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▶ To cite this version:

Joséphine Gigon, Julien Mercadier, Irvine Annesley, Antonin Richard, Andrew S. Wygralak, et al.. Uranium mobility and deposition over 1.3 Ga in the Westmoreland area (McArthur Basin, Australia). Mineralium Deposita, 2021, 56 (7), pp.1321-1344. 10.1007/s00126-020-01031-2. hal-04772490

HAL Id: hal-04772490 https://hal.science/hal-04772490v1

Submitted on 13 Feb 2025

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Uranium mobility and deposition over 1.3 Ga in the Westmoreland area (McArthur Basin, Australia)

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4 Joséphine Gigon¹, Julien Mercadier¹, Irvine R. Annesley¹, Antonin Richard¹, Andrew S. Wygralak², Roger G.

- 5 Skirrow³, Terrence P. Mernagh⁴, Ion Probe Team Nancy⁵
- 6 ¹Université de Lorraine, CNRS, CREGU, GeoRessources, F-54500 Vandœuvre-lès-Nancy, France
- ⁷ ²Northern Territory Geological Survey, PO Box 2901, Darwin, NT 0801, Australia
- ⁸ ³Geoscience Australia, GPO Box 378, Canberra, ACT 2601, Australia
- 9 ⁴Australian National University, 142 Mills Rd., Acton, ACT 2601, Australia
- 10 ⁵CRPG, UMR 7358, CNRS, Université de Lorraine, Vandœuvre-lès-Nancy F-54501, France
- 11 Corresponding author: josephine.gigon@gmail.com; +33 3 72 74 55 59
- 12 JG: <u>https://orcid.org/0000-0001-5971-9659</u>
- 13 RS: <u>https://orcid.org/0000-0002-9684-1554</u>
- 14 TM: <u>https://orcid.org/0000-0002-8555-0110</u>

15 Abstract

16 The Westmoreland area is located south of the McArthur Basin (Australia) and hosts several uranium (U) deposits and prospects. Previous studies proposed that U mineralisation formed under conditions similar to the 17 18 unconformity-related U deposits of the Alligator Rivers Uranium Field (ARUF), located north of the same 19 sedimentary basin. Detailed mineralogical, geochemical and geochronological studies on mineralised intervals from the Redtree, Junnagunna and Huarabagoo deposits, and integration with previous studies, identified at least 20 21 six generations of uranium oxides. These formed between ca. 1680 and 350 Ma, highlighting protracted mobility 22 of U over 1.3 Ga. Each generation of uranium oxide has a specific chemical composition, indicating variable 23 physico-chemical conditions for their formation throughout this period.

Although the 1680 Ma mineralising event formed at the same time and with similar mineralising fluids as the unconformity-related U deposits from the ARUF, the physico-chemical conditions differed between the two

26 areas. Deposits of the Westmoreland area lack the typical Mg- and B-metasomatism of the unconformity-related 27 U deposits in the ARUF: in the Westmoreland area, the chlorite is Mg-poor and no B minerals are observed, 28 whereas the ARUF is marked by Mg-rich chlorite (between the clinochlore and Mg-amesite fields), Mg-foitite 29 (tourmaline), and alumino-phosphate-sulphate minerals. The chondrite-normalized REE patterns of uranium 30 oxides are enriched in LREE or flat, compared with the typical bell-shape of the unconformity-related U deposits. Moreover, chlorite thermometry indicates significantly higher temperature conditions (>300°C) than in 31 32 the ARUF for the early U stages. Based on these results, it is thus unlikely that the mineralisation in the 33 Westmoreland area can be linked to significant unconformity-related U deposits in this area. Collectively, the 34 data point towards atypical ore-forming processes for basin-related U deposits in the Westmoreland area 35 compared with known processes in the ARUF and in other sedimentary basins.

36 Keywords

37 Uranium, Australia, Westmoreland, U-Pb isotopic ages, uraninite REE patterns, chlorite thermometry

38 1. Introduction

The Westmoreland area (Northern Territory and Queensland, Australia) hosts several U deposits and prospects 39 40 located in the Paleo- to Mesoproterozoic McArthur Basin and related crystalline basement (Polito et al. 2005a; 41 Ahmad et al. 2013; International Atomic Energy Agency 2018a, Figs. 1 and 2). The area hosts five deposits 42 (Redtree, Junnagunna, Huarabogoo, Outcamp and Sue) that are classified as Westmoreland-Murphy-type U 43 deposits (Lally and Bajwah 2006), and have indicated combined resources for all the deposits totalling $\sim 16\ 000\ t$ 44 U contained within 18.7 million tonnes at an average grade of 0.089% U₃O₈, and an inferred mineral resource 45 totalling ~ 7000 t U contained within 9.0 million tonnes at an average grade of 0.083% U₃O₈ (JORC and NI 43-46 101 compliant resource, Laramide Resources Ltd 2019). These deposits and related occurrences are part of a set 47 of deposits known in this large intracratonic basin, with several mineralised areas concentrating various types of 48 metals (U, Pb, Zn, and Cu, Ahmad et al. 2013). U deposits are found in the Pine Creek Orogen (northern part of 49 the basin) and in the Westmoreland area (southern part), Cu-bearing breccia pipes and minor vein-type 50 mineralisation are present in the Redbank area in the southeastern part of the basin (e.g. Knutson et al. 1979), 51 and SHMS Zn-Pb deposit are mostly found in the centre of the basin (e.g. Gigon et al. 2020).

52 The geological processes and physico-chemical conditions for the origin of the different U deposits found in the McArthur Basin have been studied for decades. The Alligator Rivers Uranium Field (ARUF) to the North hosts 53 54 the Jabiluka, Ranger, Nabarlek and Koongarra U deposits among others belonging to the unconformity-related 55 deposits type, for which the processes related to the U deposition have been studied extensively (e.g. Wilde and 56 Wall 1987; McKay and Miezitis 2001; Derome et al. 2003; Polito et al. 2004, 2005b, 2011; Lally and Bajwah 57 2006; Fisher et al. 2013; Clauer et al. 2015; Skirrow et al. 2016; Gigon et al. 2019). U deposits from the 58 Westmoreland area have comparatively attracted much less attention (Hills and Richards 1976; Fuchs and 59 Schindlmayer 1981; Ahmad and Wygralak 1989 and references herein; Rheinberger et al. 1998; Polito et al. 2005a; Mernagh and Wygralak 2011), and the previous research (e.g. Polito et al., 2005a and Mernagh and 60 61 Wygralak 2011) mainly examined if these U deposits and related occurrences could be analogues to the 62 unconformity-related U deposits known at the border of the Northern part of the McArthur Basin, but also in the Athabasca and Thelon basins (Canada; Jefferson et al. 2007; Cuney and Kyser 2015; Grare et al. 2020). 63 Unambiguously establishing such relationships with unconformity-related U deposits is indeed important 64 because unconformity-related U deposits have the highest grades and tonnages among all the U deposits, and are 65 66 therefore dominant U exploration target worldwide.

67 The results from the previous studies in the Westmoreland area indicate that the mineralogy, paragenesis and 68 geochemistry of the Redtree, Junnagunna and Huarabagoo deposits share some characteristics with the U 69 deposits of the ARUF (Polito et al. 2005a), thus suggesting that the timing and geological processes for the 70 origin of both U provinces were similar. The fluid inclusions in the Eva Mine and Jackson Pit U deposits, 71 yielded NaCl-rich and CaCl₂-rich brines and a low-salinity fluid that mixed in varying degrees (Mernagh and 72 Wygralak 2011), as also documented for the unconformity-related U deposits from the ARUF (Ypma and 73 Fuzikawa 1980; Durak et al. 1983; Derome et al. 2003, 2007; Polito et al. 2004, 2005b). These combined results 74 seem to indicate that the first-order parameters and the timing for the formation of both uranium provinces in the 75 McArthur Basin are identical. The main goal of the present work is thus to refine this comparison between the U deposits of the ARUF and the Westmoreland area by verifying the possible metallogenetic relationships between 76 77 these two deposit subtypes.

New geochemical criteria applicable to uranium oxides, based on rare earth element content in uranium oxides, allow classification of the different types of U deposits (Mercadier et al. 2011b; Frimmel et al. 2014; Alexandre et al. 2016). Such criteria have been applied to the U deposits from the ARUF, confirming their affinity with the U deposits from the Athabasca Basin (Canada) and thus their classification as unconformity-related U deposits

82 (Mercadier et al. 2011b). Application of such geochemical markers is lacking for the U deposits of the Westmoreland area and this forms the first objective of this study. The second objective is to add to the existing 83 84 data on ore mineralogy, geochemistry and age dating for different mineralised intercepts of three U deposits 85 from Westmorland area: Redtree, Junnagunna and Huarabagoo. The final objective of this approach is to provide new geochronological constraints on formation of the U deposits and occurrences of the Westmoreland area, in 86 particular the characterisation of potential multiple fluid events that favoured U mobility through time and space. 87 88 These new results are compared to previously published models for the area, to the unconformity-related 89 deposits of the ARUF, and to U deposits in other Proterozoic basins.

