 2003; Polito *et al.* 2004, 2005). Aqueous-dominated fluid inclusions have eutectic temperatures that range from -82.0 to -21.4°C and final melt temperatures from -50.3 to 0.0°C (Polito *et al.* 2004, 2005). The lowest of these temperatures indicate that some post-ore fluids were extremely saline and dominated by a Ca^{2+} -rich fluid. Other fluid inclusions contain daughter minerals that achieve homogenisation by halite disappearance between 106 and 168°C indicating the presence of a highly saline Na^+ -dominated fluid. Derome *et al.* (2003) report that drusy quartz veins that host fluid inclusions, with identical chemical and thermal properties to those described here, contain fluids dominated by Ca^{2+} and Na^+ with Ca:Na ratios ranging widely from 0.07 to 10.5 . This range of chemical and thermal characteristics can be attributed to mixing between a Na-dominated and a Ca-dominated fluid as suggested by Derome *et al.* (2003), but according to the paragenesis (Figure 4), this mixing occurred after uranium precipitation had ended and is not the cause for uraninite precipitation.

Fluid inclusion homogenisation temperatures range from 91.8 to 325.1°C . Derome *et al.* (2003) modelled pressure corrections for various basin scenarios during the Mesoproterozoic for the ARUF and demonstrated that most fluid inclusions were likely trapped at a minimum temperature of $150 \pm 20^\circ\text{C}$ assuming a geothermal gradient of $30 \pm 5^\circ\text{C}$ (Figure 6). Other fluid inclusions homogenise at a much higher temperature around $280 \pm 50^\circ\text{C}$ (Figure 6). These fluid inclusions have low salinities, and their homogenisation temperatures, uncorrected for pressure, coincide with post-ore chlorite temperature estimates around $286 \pm 41^\circ\text{C}$ indicating that these high temperatures are not unique to a specific mineral phase; nor can they be considered data outliers.

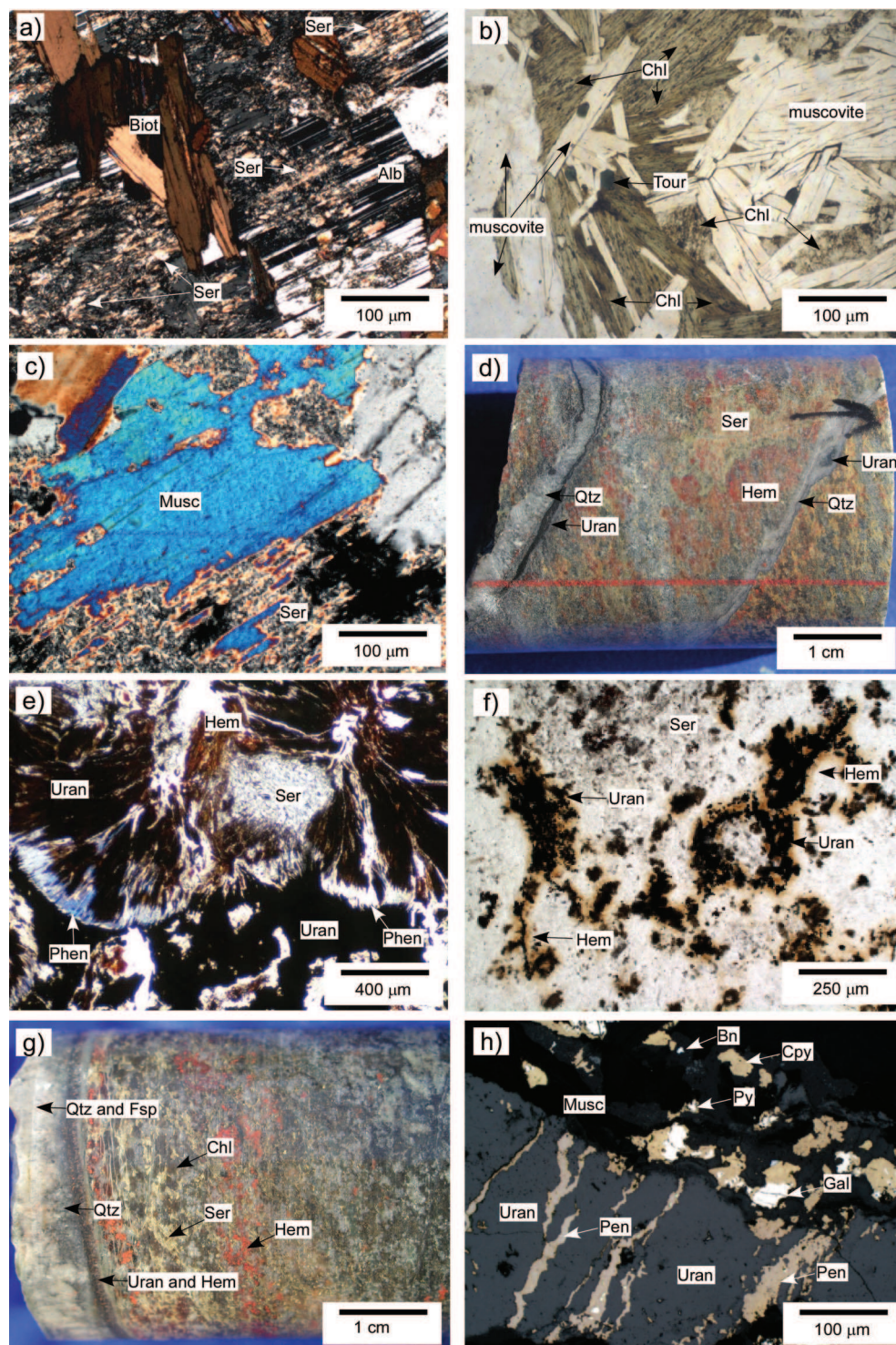
Post-ore chlorite and drusy quartz at Nabarlek, Jabiluka and Ranger preserve $\delta^{18}\text{O}$ and δD values that are distinct from those recorded by the syn-ore minerals. Post-ore chlorite indicate formation from fluids having $\delta^{18}\text{O}_{\text{fluid}}$ values of $2.1 \pm 1.5\text{‰}$ and $\delta\text{D}_{\text{fluid}}$ values of

$-45 \pm 14\text{‰}$ (Figure 7; Table 1; Polito *et al.* 2004, 2005) indistinguishable from post-ore drusy quartz which formed in equilibrium with a fluid having $\delta^{18}\text{O}_{\text{fluid}}$ values of $1.3 \pm 3.0\text{‰}$ and $\delta\text{D}_{\text{fluid}}$ values of $-47 \pm 15\text{‰}$. These values indicate formation from a variably evolved meteoric fluid with a relatively minor contribution from a marine source (Longstaffe 1987, 2000). Drusy quartz veins collected from other localities in the ARUF, including Deaf Adder and the Caramal prospects reported by Derome *et al.* (2003), also record $\delta^{18}\text{O}_{\text{fluid}}$

values of $2.5 \pm 2.1\text{‰}$ and $\delta\text{D}_{\text{fluid}}$ values of $-47 \pm 13\text{‰}$ (Kyser *et al.* 2000) demonstrating that the majority of drusy quartz veins in the ARUF appear to have formed from a fluid that was unrelated to the syn-ore fluid.

AGE DETERMINATIONS AND IMPLICATIONS

Numerous efforts have been made to determine the ages of the ARUF deposits (Hills & Richards 1976; Gulson &



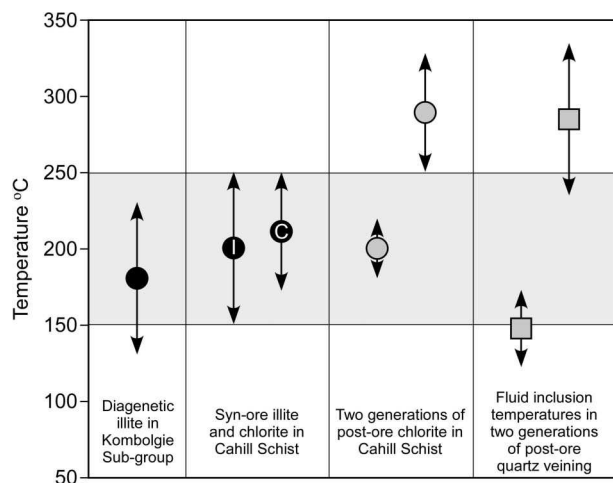


Figure 6 Temperature ranges calculated for various minerals associated with diagenesis and mineralisation in the ARUF. Temperatures were obtained from electron microprobe, X-ray diffraction and fluid inclusion homogenisation data. This diagram incorporates previously published data (Kyser *et al.* 2000; Polito *et al.* 2004, 2005).

Mizon 1980; Maas *et al.* 1986; Ludwig *et al.* 1987; Maas 1989; Polito *et al.* 2004, 2005), with the oldest age of 1737 ± 20 Ma coming from the Ranger deposit (Ludwig *et al.* 1987). However, structural and paragenetic investigations by Hein (2002) now indicate that uranium-bearing ore shoots at the Ranger No 1 and 3 pits formed after emplacement of the 1723 Ma Oenpelli Dolerite, subsequently questioning the accuracy of the 1737 ± 20 Ma mineralisation age.

Backscattered electron images show that there are multiple generations of uraninite preserved at each deposit and prospect in the ARUF (Figure 8; Polito *et al.* 2004, 2005). This is reflected by electron microprobe data and LA-HR-ICPMS analyses that give widely variable ages from Jabiluka, Nabarlek, Ranger and King River uraninite (Figures 9, 10). Electron microprobe data show that ARUF uraninite has a variable chemical

composition, comprising <90.4 wt% UO_2 , and up to 20.2 wt% PbO, 3.7 wt% CaO, 0.9 wt% FeO and 3.7 wt% SiO_2 (Figure 9, Table 2; Polito *et al.* 2004, 2005). The U:Pb ratios determined from their respective oxide proportions correlate to chemical ages that range from 1691 to 227 Ma (Figure 9, Table 2; Bowles 1990). The oldest U–Pb chemical ages from King River come from the least altered uraninite. These ages are indistinguishable from the oldest $^{207}\text{Pb}/^{206}\text{Pb}$ ages around 1680 ± 10 Ma determined by LA-HR-ICPMS analysis (Figure 10, Table 3). $^{207}\text{Pb}/^{206}\text{Pb}$ ages are generally more accurate than U–Pb ages because preservation of Pb ratios during overprinting is possible, whereas U:Pb ratios are reset every time any amount of Pb is removed from the uraninite system. The oldest $^{207}\text{Pb}/^{206}\text{Pb}$ ages of 1685, 1684, 1679 and 1673 Ma obtained from the ARUF (Table 3) are therefore interpreted to represent the approximate age (i.e. 1680 ± 10 Ma) for the formation of the ARUF deposits (Polito *et al.* 2005; Kyser 2007).

The King River uraninite has chemical compositions that sit along an alteration trend that is distinct from that at other ARUF deposits (Figure 9). The old U–Pb chemical ages and distinct chemical composition for uraninite from King River support petrological evidence that a relatively small remobilisation event, relative to the rest of the ARUF, affected this project area. These data are in contrast to every other ARUF U deposit where U–Pb ages up to a maximum 992 Ma indicate that fluid overprinting efficiently removed much of the Pb from the uraninite up to 1000 Ma and then less efficiently after that. LA-HR-ICPMS data show that this Pb removal likely occurred during one of several events in the ARUF that is recorded by $^{207}\text{Pb}/^{206}\text{Pb}$ ages that form prominent peaks at 1513, 1448, 1360, 1257, 1176, 1001, 855 and 634 Ma (Figure 10).

Crystalline illite and sericite samples extracted from the alteration assemblage within the deposits and sandstone from the Kombolgie Subgroup were analysed using the $^{40}\text{Ar}/^{39}\text{Ar}$ method of Lee *et al.* (1990). These samples often give disturbed age spectra that are bell-shaped or indicate strong superficial $^{40}\text{Ar}^*$ loss or ^{39}Ar recoil. At Nabarlek, the problem of $^{40}\text{Ar}^*$ loss and ^{39}Ar

Figure 5 (a) Plagioclase (Alb) being replaced by sericite (ser) in the presence of pristine biotite (biot) at Kuroikin, King River prospect. Sample DDH KRD775 258.0 m, cross-polarised light. This sample appears unaltered in hand specimen despite the presence of a 1 cm wide uraninite–quartz–hematite vein 12 m away at 246.0 m. (b) Plain transmitted light photomicrograph showing chlorite (chl) pseudomorphing metamorphic biotite in the distal alteration zone, with apparently unaltered muscovite adjacent at Jabiluka. Note the pore filling chlorite, which has a different chemical composition to the chlorite replacing the biotite (Figure 15). Sample N207V 546.7 m. Tour, tourmaline. (c) Sericite (ser) replacing muscovite (musc) close to mineralisation at Aurari North, King River prospect. Sample DDH 664 151.1 m, cross-polarised light. (d) Uraninite with euhedral quartz $\pm$ hematite veins surrounded by an illite/phengite/sericite and hematite selvage replacing feldspar–biotite gneisses at the Laterite Area, King River prospect. Sample DDH KRD0770 314.0 m. Uran, uraninite; Hem, hematite; ser, sericite; qtz, quartz. (e) Finely disseminated hematite within large, radial fans of clear to pale-blue illite indicating growth into open space at Nabarlek. Note the finely disseminated hematite within illite and the filamentous terminations of the illite interfingering with uraninite. Sample, Na4 40.5 m, plain transmitted light. Uran, uraninite; Hem, hematite; ser, sericite; phen, phengite. (f) Finely disseminated (1–10 μm) uraninite, red-yellow hematite and fine-grained, pervasive sericite at Aurari North, King River prospect. Sample DDH 664 151.1 m, plain transmitted light. Uran, uraninite; Hem, hematite; ser, sericite. (g) Intense red hematite, chlorite and sericite alteration developed around a quartz-feldspar-uraninite-hematite vein at Kuroikin, King River prospect. Note the narrow zone of alteration and the fresh biotite granodiorite approximately 5 cm from the mineralised vein; a characteristic that is unique at King River. Sample DDH KRD0776 314.0 m. Qtz, quartz; Fsp, Feldspar; Uran, uraninite; Hem, hematite; chl, chlorite; ser, sericite. (h) A uraninite vein (Uran) from the Two Rocks prospect cross-cut by sulfides including pyrite (Py), chalcopyrite (Cpy), galena (Gal), bornite (Bn) and pentlandite (Pen). Sample DDH MRD101 72.6 m.

recoil was prevalent and only integrated ages, which are considered equivalent to K–Ar ages could be reported for most samples (Polito *et al.* 2004). Similarly disturbed spectra are recorded from other locations throughout the ARUF, and it is rare that syn-ore illite and sericite samples preserve plateau and pseudo-plateau spectra. Illite extracted from Kombolgie Subgroup sandstones near Jabiluka (Polito *et al.* 2005) and King River

(Figure 11) has plateau and pseudo-plateau spectra that reveal old ages such as 1798 ± 13 Ma, through to 1688 ± 7 , 1683 ± 11 and 1677 ± 40 Ma. The oldest of these ages is interpreted to reflect the termination of diagenetic processes in a diagenetic aquitard within the Kombolgie Formation (Polito *et al.* 2005), whereas the *ca* 1680 Ma ages from illite in diagenetic aquifers are in agreement with the oldest $^{207}\text{Pb}/^{206}\text{Pb}$ and U–Pb ages proposed to be the timing of ore deposition.

The disturbed Ar isotopic systems recorded in most samples prevent an accurate interpretation of the data using the traditional age step method (McDougall & Harrison 1999). To make use of the step ages obtained, a Kernel density analysis (cf. Alexandre *et al.* 2009) was employed which, in relation to the $^{40}\text{Ar}/^{39}\text{Ar}$ method, assumes that mineral samples preserve discrete isotopic reservoirs with a distinct argon composition that broadly reflects the age of the fluid or thermal event that formed or reset the minerals. The Kernel technique is based on the accumulation of the individual steps over the age span of interest; in this case from approximately 1800 to 200 Ma. It assumes Gaussian distribution of the individual probabilities with each peak in the cumulative probability diagram corresponding to a discrete age (Alexandre *et al.* 2009). Application of the Kernel density analysis to the ARUF data reveals discrete peaks at *ca* 1760 ± 25 , 1729 ± 15 , 1688 ± 20 , 1629 ± 25 , 1470 ± 15 , 1382 ± 15 and 1264 ± 15 Ma (Figure 10). Each of these ages corresponds to a northern Australian basin or a magmatic event that in many cases is also recorded by uraninite in the $^{207}\text{Pb}/^{206}\text{Pb}$ system.

Figure 10 compares ages obtained from illite and sericite by the $^{40}\text{Ar}/^{39}\text{Ar}$ method of Lee *et al.* (1990) and $^{207}\text{Pb}/^{206}\text{Pb}$ ages from uraninite by the LA-ICP-MS method of Kyser *et al.* (2003). A small narrow peak at 1788 ± 10 Ma is only recorded by illite in diagenetic aquitards belonging to the Kombolgie Subgroup. This age likely represents the timing of quartz cementation of the distal fluvial, coastal and fine-grained sandstones in the basin. The 1760 ± 25 Ma age is predominantly recorded by diagenetic illite in the Kombolgie Subgroup and some sericite in the basement rocks. This age overlaps several events in northern Australia including the extrusion of the 1780–1760 Ma Nungbagarri, Seigal and Eastern Creek Volcanics as well as the 1755–1730 Ma

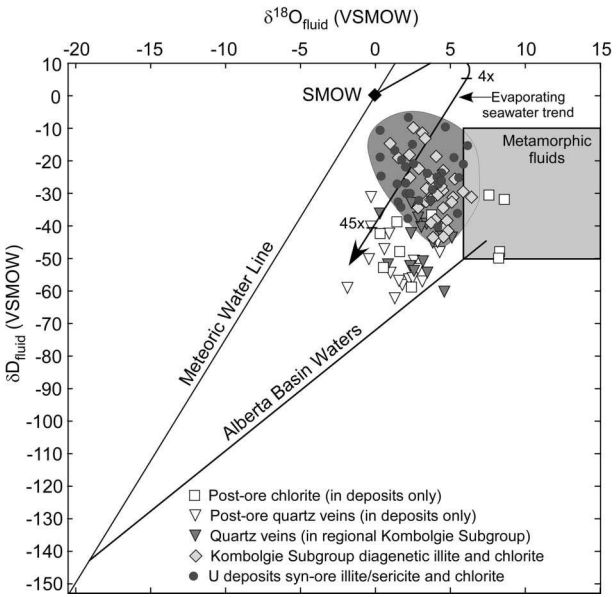


Figure 7 Calculated $\delta^{18}\text{O}$ and δD values for various fluids associated with alteration in the ARUF. The shaded area represents the calculated isotopic value of the fluid that formed both the regional Kombolgie Subgroup diagenetic minerals (illite and chlorite) and the syn-ore alteration minerals (sericite and chlorite). The compositions and calculations for each mineral are presented in Table 1 or previously published (Kyser *et al.* 2000; Polito *et al.* 2004, 2005). The regional quartz veins that cross the Kombolgie Subgroup were collected from the Caramel, Black Rock, Deaf Adder, Tin Camp and West Beatrice prospects (Kyser *et al.* 2000). The evaporating seawater trend and Alberta Basin water trend have been added to demonstrate the different origins for the syn-ore and post-ore fluids. SMOW, Standard Mean Ocean Water; MWL, Meteoric Water Line.