90 2. Geological settings

91 The Westmoreland area is adjacent to the Queensland-Northern Territory border on the south-eastern margin of the southern 92 McArthur Basin (Fig. 1). The oldest rocks in the Westmoreland region are Paleoproterozoic metasedimentary, volcanic and 93 felsic rocks from the Murphy Province that forms the basement to the McArthur Basin. The units in the Murphy Province are 94 the Murphy Metamorphics and a suite of comagmatic felsic intrusive and volcanic rocks known as the Nicholson Granite and 95 the Cliffdale Volcanics. The Murphy Metamorphics are essentially a succession of greenschist-facies metamorphosed shales 96 and greywackes (Ahmad et al. 2013). Uranium-lead data on detrital zircon suggest a maximum depositional age of 1853 ±4 97 Ma (Hollis et al. 2010, Table SM1) and the sediments were metamorphosed prior to the extrusion of the Cliffdale Volcanics 98 (Ahmad et al. 2013). The Nicholson Granite Complex (Fig. 2) is intrusive in the Murphy Metamorphics and can be divided 99 into two broad suites based on chemical and mineralogical composition, one hornblende- and/or biotite bearing granite, 100 quartz monzonite and granodiorite and one biotite- and/or muscovite bearing monzonite, granite and alkali granite. Two 101 samples of the Nicholson Granite Complex have yielded SHRIMP U-Pb zircon ages of 1856 ±3 and 1845 ± 3 Ma (Page et al. 102 2000, Table SM1). The Cliffdale Volcanics are a succession of more than 4km of rhyolite, dacitic lavas, ignimbrites and 103 intrusions that unconformably overlie the Murphy Metamorphics (Ahmad et al. 2013). SHRIMP U-Pb dating of zircons has 104 yielded an age of 1851 ± 3 Ma (Page et al. 2000, Table SM1).

The Paleo–Mesoproterozoic McArthur Basin is an up to 10 km thick sequence of mostly unmetamorphosed sedimentary and volcanic rocks deposited between 1815 and 1492 Ma that unconformably overlies the Murphy Province (Ahmad et al. 2013). The basal unit of the McArthur Basin, the Westmoreland Conglomerate (Fig. 2), is composed mainly of fluvial sandstones and conglomerates and is up to 1,800 m thick. It has been subdivided into five stratigraphic units, each unit representing a major fining-upward cycle of sedimentation (Ahmad and Wygralak 1989). Each unit comprises proximal fluvial deposits typical of debris flows, alluvial fans, and braided river systems overlain by medium- to coarse-grained, well-sorted sandstones. Angular unconformities or 112 disconformities define breaks in sedimentation, with each new cycle of pebble or boulder conglomerate 113 commonly defining the beginning of a new unit. Cobbles and coarse sand grains within the basal conglomerate 114 are dominated by reworked quartz veins, chert, and clasts of felsic to mafic volcanic rocks that were likely 115 derived from basement rocks such as the Murphy tectonic ridge or similar rocks that once existed to the North 116 (Wygralak et al. 1988; Ahmad and Wygralak 1989; Polito et al. 2005a). The detrital lithic clasts, monazite, 117 zircon, and apatite are likely to be among the source of uranium that was dissolved during peak diagenesis 118 (Polito et al. 2005a and references herein). Most of the uranium mineralisation is hosted within the upper unit of 119 the Westmoreland Conglomerate (Ptw4), which is a 80-90 m thick porous, coarse-grained sandstone, 120 conglomeratic in part (International Atomic Energy Agency 2018a). U-Pb dating of detrital zircon from the 121 Westmoreland Conglomerate has returned maximum depositional ages of 1865 ± 7 Ma for the lower section and 122 1843 ± 4 Ma for the upper section (Ahmad and Wygralak 1990, Table SM1). The Westmoreland Conglomerate 123 is conformably overlain by the mafic Seigal Volcanics (basaltic lavas), followed by dolomite, sandstone, mafic 124 and felsic volcanic rocks of the upper part of the Tawallah Group. The deposition age of the Seigal Volcanics is 125 estimated at ca. 1780 Ma by stratigraphic correlation (Scott et al. 2000; Mernagh and Wygralak 2011).

Dolerite dykes and minor sills, which may have been feeders of the lava flows, intrude along North-Est trending fault and fracture zones that intersect the Westmoreland Conglomerate (International Atomic Energy Agency 2018b). Uranium mineralisation is associated with three major northeast-trending lineaments (NE Westmoreland, Redtree and El Nashfa dyke zones, Fig. 1), with the Redtree dyke zone extending over a strike of 20 km (Ahmad et al. 2013). The dykes has not been dated but they could have the same age as the Seigal Volcanics (Ahmad and Wygralak 1989).

132 The Westmoreland region was first prospected in the 1890s for Pb-Zn, then for Cu, and finally U was mined 133 from the Peters Creek Volcanics which overlie the Westmoreland Conglomerate, 20 to 30 km west of Redtree 134 (Laramide Resources Ltd 2016). The Redtree, Junnagunna and Huarabagoo U deposits are hosted largely within 135 the shallow dipping Westmoreland Conglomerate near the unconformity with the Murphy Inlier and lie along the 136 Redtree dyke zone (Fig. 1). The U-rich zones can be oriented parallel to the volcanic rocks or sills, discordant at 137 the contact with mafic dykes, within fractures cross-cutting altered mafic volcanic rocks within fractures or 138 along shear zones in altered felsic volcanic rocks. The studied deposits belong to "mafic dykes/sills in 139 Proterozoic sandstone type" (International Atomic Energy Agency 2018b) or "Westmoreland-Murphy-type" 140 (Lally and Bajwah 2006). The deposits of the "mafic dykes/sills in Proterozoic sandstone" category are situated 141 in the Westmoreland area and within the Otish Basin (Canada). "Westmoreland-Murphy-type" deposits occur

142 within conglomerate, sandstone and mafic volcanic rocks, and are all associated with redox boundaries near the 143 contacts between different lithologies. In addition, in the studied region, uranium occurrences were classified by 144 Ahmad (1987) into five types (A-E), based on their hydrological and geological settings. Four of them are 145 present in the basin, and reported on Fig. 1. Type A deposits lie at the contact between the Westmoreland Conglomerate and the Cliffdale or Seigal Volcanics (in sub-type A1 occurrences, the contact between the 146 147 Cliffdale Volcanics and Westmoreland Conglomerate is a reverse fault; in sub-type A2 deposits, the contact 148 between the Westmoreland Conglomerate and Seigal Volcanics is conformable). In type B deposits, 149 mineralisation occurs as sub-horizontal and sub-vertical lenses in the Westmoreland Conglomerate. Type C 150 deposits are hosted by intensely altered Cliffdale Volcanics; Type D occurrences are related to fractures in the 151 lower part of the Seigal Volcanics.

The bulk of the Westmoreland resource base (\sim 50%) is located in the Redtree deposit. It flanks the Redtree dyke 152 153 zone immediately north of the northwest-trending Namalangi fault (Fig. 1). The Redtree dyke (20-30 m wide) 154 broadly extends for 7 km from Redtree to Junnagunna and spatially controls the distribution of U, indicating that mineralizing fluids used this structure as a main drain uranium mineralising fluids probably used this structure 155 156 (Laramide Resources Ltd 2019). The deposit is characterised by a mix of horizontal and vertical lenses of 157 continuous mineralisation in distinctly coarse pebble conglomerates with grades ranging from 0.15% to >2% 158 U₃O₈ (Fig. 2). Mineralisation is generally shallow, within 10m of surface and is associated with chlorite/hematite 159 alteration. The mineralisation thickens and steepens near the dyke, where it is 30-40 m thick (International 160 Atomic Energy Agency 2018a). The indicated resource is 12.86 Mt of ore at an average grade of 0.09% U₃O₈ 161 and an inferred resource of 4.47 Mt at an average grade of 0.07% U₃0₈ (JORC and NI 43-101 compliant, Laramide Resources Ltd 2016). 162

The Junnagunna U deposit occurs at a fault intersection west of the Redtree dyke zone and south of the northwest trending Cliffdale fault (Fig. 1). Mineralisation is generally flat lying on both sides of the Redtree dyke and ranges from 0.5 m to 20 m in thickness beneath the Seigal-Westmoreland contact (Fig. 2). The deposit is covered by alluvium/colluvium clays, sand and weathered Seigal Volcanics (Laramide Resources Ltd 2019). The indicated resource is 4.36 Mt ore at an average grade of 0.08% U₃0₈ and an inferred resource of 2.15 Mt at an average grade of 0.8% U₃0₈ (Laramide Resources Ltd 2016).

The Huarabagoo deposit is about 3 km NE of Redtree along the Redtree dyke zone and straddles the contact of the Seigal Volcanics with the Westmoreland Conglomerate (Figs. 1 and 2). Mineralisation occurs in 20 m thick 171 lenses, which extend to 100-200 m length and to a depth of 80 m (Laramide Resources Ltd 2019). It outcrops at the southern end and is concealed to the north under 2 to 3 m of sandy alluvium and 5 to 8 m of weathered basalt 172 173 of the Seigal Volcanics (Laramide Resources Ltd 2016). The deposit comprises a 3 km zone of vertical 174 mineralisation associated with a complex dyke geometry with vertical and horizontal branches between the two 175 principal dykes. The strongest mineralisation is associated with chlorite/hematite-altered coarse pebbly sandstones similar to Redtree. Most of the mineralisation is hosted within the Westmoreland Conglomerate 176 177 adjacent to the dykes, and the remainder is hosted in the dykes (International Atomic Energy Agency 2018b). Indicated resources are 1.46 Mt ore at an average grade of 0.08% U₃0₈ and inferred resources are 2.41 Mt at an 178 179 average grade of 0.11% U₃0₈ (Laramide Resources Ltd 2016).

180 **3. Sampling and methodology**

A multi-method approach was conducted on different minerals (uranium oxides, chlorite and apatite), from the Junnagunna, Redtree and Huarabagoo deposits. A specific focus was the selection of samples within the highestgrade parts of mineralised zones of the deposits, all located within the Westmoreland Conglomerate (Fig. 2). This selection was also based on the paragenesis for the U deposits of the Westmoreland area established by Polito et al. (2005a).

186 Five drill holes were sampled in three deposits based on their high U grade, representative of the different mineralisation styles: JDD08-003 (Junnagunna), WDD9-128 (Huarabagoo), WDD10-151 (Huarabagoo), 187 188 WDD07-2 (Redtree), and WDD08-106 (Redtree). Mineralised intersections are: at Junnagunna, 0.21% U₃O₈ in 189 JDD08-003 from 71 to 76m; at Huarabagoo, 0.16% U₃O₈ in WDD10-151 between 22 and 31m; at Redtree, 0.25% U₃O₈ in WDD07-02 from 48 to 59m and 0.2% from 70 to 75m and 0.31% in WDD08-106 from 8 to 16m 190 191 (Vigar and Jones 2009; Laramide Resources Ltd 2016). The following samples were studied: JDD08-003 at 192 74.4m, WDD07-2 at 51.7, 55.9 and 73.5m, WDD9-128 at 53.3 and 92.3m, WDD10-151 at 29.4m and WDD08-193 106 at 10.6m.

Selected thin sections were studied by optical and scanning electron microscope (SEM) to develop detailed paragenetic sequences. The chemical composition for the major elements of chlorite and uranium oxide was determined by Electron Probe Micro Analyses (EPMA). The isotopic U-Pb and Pb/Pb ages of uranium oxide and apatite were determined by Secondary Ion Mass Spectrometry (SIMS). The concentrations of minor and trace elements in uranium oxides were determined by Laser Ablation-Inductively Coupled Plasma-Mass Spectrometer
 (LA-ICP-MS). The locations of the analyses are presented in Figs. SM1–SM8.

200 3.1. Petrography

Optical microscopy was carried out on thin sections using a numerical microscope VHX-200F with a high resolution objective VH-Z500W to cover the whole sections and a Zeiss Axio Scope A1 microscope for detailed petrography (GeoRessources Lab - Vandœuvre-lès-Nancy, France). SEM images and spectra were obtained using BackScattered Electron detector (BSE) Tescan Vega3LM with a conventional tungsten source at SCMEM (GeoRessources Lab - Vandœuvre-lès-Nancy, France). The analytical conditions were an acceleration voltage of 15 kV and beam current of around 0.1–1 nA.

207 3.2. Major elements composition of uraninite and chlorite

208 The quantitative measurements of major elements in uraninite and related chlorite were performed using a 209 CAMECA SX-100 Electron Probe MicroAnalyser (EPMA) at SCMEM with a spot size of 1µm². For uraninite, 210 the calibration was based on natural and synthetic oxides and/or alloys (V for V, andradite for Ca, hematite for 211 Fe, albite for Si, apatite for P, zircon for Zr, UO₂ for U, MnTiO₃ for Mn and Ti, Al₂O₃ for Al, ThO₂ for Th, PbS 212 for Pb and YPO4 for Y). The analytical conditions were 20 nA current, accelerating voltage of 20 kV and 213 counting time of 10 s. For chlorite, the calibration was also based on natural and synthetic oxides and/or alloys 214 (V for V, andradite for Ca, orthose for K, hematite for Fe, albite for Si and Na, olivine for Mg, MnTiO₃ for Mn and Ti, Al_2O_3 for Al, and Cr_2O_3 for Cr). The analytical conditions were 12 nA current, accelerating voltage of 15 215 216 kV and counting time of 10 s. The limits of detection of each analysed element are presented in Table SM2 217 (uraninite) and Table SM12 (chlorite).

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3.3. U-Pb isotopic ages of uranium oxides and apatite

SIMS is an established approach to date apatite (Li et al. 2012; Zhou et al. 2018) and uranium oxide (Fayek et al. 2000, 2002; Luo et al. 2015; Martz et al. 2019). The U-Pb and Pb/Pb analyses were carried out at the Centre de Recherches Pétrographiques et Géochimiques (CRPG, Nancy, France) with the Cameca IMS 1280 HR2 and the Hyperion RF O⁻ source for apatite and with the Cameca IMS 1270 E7 and the duoplasmatron O⁻ source for uranium oxide. The secondary positive ions were measured with a mass resolution (M/ Δ M) of 6000 to separate peaks of interest from isobaric interferences, by ion counting in mono-collection mode on the axial electron multiplier. 226 For apatite, the primary O⁻ beam was set at an intensity of 30nA with a 10 µm spot size. Each measurement 227 consisted of a 120 s pre-sputtering, secondary beam and mass centring and 16 cycles of measurement of masses: 178.5 (background), ⁴⁰Ca⁴⁴Ca³¹P¹⁶O₄, ²⁰⁴Pb, ²⁰⁶Pb, ²⁰⁷Pb, ²⁰⁸Pb, ²³⁸U, ²³²Th¹⁶O and ²³⁸U¹⁶O. Counting times were 228 4 s per peak for ²³²Th¹⁶O and ²³⁸U¹⁶O, 6 s per peak for ²³⁸U, 8 s per peak for ²⁰⁸Pb and 18 s per peak for ²⁰⁴Pb, 229 ²⁰⁶Pb and ²⁰⁷Pb. An energy window of 30 eV was used with no energy offset. ⁴⁰Ca⁴⁴Ca³¹P¹⁶O₄ was used as a 230 231 reference mass to calculate the U and Pb contents. The Madagascar apatite was used as standard to determine the relative ionization yield of U and Pb, with the Pb/U ratio expressed as a function of UO/U ratio. The external 232 error on the ²⁰⁶Pb/²³⁸U ratios, measured on the Madagascar standard (dated at 473.5 Ma, Chew et al. 2019) for 233 each session, was propagated on the final error of each sample measurements. With the measurement of the ²⁰⁴Pb 234 isotope it is possible to calculate ²⁰⁶Pb/²³⁸U corrected ages using the measured ²⁰⁴Pb/²⁰⁶Pb ratios and the 235 236 continental common lead compositions from Stacey and Kramers (1975) at the age of crystallisation. This allows us to calculate probability density plots and weighted average plots using ²⁰⁶Pb/²³⁸U corrected ages, and compare 237 them to the common lead uncorrected Tera-Wasserburg intercept ages (Tera and Wasserburg 1972). All data 238 239 were treated using ISOPLOT software (Ludwig 2007).

For uranium oxides, the primary O⁻ beam was set at an intensity of ~5 nA and focussed to a spot of ~15 μ m in 240 241 diameter, in Gaussian mode with a raster of 10µm. The energy window was opened at 30 eV and centred on the low energy side, 5 eV below the maximum value. An offset of -50eV was applied during analyses in order to 242 243 avoid matrix effects due to mixes between uraninite and silicate in natural samples. Ions were measured by peak jumping in monocollection mode using the axial Faraday cup (FC) for ²³⁸U and ²³⁸UO and the axial electron 244 multiplier (EM) for ²⁰⁴Pb, ²⁰⁶Pb, ²⁰⁷Pb, ²⁰⁸Pb and ²³²ThO. Each analysis consisted of 8 successive cycles. Each 245 cycle began with measurement of the mass 203.5 and 203.6 for backgrounds of the FC and the EM. The 246 counting time was 4 s for 203.5, 203.6, ²⁰⁸Pb and ²³⁸UO; 3 s for ²⁴⁸ThO; 6 s for ²⁰⁶Pb and ²³⁸U; 10 s for ²⁰⁴Pb and 247 248 20 s for ²⁰⁷Pb (waiting time of 1 s). The standard used was a uraninite from Zambia (Holliger 1988), which has a 249 given concordant age at 540 ± 4 Ma (Cathelineau et al. 1990). Ages and error correlations were calculated using ISOPLOT (Ludwig 2007). Uncertainties in the ages are reported at the 2σ level. Common Pb corrections were 250 251 based on the measured ²⁰⁴Pb content using the Pb isotope composition calculated from Stacey and Kramers 252 (1975) at the inferred age of the uranium oxide crystallisation.