Table 1 Oxygen and hydrogen stable isotope values (‰ VSMOW) of quartz, chlorite and sericite from the Ranger deposit and King River prospect.

DDH	Paragenesis	$\delta^{18}\text{O}_{\text{min.}}$	$\delta\text{D}_{\text{min.}}$	Temperature ($^{\circ}\text{C}$)	$\delta^{18}\text{O}_{\text{fluid}}$	$\delta\text{D}_{\text{fluid}}$
Kuroikin KRD775 257.8m	Syn-ore sericite	7.6	–51	200 $^{\circ}\text{C}$	1.5	–34
Aurari Nth DDH664 151.1m	Syn-ore sericite	10.2	–54	200 $^{\circ}\text{C}$	4.1	–37
Ranger 3 pit 99R3-1	Post-ore quartz vein	14.7	–42	170 $^{\circ}\text{C}$	1.0	–42
Ranger 3 pit 99R3-3	Post-ore quartz vein	13.5	–31	170 $^{\circ}\text{C}$	–0.2	–31
Ranger S1 216 76.4	Post-ore chlorite alteration	5.6	–82	170 $^{\circ}\text{C}$	0.7	–53
Ranger S1 20 32	Post-ore chlorite alteration	6.4	–68	170 $^{\circ}\text{C}$	1.5	–39
Ranger S1 218 69.4	Post-ore chlorite alteration	6.6	–77	170 $^{\circ}\text{C}$	1.7	–48
Ranger S1 218 70.4	Post-ore chlorite alteration	5.4	–72	170 $^{\circ}\text{C}$	0.5	–43
Ranger S3 115 91.6	Post-ore chlorite alteration	7.4	–88	170 $^{\circ}\text{C}$	2.5	–59

Stable isotope values of the fluid from which these minerals precipitated are calculated using temperatures estimated in this paper (see text for details). Abbreviations: $\delta^{18}\text{O}_{\text{min.}}$ = $\delta^{18}\text{O}$ value of the mineral analysed; $\delta\text{D}_{\text{min.}}$ = δD value of mineral analysed. Figure 4 includes data presented here and in Polito *et al.* (2004, 2005).

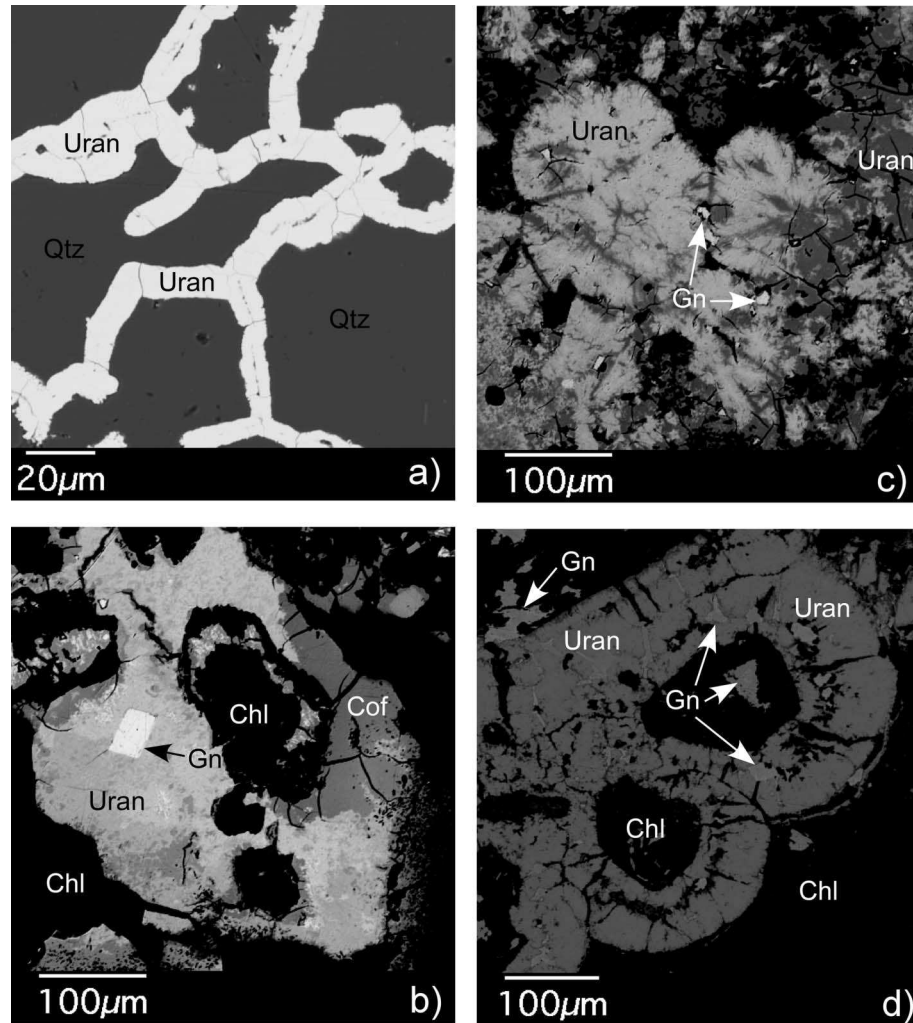


Figure 8 (a) Backscatter image of worm-like homogeneous uraninite in quartz at Aurari North; DDH 664 155.1 m. (b) Backscattered electron image of heterogeneous uraninite (light grey) displaying variable alteration to younger uraninite (various shades of intermediate grey), galena cubes and coffinite in a post-ore chlorite matrix. Sample provided by Northern Territory Geological Survey Ranger No. 1 pit. (c) Backscattered electron image of heterogeneous botroidal uraninite altering to galena cubes and a second generation of uraninite (dark grey). Sample provided by Northern Territory Geological Survey Ranger deposit; DDH 85-3 S1 219.0 m. (d) Backscatter image of uraninite botroids cross-cut by galena blebs and surrounded by post-ore chlorite. Two Rock prospect; DDH MRD101 72.6 m.

mid-Tawallah compression event that is recorded in the southern McArthur Basin (Bull & Rogers 1996). The 1729 ± 15 Ma age is recorded by both diagenetic illite and sericite within the Nimbuwah Domain and is coincident with the intrusion of the 1732 ± 11 Ma Jim Jim Granite and the 1723 ± 6 Ma Oenpelli Dolerite. A peak in Ar ages at this time confirms that these events affected both basement and basin lithologies in the ARUF. Distinct peaks at 1688 ± 17 Ma ($^{40}\text{Ar}/^{39}\text{Ar}$ from illite and sericite) and 1674 ± 15 Ma ($^{207}\text{Pb}/^{206}\text{Pb}$ from uraninite) record the timing of U mineralisation in the deposits and fluid flow out of diagenetic aquifers (Polito *et al.* 2005; Kyser 2007). These ages are coincident with a period of uplift and erosion in the southern McArthur Basin represented by bend 2 on the Australian Apparent Polar Wander Path (Idnurm 2000), and significant volcanic activity in the Mt Isa Basin (Page 1983; Southgate *et al.* 2000). $^{40}\text{Ar}/^{39}\text{Ar}$ step ages and $^{207}\text{Pb}/^{206}\text{Pb}$

uraninite ages around 1620 Ma (Figure 10) coincide with two Sm–Nd whole rock ages for U mineralisation in the ARUF (Maas 1989) suggesting that a subtle basin event occurred at this time, moving some Pb out of uraninite and partially resetting several different isotope systems. Other, younger ages around 1515, 1460, 1370, 1260, 1165, 1000 and 850 Ma (Figure 10) broadly correspond to well-documented tectonic or magmatic events. Some of these younger ages are within error, and are therefore potentially related to (1) renewed subsidence and volcanism at 1493 ± 4 Ma in the Roper Group of the McArthur Basin (Jackson *et al.* 1999; Abbott & Sweet 2000), (2) the intrusion of phonolitic dykes between 1370 and 1316 Ma (Page *et al.* 1980; Needham 1988) and the intrusion of the Derim Derim Dolerite at 1324 ± 4 Ma (Sweet *et al.* 1999b), (3) the earliest stages of amalgamation of Australia and Laurentia during the Grenville Orogeny at *ca* 1140 Ma (Wingate *et al.* 2002) and (4) the

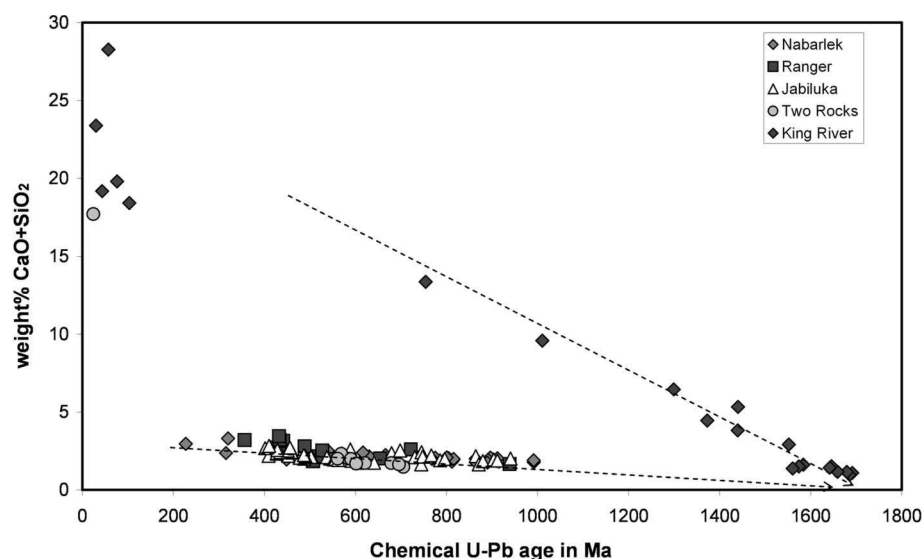


Figure 9 Relationship between U/Pb chemical ages and $\text{SiO}_2 + \text{CaO}$ contents in uraninite. Increasing concentrations of $\text{SiO}_2 + \text{CaO}$ in uraninite correlate with decreasing ages indicative of post-formation alteration by retrograde fluids. Note that the King River data gives older U–Pb ages and alters to higher $\text{SiO}_2 + \text{CaO}$ concentrations. This diagram combines previously published data (Polito *et al.*, 2004, 2005) with data in Table 2.

breakup of Rodinia between 1000 Ma (Tack *et al.* 2001) and 750 Ma (Wingate & Giddings 2000).