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3.4. Minor and trace element concentrations in uranium oxides

254 Minor and trace elements in uranium oxides, including rare earth elements (REEs), were measured with a Laser 255 Ablation-Inductively Coupled Plasma-Mass Spectrometer (LA-ICP-MS) at the GeoRessources laboratory 256 (Nancy, France). The uranium oxides were ablated using a 193 nm Geolas Q Plus system with an aperture-257 imaged Complex 103 ArF excimer laser (Microlas, Göttingen, Germany) equipped with beam homogenisation optics. The laser spot sizes were 16 and 24 μ m and the fluence was ~5 J/cm² with a repetition rate of 5 Hz. 258 Ablated particulate material was analysed by an Agilent 7500c Quadrupole ICP-MS. The transport gas was a 259 260 mixture of He and Ar, at typical flow rates of 0.5 and 0.8 l.min⁻¹ respectively. All data were acquired in raw 261 counts using time-resolved mode. Data reduction was done using Iolite software (Paton et al. 2011). Absolute 262 concentrations and limits of detection were obtained from the equation developed by Longerich et al. (1996). 263 The calibration material (external standard) was a SRM NIST 610 glass standard and SRM NIST 612 glass was analysed to control the precision and accuracy of the measurements (concentrations from Jochum et al. 2011). 264 265 The methodology was modified from Lach et al. (2013). The matrix effects when quantifying the REEs in a 266 uraninite using SRM NIST 612 are limited (< 15%; Lach et al. 2013) and are considered to be in the same range 267 for the other measured minor and trace elements in the present study, as previously proposed by Frimmel et al. (2014) and Alexandre et al. (2016). A series of 37 elements were analysed for each experiment: ¹¹B, ⁴⁷Ti, ⁵¹V, 268 ⁵³Cr, ⁵⁵Mn, ⁵⁹Co, ⁶⁰Ni, ⁶³Cu, ⁶⁶Zn, ⁶⁹Ga, ⁷⁵As, ⁸⁸Sr, ⁸⁹Y, ⁹⁰Zr, ⁹³Nb, ⁹⁵Mo, ¹²¹Sb, ¹³⁷Ba, the 14 REEs, ¹⁸¹Ta, ¹⁸²W, 269 ²⁰⁹Bi, ²³²Th and ²³⁸U. ²³⁸U was the internal standard measured by EPMA and the used concentration was adjusted 270 depending of the analysed sample: 77.1 wt% U for WDD07-2 55.9A, 76.8 for WDD07-2 55.9B, 75.6 for 271 272 WDD10-151 29.4, 75.2 for WDD08-106 10.6, 74.9 for WDD07-2 73.5B and 72.1 for JDD08-003 74.4B. The 273 analytical precision calculated for all elements using standard deviation of recorded intensity is better than \pm 274 20%. The limit of detection (LOD) varies for each element and each analysis as a function of signal intensity and 275 was calculated for each ablation from signal intensities using the 3σ criterion (Longerich et al. 1996). The LODs 276 were reported in Table SM3 when the calculated values for a given element fell below these values. Values used 277 to calculated ($\Sigma LREE / \Sigma HLREE)_N$ are chondrite-normalised with chondrite values from Anders and Grevesse 278 (1989) following elements used for calculation: and the were ΣREE=La+Ce+Pr+Nd+Sm+Eu+Tb+Dy+Ho+Er+Tm+Lu; 279

 $280 \qquad (\Sigma LREE / \Sigma HREE)_N = (La_N + Ce_N + Pr_N + Nd_N + Sm_N;) / (Er_N + Tm_N + Lu_N)..$

Correlations matrices (Tables SM4 – SM9) were generated using the ioGAS software. The chosen criteria for a positive correlation between two elements was a correlation coefficient higher than 0.9, and lower than -0.9 for an anti-correlation.

284 **4. Results**

285 4.1. Paragenesis

286 The paragenesis presented in this section is based only on the observations from the samples for this study. A 287 combined paragenesis from this study and Polito et al. (2005a) is presented in Fig. 3 and discussed in the 288 "Discussion" section, where the first uranium oxide generation (U1) and the third one (U3) are presented. In the mineralised intercepts, the early diagenesis is represented by hematite (H1) crystallisation and quartz 289 290 overgrowths (Q1) on detrital quartz (Q0) grains (Fig. 4a). This was followed by an episode of dissolution of 291 detrital quartz Q0, noted as Q⁻ in Fig. 4b, in relation to peak diagenesis. Chlorite in rosette-shape (C1, Fig. 4c), rutile (R1, Fig. 4d), clays minerals (dickite D1 and illite I1) and apatite (Ap; mostly 10-20 µm in width, Fig.5a) 292 293 form during this stage. Chlorite C1 is altered into clay minerals (I1, Fig. 4e). A second generation of quartz (Q2, 294 Fig. 4f) occurs at the end of this stage. A second generation of uranium oxide (U2) crystallises in contact with 295 apatite (Ap) in voids produced by quartz dissolution (Fig. 5a). The U2 postdates apatite as U2 fills small 296 fractures within apatite grains and surrounds them. Veins containing quartz (Q3), uraninite (U4), pyrite (Py3) 297 and chalcopyrite (ccp) crosscut the sandstone (Fig. 5c) and they are thought to be coeval with U4 emplaced in 298 voids (Fig. 5b) based on dating results. A fifth (U5) and sixth (U6) generations of uranium oxide occur (Fig. 5d), 299 filling voids. The mineral assemblage is similar for three last uranium generations, but the ages obtained account 300 for the different generations.

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4.2. Chemical composition of the uranium oxides

The chemical composition of the different uranium oxides are reported in Table SM2 for the major elements and in Table SM3 for minor and trace elements. Fig. 6 shows the chemical maps obtained by EPMA, on which the scales were determined using element concentrations measured during *in-situ* spot EPMA analyses.

305 Major elements

The U contents of the different generations of uranium oxides range from 63.2 to 90.15 wt% UO₂. The thorium contents for all the uranium oxides is low with concentrations below the detection limits (0.1 wt% Th). The different uranium oxides are characterised by variable contents of Pb, either between samples or within an individual sample (Figs. 6 and 7a). For each sample, the maximum Pb content, corresponding to the least altered zones, and the minimum Pb content which is measured in the most altered ones, are summarized in Table SM2. 311 The samples WDD10-151 29.4 (U4) and WDD08-106 10.6 (U5) have the most homogeneous Pb contents and 312 can be considered as the least altered. The Ti contents are below the detection limit (0.07 wt% TiO₂) except for samples JDD08-003 74.4B (U2) with concentrations up to 1.39 wt% TiO2, WDD07-2 55.9A and WDD07-2 313 314 55.9B (U4) up to 2.6 wt% TiO₂ and WDD07-2 73.5B (U6) up to 1.25 wt% TiO₂. The P contents are lower than 2 315 wt% P₂O₅ in sample WDD07-2 73.5B (U6), but can reach 6.93 wt% P₂O₅ in zones where U concentrations are 316 low in samples WDD07-2 55.9A and WDD07-2 55.9B (U4). The calcium and silica contents are also highly 317 variable, ranging from 0.98 to 4.16 wt% CaO with the highest values found in sample JDD08-003 74.4B (U2), WDD07-2 55.9A and WDD07-2 55.9B (U4), and from < LOD (0.1 wt% SiO₂) to 13.11 wt% SiO₂ in zones with 318 319 low U concentrations. The silica contents are higher than the calcium contents for all the samples except 320 WDD08-106 10.6 (U5). The alteration for samples WDD07-2 55.9A, WDD07-2 55.9B (U4) and WDD07-2 321 73.5B (U6) is marked by a loss in lead compensated by a gain in Si, Ca, Fe and P (Fig. 7a), which is not 322 observed for the other samples.

323 Minor and trace elements

324 Among all the minor and trace elements measured by LA-ICP-MS, 14 have contents below the limit of detection 325 (LOD) for some of the analyses. Table SM3 presents the results, with the minimal and maximal values for each 326 sample, and the maximum LOD value calculated for each element considering all samples and all analyses. The 327 diagrams comparing the contents in As, Nb, W, Ba and V (Fig. 7 b, c, d) demonstrate that the different uraninite 328 generations are chemically distinct. Arsenic contents are low in sample WDD07-2 73.5B (U6, 807–4870 ppm), 329 range between 2490 and 5130 ppm for WDD07-2 55.9A (U4), and the highest values are obtained in sample JDD08-003 74.4B (U2, 5750-17590 ppm), WDD10-151 29.4 (U4, 14300-25800 ppm) and WDD08-106 10.6 330 331 (U5, 10120–13130 ppm, Fig. 7b and Table SM3). Niobium contents range between 5.6 and 137 ppm for JDD08-332 003 74.4B (U2), between 78.2 and 147.8 ppm for WDD07-2 55.9A (U4), between 6.3 and 16.9 ppm for 333 WDD10-151 29.4 (U4), between 1.56 and 2.7 ppm for WDD08-106 10.6 (U5), and between 0.24 and 108.8 ppm 334 for WDD07-2 73.5B (U6). Tungsten contents range between 67.3 and 156.6 ppm for WDD10-151 29.4 (U4 in 335 veins) and between 551 and 8350 ppm for the other samples. Barium contents range between 227 and 2280 ppm 336 for JDD08-003 74.4B (U2), between 101 and 283 ppm for WDD10-151 29.4 (U4), between 32.6 and 856 ppm 337 for WDD07-2 55.9A (U4), between 69.4 and 77.6 ppm for WDD08-106 10.6m (U5), and between 584 and 7780 338 ppm for WDD07-2 73.5B (U6). Vanadium contents range between 469 and 1637 ppm for WDD07-2 55.9A 339 (U4), and between 1418 and 16510 ppm for JDD08-003 74.4B (U2), WDD10-151 29.4 (U4 in veins), WDD08-340 106 10.6 (U5), and WDD07-2 73.5B (U6).

341 Rare Earth Elements

342 The REE chondrite-normalised patterns of the uranium oxides from the different samples are variable, but are all 343 characterised by a relatively high concentrations in REEs (between 537 and 40169 ppm ΣREE , Fig. 7e) and an 344 enrichment in LREE and MREE compared to HREE. The REE patterns are homogeneous for a given sample, 345 except for samples JDD08-003 74.4B, WDD07-2 55.9A and WDD07-2 55.9B (Fig. 8). The sample JDD08-003 346 74.4B (U2) is marked by a variation of MREE and HREE (1709 to 42014 ppm Gd and 154 to 6778 ppm Lu for 347 example), whereas sample WDD07-2 55.9A and WDD07-2 55.9B (U4) are marked by a variation of LREE (19 348 to 2493 ppm La for example). Increased LREE contents correlate to higher degrees of alteration in samples 349 WDD07-2 55.9A and WDD07-2 55.9B, whereas no link between alteration and MREE and HREE variations 350 could be established in sample JDD08-003 74.4B. Some of the samples display a negative Eu anomaly (WDD07-2 55.9A and WDD07-2 55.9B, WDD10-151 29.4, WDD07-2 73.5B). 351

352 *Correlations between elements*

353 Some minor and trace elements are correlated in all the samples, whereas some others are only correlated in 354 some of the uraninite generations. The correlation matrices are presented in Tables SM4-SM9. In all samples the 355 following elements have a positive correlation: Co and Ni; the REEs; and Ba and Ga. In sample JDD08-003 356 74.4B, B is correlated to V, Zn, Ga and Ba; Mn is anti-correlated to the REEs; Bi is correlated to Co and Ni 357 (Table SM4). In sample WDD07-2 55.9A, V is correlated to Ba and the LREE; Mn to Sb; Cu to Ga, Sr and Ba 358 (Table SM5). In sample WDD07-2 55.9B, the REEs, Zr and Th are correlated; B is correlated to Mn and Sb, and 359 anti-correlated to Sr (Table SM6). In sample WDD10-151 29.4, B is correlated to the REEs but anti-correlated to 360 V and Mn (Table SM7). In sample WDD08-106 10.6, As is correlated to the HREE and W, and anti-correlated 361 to V, Mn and Co; Mo is anti-correlated to the LREE (Table SM8). In sample WDD07-2 73.5B, Co is correlated 362 to Cu and Ga; Ti to Nb; B to Co; Ba to Ga and Sr (Table SM9).

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- 364

4.3. Isotopic ages of apatite and uranium oxides

365 Apatite

Apatite predates uranium oxides, as observed in sample JDD08-003 74.4B (Fig. 9a). A significant number of analyses present anomalous U and/or Pb contents correlated to anomalous Pb/Pb or U/Pb ratios (Table SM10 and Fig. 9b). These anomalous values are probably linked contribution from uraninite U2 during the analyses of some apatite grains, due to the relatively small size of the apatite grains. When discarding such data, the eight remaining analyses are marked by variable concentrations of common Pb and degrees of discordance, with $^{204}Pb/^{206}Pb$ and $^{207}Pb/^{206}Pb$ ratios ranging respectively from 0.0015 to 0.0350 and 0.1281 to 0.5846 (Table SM10). The analyses plot in a discordant position on a Tera-Wasserburg diagram with the calculated lower intercept giving a poorly-defined age of 1685 ± 65 Ma (MSWD=1.4; 8 data points).

374 Uranium oxides

375 The data for the different uranium oxides dated in this study are given in Table SM11. The majority of the U-Pb 376 isotopic ratios are discordant in the Wetherill Concordia diagram (Fig. 10), indicating that all the uranium oxides 377 suffered from post-crystallisation alteration. The least altered zones (i.e. richest in Pb) were selected for the 378 isotopic dating based on EPMA analyses and mapping (Fig. 6). All the uranium oxides have a contribution of 379 common lead, with ²⁰⁴Pb/²⁰⁶Pb ratios varying between 0.000001 and 0.001926. Sample WDD10-151 29.4 (U4) 380 has the highest common lead contribution, with ²⁰⁴Pb/²⁰⁶Pb ratios between 0.000763 and 0.0011327, followed by 381 samples JDD08-003 74.4B (U2, 204Pb/206Pb: 0.000002-0.001926), WDD07-2 55.9A and WDD07-2 55.9B (U4, ²⁰⁴Pb/²⁰⁶Pb: 0.000010–0.000171), WDD08-106 10.6 (U5, ²⁰⁴Pb/²⁰⁶Pb: 0.000030–0.000057) and WDD07-2 73.5B 382 (U6, ²⁰⁴Pb/²⁰⁶Pb: 0.000003–0.000122). All the analyses were corrected for common lead using the common lead 383 384 contribution calculated at their crystallisation age: 1030 Ma for JDD08-003 74.4B (U2), 730 Ma for WDD10-385 151 29.4 and WDD07-2 55.9A and WDD07-2 55.9B (U4), 550 Ma for WDD08-106 10.6 (U5), and 350 Ma for 386 WDD07-2 73.5B (U6).

The 54 analyses of the uranium oxides from sample JDD08-003 74.4B (U2) are moderately to strongly discordant and give an upper intercept at 1035 ± 57 Ma and a lower intercept at the origin, indicating a recent alteration event (Fig. 10a). The data are scattered with a MSWD of 76, with a majority of analyses with U/Pb ratios close to the origin.

The uranium oxides from samples WDD07-2 55.9A and WDD07-2 55.9B (U4) give a global upper intercept age at 661 ± 39 Ma (49 analyses, lower intercept: 12 ± 11 Ma) but the data are highly scattered (MSWD = 170) and present highly variable discordancy behaviour, from almost concordant to strongly discordant (Fig. 10b). When using the less discordant data (25 analyses), the data are less but still scattered (MSWD = 84), with an upper intercept age at 738 ± 44 Ma and a lower intercept at 79 ± 45 Ma.