PEARCE ELEMENT RATIO/GENERAL ELEMENT RATIO ANALYSIS AND EXPLORATION IMPLICATIONS

Whole-rock geochemical interpretation on samples from the ARUF has previously been carried out by Ferguson & Winer (1980) and Binns *et al.* (1980b). The application of cluster analysis to 143 Cahill Formation samples (Ferguson & Winer 1980) found that U correlates with As, Nb, La, Ce, Mo, Pb and Co at a 99% confidence level when it is in excess of 500 ppm. In samples where U is in excess of 40 ppm, As, Nb, Mo, Pb, Sc, Co (and Cu and W at Jabiluka) show a good correlation. In other samples where U is <40 ppm, correlation with other elements is poor. The conclusion by Ferguson & Winer (1980) was that U, As, Nb, La, Ce, Mo, Pb, Co, W and Sc were transported in the mineralising fluids. Binns *et al.* (1980a) took whole rock data from the ARUF to the next level and sought to determine a geochemical signature that could be used in exploration for concealed U deposits. These authors combined petrological knowledge with geochemical data to determine which element enrichments and losses best represent alteration and mineralisation. The conclusions by Binns *et al.* (1980b) are sound, but significantly complicated by division of the Cahill Schist into 11 subgroups, the major ones being: (1) Pelitic Schist, (2) Psammitic Schist, (3) Graphite Schist, (4) Chlorite Schist, (5) Graphitic-Chlorite Schist and (6) Greisen.

In this paper, Pearce Element Ratios and General Element Ratios (PER/GER) were employed to: (1) determine if the large, but texturally subtle distal alteration halos could be identified using whole-rock

geochemical data, and (2) establish/confirm the trace-element geochemical signature of the uranium deposits.

PER/GER analysis allows comparisons between unaltered and altered samples by dealing with the addition or loss of material without being affected by errors created by closure. Closure arises in geochemical data when the sum total of all components is scaled to equal 100%. Summing all components to 100% effectively masks compositional variation when the size of the rock changes during material transfer. For example, the addition of one element during alteration associated with brittle deformation will lower the concentration of other elements in the rock, even though there has been no material loss. Closure can be avoided by converting the data to molar element ratios (otherwise termed PER) by using an element that did not participate in the material transfer (i.e. a conserved element) as the denominator element. In this way, the relative material transfers that took place to change the rock composition can be understood (Pearce 1968; Russell & Stanley 1990; Stanley & Madeisky 1994). The limitation of PER/GER analysis is that only one lithology can be interrogated at a time. In this paper, only samples that represent the Cahill Schist were reviewed. This implies that all regional Cahill Schist samples and only Jabiluka, Ranger and Koongarra uranium deposit samples were incorporated into the study. Nabarlek and King River are not considered further, despite both deposits having a petrographically similar, if not indistinguishable alteration assemblage, to that identified at the other deposits in the ARUF.

Whole-rock geochemical data from the Cahill Formation in the ARUF were obtained from open file and confidential company reports and Geoscience Australia's OZROCK database (<<http://www.ga.gov.au/gda/>>). They were lithogeochemically assessed using PER/GER analysis (Pearce 1968; Stanley & Russell 1989; Russell &

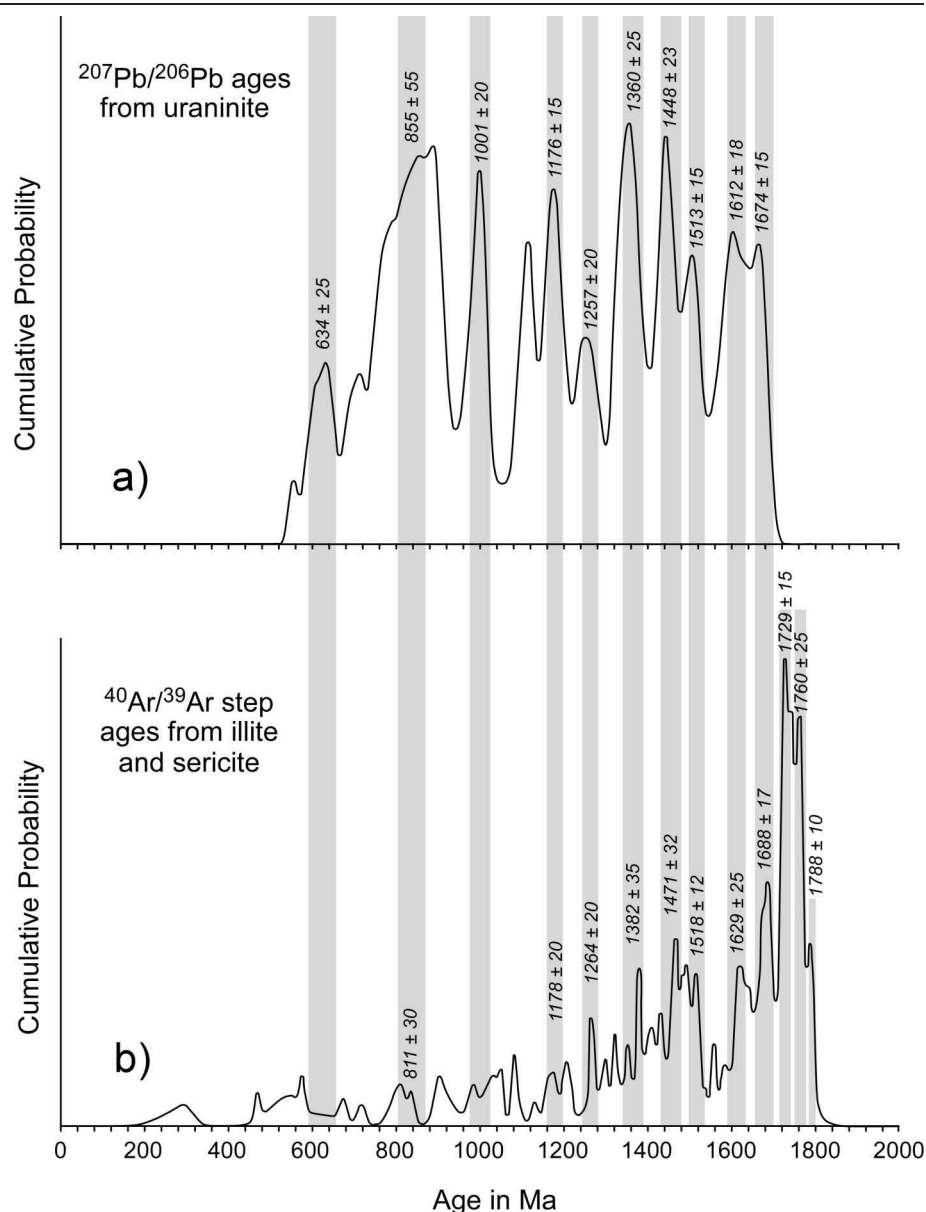


Figure 10 Cumulative probability diagrams for all $^{40}\text{Ar}/^{39}\text{Ar}$ analyses calculated using the individual step ages and error margins and $^{207}\text{Pb}/^{206}\text{Pb}$ ages and error margins from the ARUF (Table 3; Kyser *et al.* 2000; Polito *et al.* 2004, 2005), as indicated. The grey bands show the major perturbation events that affected the two different isotopic systems. The ages of the individual peaks are indicated.

Stanley 1990; Pearce & Stanley 1991; Stanley & Madeisky 1994). Prior to interrogation, the data were put through a rigorous QA/QC assessment to ensure that they were geochemically robust. Samples that had insufficient geographic locations, limited or no trace-element data, whole-rock total values that did not add up to $100 \pm 2\%$ and major element concentrations that suggested that the rock type was not representative of the Cahill Schist (i.e. samples of massive carbonate, massive chert, amphibolite or dolerite) were removed from the database.

Pearce Element Ratios are formed by dividing the molar concentration of a major element by that of a conserved element (typically a high field strength, immobile element) that has been unaffected by material transfer (Stanley 1993). Binary plots between Ti (as TiO_2), P (as P_2O_5), Nb, Zr, Y, Ce, La, and Al (as Al_2O_3) reveals that Ti is the most conserved element in the data set (Figure 12), displaying no evidence for addition or loss during metamorphism or alteration.

PER and GER diagrams were constructed with the aim to demonstrate alteration of fresh schist through to mineralised schist. The diagrams focussed on representing the alteration associated with the formation of illite and chlorite. Whereas PER diagrams display overall trends, GER diagrams allow multiple minerals of different compositions to be plotted as nodes on a single diagram. In GER diagrams, samples 'trend' towards or away from mineral nodes depending on the abundance of that mineral in the sample. Four diagrams accurately describe the process of alteration around unconformity-type deposits in the ARUF (Figures 13, 14). They are:

1. PER diagram of Al/Ti vs $(2\text{Ca} + \text{Na} + \text{K})/\text{Ti}$;
2. GER diagram of Na/Al vs $(\text{Na} + \text{K})/\text{Al}$;
3. GER diagram of K/Al vs $(\text{Na} + \text{K})/\text{Al}$; and
4. GER diagram of $(\text{Fe} + \text{Mg})/\text{Al}$ vs $(\text{Na} + \text{K})/\text{Al}$.

The Al/Ti vs $(2\text{Ca} + \text{Na} + \text{K})/\text{Ti}$ plot (Figure 13a) models feldspar destruction in the Cahill Schist to

Table 2 Chemical composition and calculated chemical U–Pb ages from electron microprobe analyses of variably altered uraninite at King River and Two Rocks.

Sample	SiO ₂ wt. %	PbO wt. %	UO ₂ wt. %	FeO wt. %	CaO wt. %	TiO ₂ wt. %	Total wt. %	Chemical age (Ma) ^a
King River 664 155.1 m	0.37	18.70	73.27	0.11	1.25	0.03	93.73	1584
King River 664 155.1 m	0.30	18.54	73.07	0.19	1.20	0.02	93.32	1574
King River 664 155.1 m	0.25	18.44	73.33	0.09	1.12	0.03	93.26	1560
King River 664 155.1 m	0.46	19.36	72.98	0.07	1.06	0.00	93.93	1645
King River 664 155.1 m	0.21	19.97	73.51	0.06	0.76	0.00	94.51	1685
King River 664 155.1 m	0.23	20.15	73.89	0.05	0.86	0.00	95.18	1691
King River 664 155.1 m	0.30	20.01	73.91	0.06	0.86	0.05	95.19	1679
King River 664 155.1 m	0.31	19.82	74.10	0.09	0.85	0.04	95.20	1659
King River 664 155.1 m	0.33	19.59	74.04	0.31	1.09	0.02	95.37	1641
King River 664 155.1 m	20.91	0.30	61.65	0.00	2.47	0.02	85.35	30
King River 664 155.1 m	16.22	0.46	66.29	0.09	2.96	0.87	86.90	43
King River 664 155.1 m	3.31	15.93	71.99	0.24	1.14	0.12	92.72	1373
King River KRD770 313.2 m	5.13	7.46	78.34	0.04	3.40	0.06	94.42	590
King River KRD770 313.2 m	4.74	7.14	78.40	0.00	3.54	0.08	93.89	565
King River KRD770 313.2 m	5.19	6.67	78.77	0.01	3.75	0.08	94.47	526
King River KRD770 313.2 m	4.62	7.37	78.88	0.00	4.02	0.05	94.93	579
King River KRD770 313.2 m	8.56	8.73	71.81	0.05	4.79	0.13	91.98	754
King River KRD770 313.2 m	15.76	0.84	68.27	0.11	4.04	0.10	89.12	77
King River KRD770 313.2 m	4.59	11.90	73.08	0.21	4.98	0.14	93.93	1010
King River KRD770 313.2 m	23.95	0.55	59.28	0.01	4.31	0.40	88.52	57
King River KRD770 313.2 m	1.80	17.99	71.90	0.04	1.12	0.19	93.03	1552
King River KRD770 313.2 m	3.69	17.15	73.84	0.02	1.64	0.14	96.54	1440
King River KRD770 313.2 m	1.98	17.53	75.53	0.06	1.83	0.16	97.08	1440
King River KRD770 313.2 m	2.71	15.68	74.89	0.03	3.74	0.12	97.34	1299
King River KRD770 313.2 m	14.34	1.08	64.80	0.07	4.07	0.08	84.46	103
Two Rocks MRD101 72.6 m	0.34	7.62	80.79	0.31	1.66	0.35	91.07	585
Two Rocks MRD101 72.6 m	16.38	0.23	57.31	0.06	1.33	0.04	75.33	24
Two Rocks MRD101 72.6 m	0.89	7.44	81.09	0.57	1.42	0.35	91.75	569
Two Rocks MRD101 72.6 m	0.43	7.17	75.34	0.47	1.57	0.26	85.22	590
Two Rocks MRD101 72.6 m	0.29	8.19	74.89	0.40	1.43	0.25	85.45	679
Two Rocks MRD101 72.6 m	0.25	7.77	68.37	0.23	1.23	0.20	78.05	705
Two Rocks MRD101 72.6 m	0.30	7.62	67.93	0.42	1.35	0.23	77.83	696
Two Rocks MRD101 72.6 m	0.34	6.85	68.38	0.38	1.38	0.23	77.54	621
Two Rocks MRD101 72.6 m	0.32	8.06	83.08	0.16	1.37	0.26	93.25	602
Two Rocks MRD101 72.6 m	0.32	7.56	83.61	0.44	1.68	0.28	93.90	561

^aChemical age calculated using the equation: age (Ma) = Pb × 104/(1.612U) (Bowles 1990).

muscovite/sericite, $m = 0.333$, then illite, $m = 0.273$ and finally chlorite $m = 0$. Most samples collected from within or immediately around the deposits plot below the muscovite/sericite control line demonstrating that they are easily identified from unaltered samples that contain pristine feldspar. Samples that plot above muscovite/sericite control line and below the feldspar control line, $m = 1$, contain both feldspar and muscovite, which approximately represents unaltered Cahill Schist. At the other end of the spectrum, totally altered, pure chlorite-bearing samples plot along $m = 0$ because they do not contain any Na or K. However, most chlorite-dominated samples plot above $m = 0$ due to the co-occurrence of chlorite with illite and sericite. It is noteworthy that any alteration mineral that does not contain Na or K, for example kaolinite, could also plot along $m = 0$, but petrological observations show that few unweathered samples contain kaolinite, whereas all altered samples contain chlorite. Samples that plot above the feldspar control line in Figure 13, $m = 1$, indicate the presence of other minerals that contain Na, Ca or K, such as amphibole or carbonate.

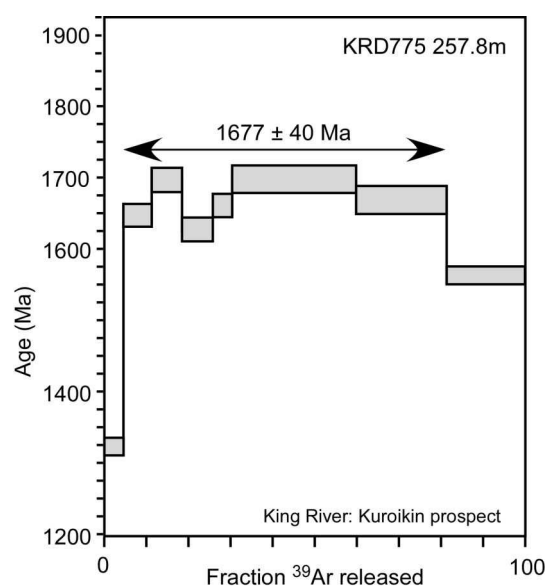
The GER diagram of K/Al *vs* (Na + K)/Al (Figure 13b) is used to demonstrate mineralogical control relative to Na and K in the system. The diagram plots K-feldspar, muscovite and illite, along a slope of 1 and by the absence of both Na and K, chlorite at the origin. This diagram shows that Na is removed from the Cahill Schist during alteration to illite. The horizontal trajectory of the Na-bearing samples toward the muscovite and illite nodes instead of a downward curve toward the chlorite node support the petrological findings that illite replaces all Na-bearing phases such as albite before chlorite begins to appear.

The GER diagram of Na/Al *vs* (Na + K)/Al (Figure 13c) models the alteration of specific feldspars, in this case albite and K-feldspar, to sericite and then chlorite. The figure supports thin-section observations and confirms the hypothesis that albite is the dominant Na-bearing phase being geochemically modelled. Figures 13b and c demonstrate that Na is lost from the system early and eventually K is totally removed, presumably in samples that are chlorite-dominated. This conclusion indicates that geochemical modelling

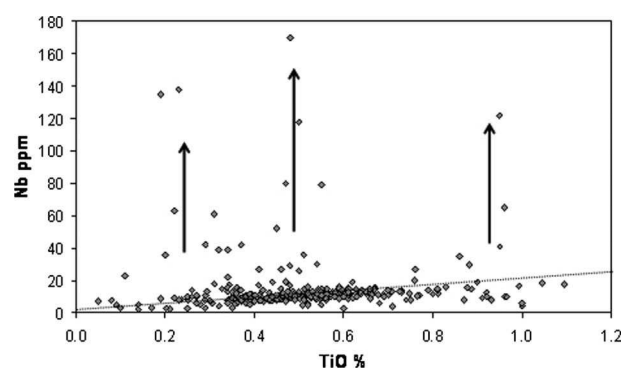
Table 3. LA-HR-ICPMS acquired U–Pb and isotopic data from uraninite samples from King River with calculated $^{235}\text{U}/^{207}\text{Pb}$, $^{238}\text{U}/^{206}\text{Pb}$ and $^{207}\text{Pb}/^{206}\text{Pb}$ ages.