- The 20 analyses of the uranium oxides from sample WDD10-151 29.4 (U4) are discordant but grouped (MSWD = 4.2) and give an upper intercept at 725 \pm 23 Ma when anchoring the lower intercept at 0 \pm 5 Ma (initial calculated lower intercept at -14 \pm 6 Ma, Fig. 10c).
- The 59 analyses of the uranium oxides from sample WDD08-106 10.6 (U5) are discordant but grouped (MSWD = 4.2) with an upper intercept at 554 ± 6 Ma when anchoring the lower intercept at 0 ± 5 Ma (initial calculated lower intercept at -16 ± 15 Ma, Fig. 10d).
- The 85 analyses of the uranium oxides from sample WDD07-2 73.5B (U6) are highly scattered (MSWD = 139) and present a variable discordancy degree, giving an upper intercept of 350 ± 21 Ma when anchoring the lower intercept at 0 ± 5 Ma (Fig. 10e).
- All the samples except two (WDD07 2 55.9A and WDD07-2 55.9B) are characterised by a lower intercept at almost 0 Ma, indicating that alteration of the uranium oxides is recent.
- 407

4.4. Chlorite composition and crystallisation temperatures

408 All the analysed chlorite grains have high iron contents (30.26-45.31 wt% FeO), low magnesium contents (2.01-11.99 wt% MgO), and plot along the Fe²⁺-Mg²⁺ chamosite-clinochlore substitution trend (Fig. 11a). Silica 409 content ranges from 21.14 to 27.44 wt% and Al between 16.18 and 22.15 wt% Al₂O₃. Titanium, Mn, Ca, Na and 410 411 K contents are consistently below detection limits except when chlorite is mixed with illite (Table SM12). 412 Chlorite has a chamositic composition (Fig. 11b, Polito et al. 2005a; this study). To estimate temperature, the 413 model developed by Bourdelle et al. (2013) was used because it was designed for diagenetic to low-grade metamorphic conditions and it avoids the problem of estimating the oxidation state of iron in chlorite. The 414 results are presented in Table SM12, where the "Linear T°" column corresponds to the basic linear model for 415 $T^{\circ}C < 350 \ ^{\circ}C$, the "Quadratic T° " is based on a quadratic equation used above 350 $^{\circ}C$, and the "Final T^{\circ}" 416 417 column gives the linear T° when it is below 350 °C and the quadratic one for temperatures above 350 °C. On 99 418 analyses, 13 range between 159 and 300 °C, most of the calculated temperatures are above 300 °C and 42 419 analyses could not be processed due to the sum of $Si + Ti + Al + Fe^{2+} + Mn + Mg + Ca + Na + K - 4$ (in *apfu*) 420 higher than 6. Other thermometers were tested such as the ones developed by Cathelineau (1988) and by Jowett 421 (1991) that also gave calculated temperatures higher than 300 °C.

422 **5. Discussion**

423 424

5.1. Timing of the U mineralisation in the Westmoreland area and its correlation to the geodynamic evolution of Australia

425 The paragenesis defined during this study is based on eight representative samples from the mineralised areas of the Redtree, Junnagunna and Huarabagoo U deposits and was combined with the results of Polito et al. (2005a), 426 427 because the latter paragenetic sequence takes into account a larger diversity of samples from barren, altered and 428 mineralised zones (Fig. 3). Some of their mineral assemblages were not observed in this study, mainly Fe-, Ti-429 oxides in the detrital minerals, dickite (D1) and pyrite (P1) linked to the ca. 1680 Ma diagenetic event. In the 430 present study, the dickite was not identified and the clay mineral replacing chlorite C1 was illite. A first generation of micron-sized grains of uranium oxide U1 (referred to as "uraninite" in Polito et al. 2005a) is 431 432 texturally associated with illite I2, hematite H2 and rutile R2. Polito et al. (2005a) described another generation 433 of uranium oxide (noted U3 in the combined sequence) that crystallised in voids or in veins, possibly associated 434 with pyrite (Py2) and galena. These two generations of uranium oxides were not observed in this study.

435 The U-Pb isotopic ages for the uranium oxides range from 1035 ± 57 Ma (sample JDD08-003 74.4B) to $350 \pm$ 21 Ma (WDD07-2 73.5B), with 4 distinct stages at ca. 1035 Ma, 730 Ma, 550 Ma and 350 Ma (Fig. 10). These 436 ages are younger than the oldest ages calculated for the Westmoreland area on very fine-grained intergranular 437 uraninite U1 inclusions (6–8µm) in hematite at Redtree, with $^{207}Pb/^{206}Pb$ ages of 1606 ± 80 Ma and 1655 ± 83 438 439 Ma from two analyses obtained by LA-HR-ICPMS (Polito et al. 2005a). Although the common lead contribution in U1 was not measured, this old age is younger than, but falls within error of the 1680 ± 21 Ma 40 Ar/ 39 Ar age 440 441 measured for diagenetic illite extracted from the Westmoreland Conglomerate (Polito et al. 2005a) and the 1685 442 \pm 65 Ma age measured for apatite in association with uranium oxides at the Junnagunna deposit (sample JDD08-443 03 74.4B; Fig. 9). Therefore, it can be proposed that a diagenetic or hydrothermal fluid event was active in the 444 area at ca. 1680 Ma, during which clay minerals, apatite and a first stage of uranium oxide crystallised. However, 445 as the U1 ages are younger than the age of the illite and apatite, and considering the associated errors, U1 could 446 also have formed shortly after this major fluid event, or at the end of it. From a geodynamic point of view, the 447 ca. 1680 Ma period in the Westmoreland area can be correlated to the onset of magmatism in the Warumpi 448 Province (Scrimgeour et al. 2005) and to a phase of rifting and intrusion of the 1678 Ma Carters Bore Rhyolite 449 and 1670 Ma syn-extensional Sybella Batholith in the Mount Isa Province (Gibson et al. 2016). This stage was 450 followed by at least five successive events of U mineralisation at ca. 1035 Ma, 870 Ma (Polito et al. 2005a), 730

451 Ma, 550 Ma and 350 Ma, the ages evaluated in this study correlating with the younger events of Polito et al. 452 (2005a, Fig. 10f). These events could either have remobilised pre-existing mineralisation or a new stock of 453 uranium could have been brought to the deposit site, and these two possibilities are discussed in section 5.2. 454 Hypotheses can relate these events to the geodynamic evolution of the McArthur Basin or Australia in a more 455 global view. The timing of the ca. 1035 Ma event corresponds to the 1085–1040 Ma Giles Event, which included a failed east-trending intracontinental rift in the western Musgrave Province and the extensive 1075 Ma 456 457 Warakurna Large Igneous Province (Wingate et al. 2004; Evins et al. 2010; Howard et al. 2011; Ahmad and Munson 2013). The ca. 870 Ma event corresponds to initiation of the Centralian Superbasin at about 840 Ma 458 459 related to extension, rifting and crustal sagging that may have been an early expression of the breakup of the 460 Neoproterozoic Rodinia Supercontinent (Lindsay 2002; Greene 2010). The 730 Ma phase corresponds to a 461 period of tectonic quiescence, with little evidence for major tectonic events (Ahmad and Munson 2013). As all 462 the other ages can be correlated with major tectonic events, it can be proposed that a same situation is valid for 463 730 Ma: a currently undefined geodynamic event favoured U mobility. The last two uraniferous events (ca. 550 464 and 350 Ma) take place when central Australia was affected by major intraplate tectonism, that dissected the 465 Centralian Superbasin, exhumed rocks from deep crustal levels, and resulted in large amplitude east-trending gravity anomalies in central Australia (Ahmad and Munson 2013). The first of these events, the Petermann 466 467 Orogeny, was focused in the Musgrave Province at 580–530 Ma and overlaps to the ca. 550 Ma event. Then, the 468 Alice Springs Orogeny was a major intraplate orogenic event that variably affected large areas of central 469 Australia from 450 to 300 Ma and resulted in the exhumation of the Arunta Region (Collins and Teyssier 1989; 470 Haines et al. 2001) which could be related to the ca. 350 Ma mineralisation. The temporal correlation between 471 the ages of the different uranium oxides in the Westmoreland area and successive geodynamic events having 472 affected Australia demonstrates that these events favoured fluid circulations in the McArthur Basin and related 473 basement, and uranium mobility during more than 1.3 Ga. However, none of the uraninite generations identified 474 so far record the Isan Orogeny (1640-1490 Ma, with the peak around ca 1590 Ma, Murphy et al. 2011) that 475 affected the nearby Mount Isa Basin.

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5.2. Geochemistry of the successive U mineralisation: changing physico-chemical conditions in the Westmoreland area

The chemical composition of the ca. 1680 Ma uranium oxide U1 ("uraninite" in Polito et al. 2005a) is unknown, precluding comparison with later generations. The chemical compositions of the different generations of uranium oxides identified during this study are specific for each sample in terms of chondrite-normalised REE

481 patterns and minor to trace element concentrations (Figs. 7 and 8). Such results indicate at first order that the 482 physico-chemical conditions for the formation of these different uranium oxides were different (Mercadier et al. 483 2011b; Frimmel et al. 2014; Alexandre et al. 2015). Although the origin of this variability is not precisely 484 constrained (i.e., the chemical composition of the mineralising fluids, the source(s) of the fluids or of the metals, 485 or the pressure-temperature conditions; see Mercadier et al. 2011b), such variability suggests that the physicochemical conditions within the studied deposits have not been constant through time. The successive generations 486 487 of uranium oxides do not show any U-Pb isotopic signatures that indicate that they have been altered and/or remobilised by later fluid events; all the lower intercepts of the Discordia lines are younger than 100 Ma (Fig. 488 489 10). When combined with the variability in the geochemical signatures of the different generations of uranium 490 oxides, these results do not allow conclusively define if U in the Westmoreland area was deposited at ca. 1680 491 Ma then locally remobilised during five fluid events under different conditions, or if each successive fluid event 492 brought a new batch of uranium and related metals to the deposits. The latter option is preferred, as 493 remobilisation of primary uranium in Canadian unconformity-related deposits leads to the formation of uranium 494 oxides in which some of the geochemical markers, like REE patterns, are preserved across the generations 495 (Mercadier et al. 2011a; Martz et al. 2019). The significant variations in the geochemical signatures of the 496 uranium oxides probably indicate new U inputs for each successive generation. In addition to unique chemical 497 signatures, each U-forming event in the Westmoreland area is characterised by a specific mineral assemblage 498 that reflects different formation conditions, or degrees of post-ore alteration (Polito et al. 2011). The U 499 mineralisation currently observed in the Westmoreland area is thus the result of a protracted history spanning at 500 least 1.3 Ga (1.68 to 0.35 Ga) of fluid circulations for which the physico-chemical conditions have varied, in 501 response to specific geodynamic events.