Sample no.	Mineral	$^{207}\text{Pb}/^{235}\text{U}$	Isotopic ratios		$^{207}\text{Pb}/^{235}\text{U}$	Calculated ages (Ma ^a)	
			$^{206}\text{Pb}/^{238}\text{U}$	$^{207}\text{Pb}/^{206}\text{Pb}$		$^{206}\text{Pb}/^{238}\text{U}$	$^{207}\text{Pb}/^{206}\text{Pb}$
KRD770 313.2 m	Uraninite	1.710224	0.121179	0.102206	1012	737	1665
KRD770 313.2 m	Uraninite	0.973521	0.076390	0.092706	690	475	1482
KRD770 313.2 m	Uraninite	0.838520	0.060748	0.100579	618	380	1635
KRD770 313.2 m	Uraninite	1.037500	0.074540	0.101265	723	463	1647
KRD770 313.2 m	Uraninite	0.739702	0.054490	0.098136	562	342	1589
KRD770 313.2 m	Uraninite	0.913156	0.067205	0.102756	659	419	1673
KRD664 155.1 m	Uraninite	1.280599	0.097940	0.094388	837	602	1516
KRD664 155.1 m	Uraninite	0.276802	0.028571	0.071138	248	182	961
KRD664 155.1 m	Uraninite	1.136620	0.091155	0.091038	771	562	1447
KRD664 155.1 m	Uraninite	1.044441	0.078422	0.094146	726	487	1511
KRD664 155.1 m	Uraninite	0.221888	0.020337	0.072078	204	130	988
KRD664 155.1 m	Uraninite	1.046956	0.078498	0.094334	727	487	1515
KRD664 155.1 m	Uraninite	0.937146	0.065747	0.098052	671	411	1587
KRD666 226.9 m	Uraninite	0.167727	0.017362	0.089815	157	111	1422
KRD666 226.9 m	Uraninite	0.111250	0.013992	0.098733	107	90	1600
KRD666 226.9 m	Uraninite	0.177835	0.016936	0.103250	166	108	1684
KRD664 194.7 m	Uraninite	0.941698	0.069548	0.099298	673	433	1611
KRD664 194.7 m	Uraninite	0.782114	0.058401	0.098136	587	366	1589

^a $^{206}\text{Pb}/^{238}\text{U}$, $^{207}\text{Pb}/^{235}\text{U}$ and $^{207}\text{Pb}/^{206}\text{Pb}$ ages calculated using equations reported by Ludwig (1993).

**Figure 11** $^{40}\text{Ar}/^{39}\text{Ar}$ age spectra for sericite from Kuroikin at King River prospect. Sample extracted from DDH KRD775 257.8 m.

can monitor the destruction of albite $\pm$ K-feldspar in the Cahill Schist and can therefore be used as a vector toward mineralisation in the ARUF. The potential of this geochemical tool is enhanced with Figure 13c being presented as a bubble plot where the bubble size is proportional to U concentration in the sample. In this context, the bubble plot shows that unaltered through to weakly altered samples do not contain significant concentrations of U, whereas altered samples that plot between the muscovite/sericite node and the chlorite node contain highly anomalous concentrations of U.

**Figure 12** Scatterplot of TiO_2 vs Nb showing that Ti is the least mobile and therefore the most conserved element during hydrothermal alteration. Note that despite the wide concentration range for TiO_2 , Nb was gained or redistributed during mineralisation as indicated by the arrows.

The final GER diagram of $(\text{Fe} + \text{Mg})/\text{Al}$ vs $(\text{Na} + \text{K})/\text{Al}$ builds on the previous PER and GER diagrams and details the formation of chlorite from illite and muscovite (Figure 14). This diagram, presented as a bubble plot, supports observations that U precipitation was coincident with illite and chlorite formation. It also indicates that chlorite chemistry is variable. The mineralogical details of chlorite portrayed in this model are supported by electron microprobe data gathered from Jabiluka, Nabarlek and King River (Figure 15; Polito *et al.* 2004, 2005). However, the effect of post-ore overprinting by chlorite on the deposits cannot be determined using lithogeochemistry. Further, Figure 14 is not able to demonstrate that advanced alteration in the ARUF deposits results in the formation of illite, phengite and hematite from pre-existing chlorite (Polito *et al.* 2004, 2005). In both cases, these samples will plot

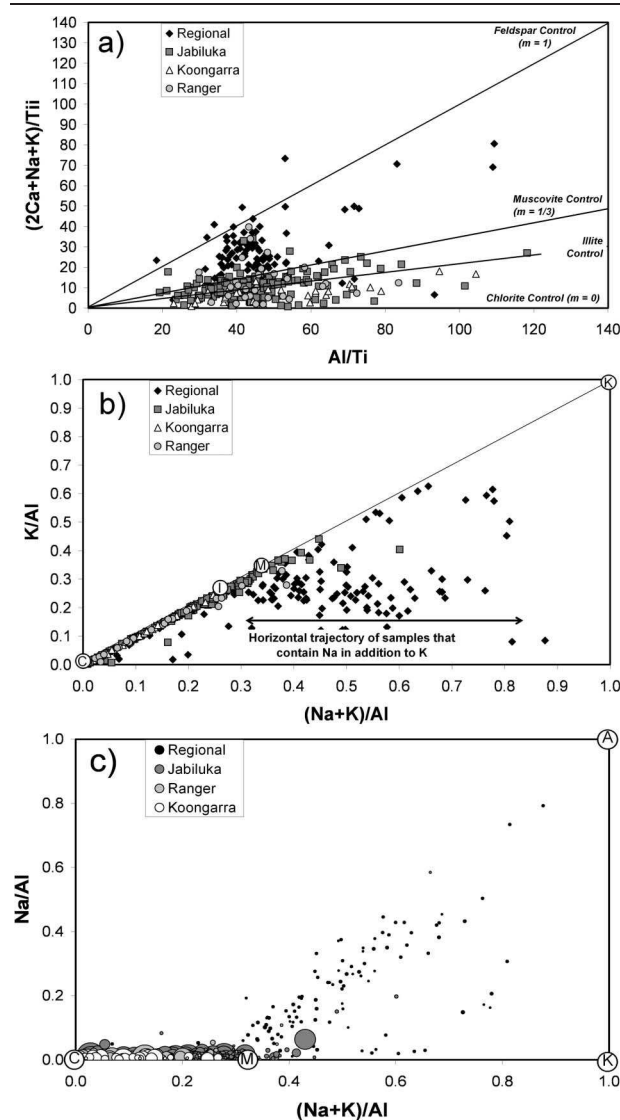


Figure 13 Pearce Element Ratio diagrams (a) Al/Ti vs $(2\text{Ca} + \text{Na} + \text{K})/\text{Ti}$ modelling feldspar destruction to muscovite (sericite), then illite and finally chlorite. Note the regional (unaltered samples plotting between the feldspar and muscovite control lines indicating no, minimal or trace alteration to sericite. Samples that plot above the feldspar control line contain excess Ca, Na or K, which is most likely due to carbonate. (b) K/Al vs $(\text{Na} + \text{K})/\text{Al}$ modelling the destruction K-feldspar (K), muscovite (M) and illite (I) along a slope of 1 to chlorite (C) at the origin. The presence of Na-feldspar is shown by the samples plotting to the right of the control line. Note the horizontal trajectory of the Na-bearing samples toward the muscovite and illite nodes instead of a downward curve toward the chlorite node. (c) Bubble plot of Na/Al vs $(\text{Na} + \text{K})/\text{Al}$ modelling the alteration of albite (A) and K-feldspar (K) to sericite (M) and then chlorite (C). Bubbles are proportional to U concentrations. The figure confirms that albite is the dominant Na-bearing phase being geochemically modelled. Note that K is totally removed in several samples, presumably in samples that are chlorite-dominated.

somewhere between the chlorite and illite nodes. Nevertheless, the lithogeochemical modelling of the ARUF alteration assemblage broadly portrays the paragenetic

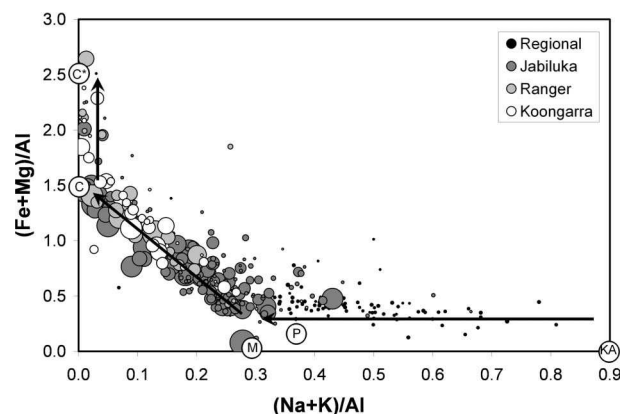


Figure 14 General Element Ratio diagram of $(\text{Fe} + \text{Mg})/\text{Al}$ vs $(\text{Na} + \text{K})/\text{Al}$ presented as a bubble plot detailing the formation of chlorite (C) from illite (I) and muscovite (M) which are alteration products of albite and K-feldspar (KA). Note that the that chlorite chemistry is variable and that the bubbles do not increase in size toward the chlorite nodes indicating that U precipitation was coincident with illite and chlorite formation. It is also noteworthy that several samples displaying significant sericite and chlorite alteration do not contain appreciable U which down-grades the application of simple multi-element path-finder analysis. This diagram is not able to demonstrate that advanced alteration in the ARUF deposits results in the formation of phengite (P) from pre-existing chlorite.

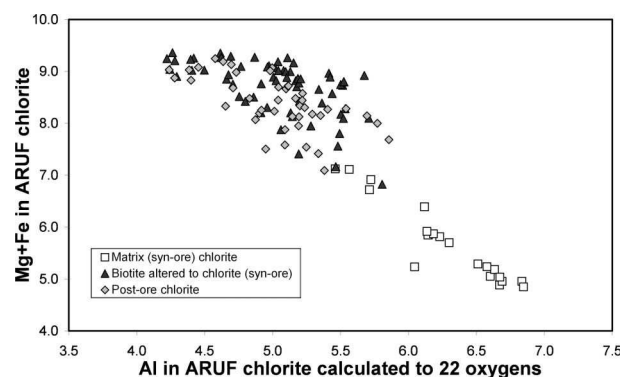


Figure 15 Scatterplot diagram showing the chemical variation of different chlorite types around the Jabiluka and Nabarlek deposits and the King River prospect. Elements expressed in units per weight formula calculated to 22 oxygens. This diagram includes some previously published data (Polito *et al.* 2004, 2005).

studies by Polito *et al.* (2004, 2005) and therefore has purpose as an alteration index based on the GER plots described here.

The alteration index, $(\text{Na} + \text{K})/\text{Al}$, is based on the molar proportions of these elements as represented in Figure 13b and c, and best represents alteration within and around the ARUF deposits. Significantly, this index outlines >20 altered samples that were previously identified as being background samples on the basis of their U concentrations (Figure 13a–c). In an exploration situation, these samples would require follow-up. In this case, the samples are in the Kakadu National Park. Alteration index $(\text{Na} + \text{K})/(\text{Fe} + \text{Mg})$, which is based on

the molar proportions of elements in Figure 14, enhances the anomalies in Figure 13 and helps to focus on the samples that may be closer to mineralisation. This index is best used with path finder elements to indicate proximity to ore (Figure 15), but must be used with caution because it has the potential to downgrade samples that only reflect illite alteration of feldspar. Therefore, it is best used in conjunction with alteration index $(\text{Na} + \text{K})/\text{Al}$.

The combination of alteration index $(\text{Na} + \text{K})/(\text{Fe} + \text{Mg})$ with all available pathfinder elements in the database reveal that U, Pb, B, Mg, Nb, Mo, Fe, Cu, Ni, As, P, Sc, W, Ce, Li, Nd and Y are positively anomalous in the deposits relative background, whereas Na, Rb, Sr, La and, where chlorite alteration was complete, K, were lost from the system. These pathfinder elements reflect alteration associated with both syn-ore and post-ore minerals in the U deposits, but discrimination between syn-ore and post-ore additions is not possible. Based on the paragenesis, U, Ce, Li, Nb, Nd, Y, Mo and B were introduced during the U mineralisation process as evidenced by the formation of uraninite with REE and trace amounts of tourmaline. Magnesium and Fe may have been added to form chlorite and hematite at this time, but the paragenesis records that the most significant introduction of Mg and Fe as chlorite and Fe-bearing sulfides occurred after U mineralisation ended. Paragenetic studies on all of the deposits in the ARUF show that radioactive Pb as a decay product of U, and Fe, Cu, Ni and As in sulfides were post-ore alteration additions. A small percentage (i.e. $< 1\%$) of highly altered samples show the addition of P, suggesting that apatite was locally formed in the deposits. However, the timing of apatite formation in the deposits is poorly documented.

DISCUSSION

Petrological and geochemical data collected from four well-documented U deposits (Jabiluka, Ranger, Koon-garra, Nabarlek), one prospect (King River) and a single drill intersection (Two Rocks) in the ARUF record unmistakable similarities in regards to alteration assemblages, timing of formation and source of fluids. This is despite the diversity in host rock lithologies. In the overlying Kombogie Subgroup sandstones, diagenetic minerals several kilometres from any known U mineralisation preserve a geochemical signature that is identical to that recorded by the alteration minerals in the ore deposits (Kyser *et al.* 2000; Polito *et al.* 2004, 2005; Kyser 2007). This suggests that a link exists between the deposits and the overlying sandstones. When the petrological and geochemical data from the sandstones, the basement rocks and the deposits is viewed holistically, a sound geological model that describes how the basin evolved and the deposits formed is generated and a method to explore for the deposits can be applied through lithogeochemistry.

The model describes the Kombogie Subgroup being deposited over the Nimbuwah Domain shortly after 1822 Ma, then experiencing a prolonged and locally complex diagenetic history. Argon ages show that the Kombogie

Subgroup sandstones and conglomerates were compartmentalised into diagenetic aquifers and diagenetic aquitards during burial diagenesis well before the intrusion of the Oenpelli Dolerite at 1723 Ma (Polito *et al.* 2005; Kyser 2007). In fact, $^{40}\text{Ar}/^{39}\text{Ar}$ ages and oxygen isotope analysis demonstrate that diagenesis at shallow depths in well-sorted, aeolian and marine sandstones resulted in the formation of aquitard units as early as 1798 Ma (Polito *et al.* 2005; Hiatt *et al.* 2007). The oxygen isotope data in particular show that the porosity-occluding quartz cement that characterises the diagenetic aquitards formed from mixed marine and meteoric fluids between 1 and 5 km depth (Hiatt *et al.* 2007).

In contrast to the diagenetic aquitards, the poorly sorted, fluvial clastic sediments within the Kombogie Subgroup experienced framework dissolution in the presence of marine-dominated basinal fluids during deep burial diagenesis. The abundance of illite $\pm$ chlorite replacing detrital grains in these rocks distinguishes them as the diagenetic aquifers. Temperature estimates from diagenetic illite indicate that the diagenetic aquifers likely formed between 4 and 8 km depth assuming a minimum geothermal gradient of 24°C (cf. Polito *et al.* 2006b). Petrological observations combined with $^{40}\text{Ar}/^{39}\text{Ar}$ ages from diagenetic illite (Figure 10; Polito *et al.* 2005) reveal that several diagenetic aquifers experienced a clay-forming fluid flow event during the 1755 to 1730 Ma mid-Tawallah compression event (Bull & Rogers 1996). A lack of younger ages in these diagenetic aquifers indicates that they were neither recharged nor overprinted by other, younger fluids. When some of these diagenetic aquifers near King River came in contact with the Oenpelli Dolerite some 10 to 20 million years later, the diagenetic illite and kaolinite were metamorphosed to pyrophyllite and muscovite. The presence of pyrophyllite reflects high-temperature contact metamorphism and indicates not only that these sequences were dry before the intrusion of the dolerite, but that they would not have been able to contribute to the U mineralisation event at *ca* 1680 Ma.

Other poorly sorted sandstone and conglomerate sequences, also recognised as being diagenetic aquifers, record younger $^{40}\text{Ar}/^{39}\text{Ar}$ ages that peak around 1680 and 1620 Ma (Figure 10; Polito *et al.* 2005). Evidence for fluids in these diagenetic aquifers, post-mid-Tawallah compression is recorded by the desilicification of sandstones adjacent to the 1723 Ma Oenpelli Dolerite and the formation of doubly terminated quartz crystals in silicified sandstone and massive veins and breccia pipes comprising euhedral quartz, tens of metres from the contact with the dolerite (Kyser *et al.* 2000). These features formed due to a change in silica solubility caused by convective circulation of available hot water. Fluid inclusions in the doubly terminated quartz crystals record temperatures up to 400°C and highly evolved Na–Mg–Ca–Cl brines with salinities of up to 22 wt% NaCl equivalent (Kyser *et al.* 2000). This brine composition is indistinguishable from that recorded by syn-ore quartz veins at Jabiluka (Polito *et al.* 2005) suggesting that the Kombogie Subgroup diagenetic aquifers had the same fluid-chemical characteristics as the U ore fluids before the U deposits formed. The spatial relationship between the diagenetic aquifers and the

location of the unconformity-related U deposits (Figure 2) supports the thesis that the mineralising fluids within the basin were confined to diagenetic aquifers and that where these diagenetic aquifers intersected favourable basement rocks, U was precipitated as uraninite (Hiatt & Kyser 2002).

U–Pb and $^{207}\text{Pb}/^{206}\text{Pb}$ ages from least altered uraninite and $^{40}\text{Ar}/^{39}\text{Ar}$ ages from syn-ore sericite in the deposits indicate that U precipitation first occurred at

1680 ± 10 Ma. This age is coincident with $^{40}\text{Ar}/^{39}\text{Ar}$ ages recorded by illite that formed in Kombolgie Subgroup diagenetic aquifers, suggesting that a fluid flow event in the Kombolgie Subgroup sandstones occurred at the same time as U mineralisation in the unconformably underlying basement (Figure 10). Other commonalities between the Kombolgie Subgroup diagenetic aquifers and the syn-ore alteration assemblages within the ARUF U deposits and prospects are recorded by stable isotopes.