502 503

5.3. Chemical composition of the clay minerals and related temperature of crystallisation

Published data on C1 chlorite from the Westmoreland U deposits indicate that this unique generation of chlorite is Fe-dominated but has variable Al contents and Mg/Fe ratios depending on their location (Westmoreland Conglomerate and Seigal Volcanics), plotting between the chamosite, clinochlore and Mg-amesite compositions (Fig. 11b; Polito et al. 2005a). The variation in chemical composition leads to variable range of calculated temperatures of formation from 196 °C to 364 °C with the Cathelineau and Nieva (1985) and Cathelineau (1988) methods (Polito et al. 2005a); and from 110 °C to 406 °C with the Bourdelle et al. (2013) method used for this study. All the new analysed C1 chlorite, regardless of the location or deposit, lie in the chamosite field with

511 related temperature of formation mostly above 300 °C (Fig. 11, Table SM12). Polito et al. (2005a) proposed that 512 the Fe-rich composition of the chlorite yielding the highest temperatures of crystallisation (364 °C) was linked to 513 the proximity of the Seigal Volcanics that provided excess Fe during crystallisation of the chlorite. In addition, the Fe²⁺ in the Fe-rich chlorite adjacent to the mafic dykes and volcanic or mafic rocks themselves may have 514 515 been the chemical reductant that reduced the hexavalent uranium to precipitate uraninite (Wall 2006; International Atomic Energy Agency 2018a). The distance between our samples and the Seigal Volcanics is 516 517 unknown, but the same process could have resulted in the high Fe contents and the high temperature of 518 crystallisation. Temperature estimates from fluid inclusions in the Redtree, Junnagunna and Huarabagoo deposits 519 are lacking, but fluid inclusions were analysed from two related U deposits (Eva Mine and Jackson Pit) in the 520 basement below the Westmoreland Conglomerate (Mernagh and Wygralak 2011). Based on their description and 521 relation to other minerals, we assume that the quartz veins in which the fluid inclusions were studied are genetically comparable to the Q₂ drusy vein described by Polito et al. (2005a) which are co-genetic or post-date 522 C1 chlorite. Temperatures of homogenisation for the different types of fluid inclusions are between 99 to 380 °C, 523 524 with two populations: 99-270 °C and 190-380 °C (Mernagh and Wygralak 2011). Assuming that data obtained 525 by Mernagh and Wygralak (2011) can be extrapolated to the Redtree, Junnagunna and Huarabagoo deposits, both chlorite and fluid inclusions indicate abnormally high thermal conditions (T>300 °C) during diagenesis at 526 527 ca. 1680 Ma and reflect three possible origins. Firstly, the high temperatures could be due to a high regional 528 geothermal gradient. Secondly, the fluids could be heated by the Seigal Volcanics whose age is not constrained 529 but could be correlated to ca. 1780 Ma volcanic rocks (Mernagh and Wygralak 2011). Thirdly, fluids heated in 530 the basement could have been injected into the basin. These high thermal conditions were probably short-lived 531 because temperature estimates for illite I1 and illite I2, which post-date chlorite C1 and is co-genetic of U1 532 uranium stage at 1655 Ma, respectively, range from 150 °C to 250 °C (Polito et al. 2005a).

533 The temperature estimates and ages for the first U mineralising event are however loosely-constrained and the 534 possibility that this U stage formed at ca. 1680 Ma in direct relation with the emplacement of the Seigal 535 Volcanics has to be reconsidered, in particular due to spatial correlation between the U mineralisation and Seigal 536 Volcanics (Fig. 2) and the possible older age of the volcanic rocks. Dyke emplacement driving formation of 537 basin-hosted U mineralisation in Proterozoic basins such as McArthur Basin has been proposed for the Matoush 538 and Camie River U deposits in the Otish Basin, Canada (Alexandre et al. 2015; Lesbros-Piat-Desvial et al. 2017), with temperatures of formation exceeding 300 °C. Such implications should be studied in more detail for 539 540 the Westmoreland area.

541 No information regarding the type of alteration, nature of the fluids or temperatures can be currently provided for 542 the five generations of uranium oxides postdating the ca. 1680 Ma U1 of Polito et al. (2005a), as these generations are not genetically associated with clay minerals, and fluid inclusions study in quartz Q3 associated 543 544 with uraninite U4 are underway. A first-order assessment could be proposed based the REE contents measured in 545 the uranium oxides (Mercadier et al. 2011b; Frimmel et al. 2014; Alexandre et al. 2015; Martz et al. 2019), but must be considered with caution. As \sum REE for U2 (sample JDD08-003 74.4B) formed at ca. 1030 Ma is above 1 546 wt% (Fig. 6e), a temperature estimate > 350 °C is proposed, whereas the following generations of uranium 547 oxides were probably formed at a temperature < 350 °C. 548

549 550

5.4.

unconformity-related U deposits

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5.4.1. Comparison at the scale of the McArthur Basin: Westmoreland area vs ARUF

Comparison between the uranium deposits in the Westmoreland area and

The Alligator Rivers Uranium Field (ARUF) of the McArthur Basin hosts numerous unconformity-related U deposits such as Ranger, Nabarlek, Jabiluka and Koongarra (Polito et al. 2004, 2005b, 2011; Jefferson et al. 2007; Cuney and Kyser 2015; Skirrow et al. 2016). As these deposits and those in the Westmoreland area share similar first-order settings and are all associated with the McArthur Basin, it is critical to establish if they could be linked to the same ore-forming events and/or ore-forming processes.

Deposits in the ARUF are located exclusively in the basement rocks, near the unconformity between an Archean to Paleoproterozoic basement complex and the McArthur Basin, are structurally-controlled, and have grades mostly above 0.1% U₃O₈ (Rawlings 1999; Jackson et al. 2000). Deposits in the Westmoreland area are structurally-controlled, spatially-related to volcanic dykes and sills in the basin and have lower grades (Hein 2002; Polito et al. 2005a; Skirrow et al. 2016; Laramide Resources Ltd 2019).

562 The ARUF and the Westmoreland area were affected by a major diagenetic/hydrothermal fluid circulation event 563 at ca. 1680–1600 Ma (Fig. 12). This episode is recorded in multiple isotopic ages measured on illite, apatite and 564 uranium oxides for the two regions (Maas 1989; Polito et al. 2004, 2005a, b; Clauer et al. 2015; Skirrow et al. 2016). The fluids of the diagenetic/hydrothermal event in the ARUF and Westmoreland area at 1680-1600 Ma 565 were basin-derived brines and low-salinity fluids (Ypma and Fuzikawa 1980; Wilde et al. 1989; Derome et al. 566 2003, 2007; Polito et al. 2004, 2005b, 2006; Mernagh and Wygralak 2011) . The isotopic and fluid inclusions 567 data at Westmoreland show the involvement of NaCl-rich and CaCl₂-rich brines derived from evaporated 568 569 seawater, comparable to the two brines related to formation of the unconformity-related deposits in the ARUF 570 (Derome et al. 2007). The mixing between low-salinity fluids and brines is a key process for ore deposition in 571 both areas (e.g. Derome et al., 2003, 2007; Mernagh and Wygralak, 2011). Although contemporaneous and 572 related to the same types of fluids, the fluids at ca. 1680 Ma in the ARUF and Westmoreland area are physico-573 chemically different based on the differences in the mineralogy and chemistry of the minerals which crystallised 574 at that time in both areas. The most recent studies on unconformity-related U deposits from the ARUF, in particular for the Ranger U deposit, show significant differences with the deposits in the Westmoreland area in 575 576 terms of the composition of chlorite (Skirrow et al. 2016; Gigon et al. 2019) and the presence and composition of hydrothermal tourmaline (Gigon et al. 2019). 577

The unconformity-related U deposits in the ARUF at ca. 1680-1600 Ma formed through Mg- and B-578 579 metasomatism (Fisher et al. 2013; Skirrow et al. 2016; Gigon et al. 2019), but such metasomatism is lacking in 580 the Westmoreland area. The absence of hydrothermal Mg-tourmaline with the uranium oxides in the 581 Westmoreland area is a first striking difference, as this mineral is a pathfinder for unconformity-related U 582 deposits in the ARUF (Skirrow et al. 2016; Gigon et al. 2019). The second difference is the chlorite composition: 583 the chlorite in the Westmoreland area are dominantly chamosite (Fe-rich), compared to the clinoclore to sudoite 584 composition (Mg-rich) of the diagenetic/hydrothermal chlorite in the unconformity-related deposits in the ARUF 585 (Fig. 11b, Polito et al. 2004, 2005b; Beaufort et al. 2005; Gigon et al. 2019). As a consequence, the 586 diagenetic/hydrothermal chlorite composition for the ARUF yields temperatures between 101 °C and 163 °C, with mean temperature of 128 °C and median temperature of 124 °C (Gigon et al. 2019), considerably lower 587 588 than the > 300 °C calculated in this study.