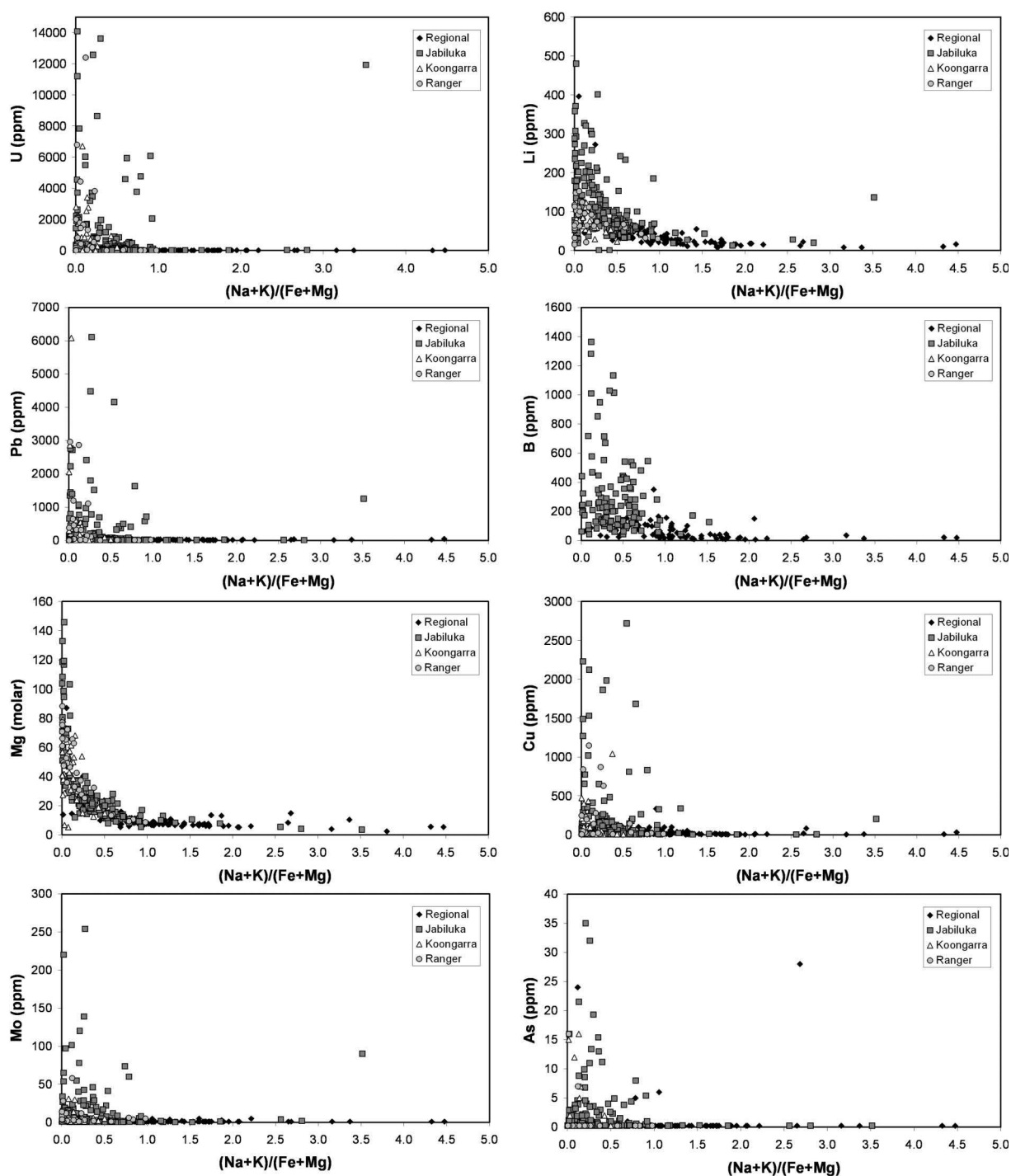


Figure 16 GER of $(\text{Fe} + \text{Mg})/\text{Al}$ vs $(\text{Na} + \text{K})/\text{Al}$ vs element concentration diagrams demonstrating that U, Li, Pb, B, Mg, Cu, Mo and As are elevated in the alteration zone, represented here by the smaller values along the x-axis. Elements that display similar properties but not shown here include that Br, Ce, Y, V and Nd.

The $\delta^{18}\text{O}_{\text{fluid}}$ and $\delta\text{D}_{\text{fluid}}$ values from syn-ore sericite and chlorite in the deposits are identical to those of illite in the Kombolgie Subgroup diagenetic aquifers (Figure 7).

The King River prospect (Figure 3), distinguished by its centimetre to metre scale alteration halos around disseminated and veinlet style uraninite and lacking any significant post-ore alteration, is unique and provides an insight into the U deposits at around 1680 Ma. The relatively unaltered alteration assemblage at the King River prospect demonstrates that chlorite and sericite/illite in approximately equal abundance together with uraninite, minor red hematite and, locally, quartz was the original alteration assemblage. This may have also been the case at other ARUF deposits, but all other ARUF deposits are characterised by an abundance of post-ore chlorite and drusy quartz $\pm$ carbonate $\pm$ sulfide veins. The domination by chlorite at some localities gives the impression that chlorite is the primary gangue mineral and a key vector into mineralisation. However, the alteration assemblage at the King River prospect suggests that such a chlorite-dominated assemblage may be a 'red-herring.' In this case, stable isotopes may aid decision making by explorers who intersect large chlorite-dominated alteration assemblages that do not appear to be associated with any significant U mineralisation (Figure 7).

PER and GER plots support petrographic observations that sericite/illite formed in response to albite and K-feldspar destruction and was then followed by chlorite precipitation in voids and as an alteration product of amphibole and biotite. This is best demonstrated in Figure 13b, where a horizontal trajectory of Na-bearing samples, trending toward the muscovite and illite node, dominates. Any indication of a downward curve toward the chlorite node, which would indicate co-precipitation with illite, is minimal. Consequently, the lithogeochemical plots cannot be used as evidence that chlorite and sericite/illite occurred in approximately equal abundance when uraninite first formed. However, Figure 13c does demonstrate that U mineralisation occurs equally in illite/sericite-dominated assemblages and illite-chlorite assemblages. There is no evidence to suggest that the highest U grades correspond to the most chlorite-dominated rock.

Whereas several papers have highlighted the importance of identifying chlorite alteration around unconformity-related U deposits during exploration, this paper advocates the importance of identifying Na-removal and illite/sericite formation remote from the mineralised zone. This is because chlorite alteration is best developed within the proximal halo and just beyond the distal/proximal boundary. Chlorite should be easy to identify in hand specimen, and a decision to follow-up its discovery should be relatively easy to make. In contrast, the cryptic illite/sericite alteration occurring up to several hundred metres away from most deposits can be difficult to identify in hand specimens, especially when radioactivity, U or other trace-element anomalism is absent. In this scenario, the sample is easily interpreted to be devoid of a mineralised signature. However, this paper demonstrates that PER and GER plots can identify a subtle alteration that may be associated with a U deposit several hundred metres

from mineralisation and therefore permits an informed decision regarding follow-up exploration methods.

Trace-metal abundance may also be used to vector into the deposit (Figure 16), but this study has shown that this only applies once inside the proximal alteration zone. This study complements the work by Ferguson & Winer (1980) and Binns *et al.* (1980b) in finding that U, Pb, Mo, Cu, B, Br, Ce, Y, Li, V and Nd are associated with the most intensely altered samples. However, it must be noted that PER/GER cannot discriminate between syn-ore and post-ore alteration, and therefore a range of trace elements, not all associated with U mineralisation, will be detected in the altered samples. The combination of syn-ore and post-ore elements in the mineralised zones should not discourage explorers from using any of these pathfinder elements to locate U deposits. Several studies have now demonstrated that the unconformity-related U deposits are associated with long-lived basement-rooted structures that were likely active during basin-fill, sandstone diagenesis and U mineralisation (Kyser *et al.* 2000; Alexandre *et al.* 2005, 2009; Polito *et al.* 2005, 2006b; Southgate *et al.* 2006; Kyser 2007). In most cases, these structures have been reactivated several times since initial uraninite precipitation as testified by the typically young ages determined for most U deposits in Australia and Canada (Hills & Richards 1976; Gulson & Mizon 1980; Fayek & Kyser 1997; Fayek *et al.* 2000, 2002; Kyser *et al.* 2000; Polito *et al.* 2004, 2005; Alexandre *et al.* 2005, 2009). Therefore, the conclusion is that elements known to be associated with post-ore alteration may be useful in unconformity-related U exploration because they too indicate formation in a long-lived structure that was originally active when U mineralisation formed. However, the onus is on the explorer to exercise caution.

CONCLUSIONS

Holistic basin analysis and detailed deposit studies in the Alligator Rivers Uranium Field demonstrate that the unconformity-related U deposits formed from fluids that resided and matured within the basin's sediments over a long period of time. Indeed, >100 million years passed between the first formation of diagenetic illite in diagenetic aquitards at 1798 Ma and the precipitation of U in the deposits at 1680 Ma. Periods of tectonic activity punctuate the state of the basin around 1755 Ma and 1723 Ma, and inform us that fluids were present in the basin at this time, but also had chemical characteristics similar to those that would eventually be recorded by alteration minerals in the deposits.

The data presented in this paper permit the development of a simple but robust model that likely has application to other basins that are considered prospective for unconformity-type U deposits. The model demonstrates that thick successions of poorly sorted, proximal fluvial sediments are a vital part of the basin's stratigraphy if it is to form a uranium deposit. These are the sediments that undergo diagenesis to become the fluid-bearing diagenetic aquifers when buried to depths between 4 and 8 km. If these sediments are not buried to at least 4 km, they do not undergo diagenetic

transformation and will not release the metals required to form world class U deposits. On the other hand, well-sorted, marine or distal fluvial sediments are consistently cemented by quartz at burial depths <4 km making them the diagenetic aquitards. A basin filled with only these sediments will not form U deposits.

The deposits, then, are a reflection of the basin's fluid chemistry. Petrological variations from deposit to deposit may reflect the characteristics of the host-rock, but appear to be more about the degree of post-ore alteration. Regardless of the host-rock reductant present, the deposits are consistently characterised by uraninite, illite/sericite, hematite and chlorite that formed at around 200°C. These syn-ore minerals are overprinted by later chlorite and quartz-carbonate veins, which have incorrectly been associated with the ore-forming process in the past and as such have been 'red-herrings' for explorers. Consequently, PER/GER plots that can identify the alteration of feldspar to illite/sericite several hundred metres away from the mineralised proximal zone may be a more useful tool for explorers who need to focus on this type of cryptic alteration.

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