589 The present study identifies for the first time multiple U generations between 1035 and 350 Ma in three deposits 590 of the Westmoreland area. The succession of fluid events also characterises the unconformity-related U deposits 591 from the ARUF, with multiple ages ranging from 1680 Ma to Present measured on both uranium oxides and illite 592 (Hills and Richards 1976; Ludwig et al. 1987; Maas 1989; Polito et al. 2004, 2005b; Ahmad and Munson 2013; 593 Clauer et al. 2015; Skirrow et al. 2016). These post-1680 Ma ages in the ARUF mainly fall between 1600 and 594 1300 Ma for the clay minerals and some uranium oxides, but are dominantly younger than 900 Ma for the 595 uranium oxides. Based on the available datasets, the post-1680 Ma ages measured for the ARUF do not totally 596 correlate with the five post-1680 Ma ages measured at the Westmoreland area, which could reflect that the fluid 597 events in the two areas are partially disconnected in time from 1680 Ma to Present. The nature of the fluid(s) and 598 physico-chemical conditions of these events in the ARUF are not well constrained.

599 It is currently not possible to link the physico-chemical conditions for the U1 generation in the Westmoreland 600 area with the primary uraninite in the ARUF as no analyses were done on U1. Both primary mineralisation could 601 have been formed in a similar system, but under higher temperature conditions in the Westmoreland area. In this 602 area, the ultimate origin of the different generations and the physico-chemical conditions prevailing between 603 1035 and 350 Ma also remain to be constrained. The chondrite-normalised REE patterns of uranium oxides 604 which are diagnostic features of deposit types, differ significantly between the ARUF and the Westmoreland 605 area. The different generations of uranium oxides in the Westmoreland area do not exhibit the "bell-shape" (i.e. 606 high concentrations of Tb and Dy) typical of unconformity-related U deposits from the ARUF such as Nabarlek 607 and Koongarra (Mercadier et al. 2011b). Even if some post-crystallisation modification of the REE patterns may 608 have contributed to the LREE enrichment (as described in the Athabasca Basin by Mercadier et al. 2011a and 609 Martz et al. 2019), such phenomenon is not sufficient to explain the major differences of the REE patterns 610 between the two areas. The uranium oxides from the Westmoreland area have significantly higher contents in 611 common lead (Table SM11) when compared to typical uranium oxides from unconformity-related uranium 612 oxides (Skirrow et al. 2016; Martz et al. 2019), which is also an indicator of differences in their conditions of 613 formation. It can be concluded that the formation conditions of the different U generations in the Westmoreland 614 area did not correspond to those known for unconformity-related U deposits in the ARUF.

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5.4.2. Comparison between U deposits in the Westmoreland area and U deposits in Canadian Proterozoic basins

617 Several Paleo- to Mesoproterozoic basins in Canada host U mineralisation: the Athabasca Basin (Saskatchewan),
618 the Thelon Basin (Nunavut) and the Otish Basin (Québec).

619 The U mineralisation in the Athabasca Basin is of unconformity-related type, like the one in the ARUF 620 (Jefferson et al. 2007; Cuney and Kyser 2015). The ca. 1680-1600 Ma diagenetic/hydrothermal episode defined 621 in the Westmoreland area is documented in the Thelon Basin, where apatite is dated at 1667 ± 5.1 Ma (Davis et 622 al. 2011). In the Athabasca Basin, this fluid event has been recorded by TIMS analyses of hydrothermal zircon 623 tips (uranium-rich, thorium-depleted) in a leucogranite sampled 200 meters east of the McArthur River highgrade U deposit with ages from 1680 to 1550 Ma. This was interpreted as the approximate timing of the 624 625 thermotectonic event(s) and associated fluids that produced the new U-rich zircon (Annesley et al. 2007). In 626 addition with the 1650-1700 Ma diagenetic apatite age (Cumming et al. 1987), such results confirm a global 627 fluid event at 1680–1600 Ma affecting the Athabasca, Thelon and McArthur basins which could have belong to 628 the same super basin at that time (Furlanetto et al. 2016). No U mineralisation has however been dated at 1680

Ma for the Athabasca and Thelon basins, with maximum ages at ca. 1590 Ma (Alexandre et al. 2009) and ca. 1500 Ma (Sharpe et al. 2015) respectively. Recent data on U mineralisation in the Thelon Basin indicate that these deposits had a polyphased history over hundreds of Ma in which unconformity-related U mineralisation formed from ca. 1500 Ma, followed by low-T and meteoric-related redox fronts (Sharpe et al. 2015; Chi et al. 2017; Shabaga et al. 2017; Grare et al. 2020).

634 In the Otish Basin, host of the Matoush and Camie River U deposits, no significant hydrothermal activity is 635 known at ca. 1680 Ma. This basin formed earlier than the Athabasca and Thelon basins with a minimum deposition age of 2172 to 2162 Ma (Hamilton and Buchan 2016; Milidragovic et al. 2016). The oldest reported 636 age of fluid circulation and mineralisation is ca. 1725 Ma for the Camie River U deposit (Beyer et al. 2012; 637 638 Lesbros-Piat-Desvial et al. 2017). This ca. 1725 Ma U mineralisation was initially interpreted to be 639 unconformity-related type (Bever et al. 2012), but recent research contradicts this interpretation because of a 640 Paleoproterozoic, higher-temperature hydrothermal event, whose origin remains speculative (Lesbros-Piat-641 Desvial et al. 2017). The timing of formation of the Matoush deposit is loosely constrained (1695 \pm 110 Ma; Alexandre et al. 2015), and its main characteristics (spatial association with a dyke, alteration mineralogy, 642 643 association between uraninite and eskolaite) do not support classification as an unconformity-related type 644 deposit, although the U-bearing fluids were oxidising basinal fluids (Alexandre et al. 2015), like for the 645 Westmoreland area.

646 The chemical composition of uranium oxides analysed in this study records major differences for minor and 647 trace elements (including REEs) compared to typical unconformity-related U deposits from the Athabasca (Mercadier et al. 2011b; Alexandre et al. 2016; Martz et al. 2019) or Thelon basins (Grare et al. 2020). This is 648 649 particularly visible when comparing the chondrite-normalised REE patterns, as the different generations of 650 uranium oxides from the Westmoreland area do not have the typical bell-shape pattern of the unconformityrelated U deposits (Fig. 8). Differences are also visible for numerous minor and trace elements like As, W, Ba 651 652 and V (Fig. 7c and d) compared with McArthur River and Cigar Lake U deposits of the Athabasca Basin 653 (Alexandre et al. 2015; Martz et al. 2019). The majority of the minor and trace elements have significantly 654 higher concentrations in the uranium oxides from the Westmoreland area, which could indicate a higher temperature of formation and/or different sources of metals. The complete geochemistry of the uranium oxides 655 656 from the Otish Basin is lacking, but the REE patterns of the ca. 1725 Ma uranium oxides from the Camie River 657 are rather similar to those measured for different generations of uranium oxide from the Westmoreland area (Fig. 658 8). Such results allow consideration that the physico-chemical conditions for the formation of the U

mineralisation in the Westmoreland area were probably more similar to those of Matoush or Camie River U deposits of the Otish Basin than those of the Athabasca and Thelon basins. Temperatures estimates from the Matoush (~300 ±50 °C, based on illite and chlorite crystal chemistry, Alexandre et al. 2015) and Camie River deposits (275 to 300 °C, Beyer et al. 2012 and 320 °C, Lesbros-Piat-Desvial et al. 2017) are comparable to the Westmoreland area, but significantly higher than values reported from the Athabasca and Thelon basins (e.g. Kotzer and Kyser 1995; Richard et al. 2016; Chi et al. 2017; Grare et al. 2020).

665 Other similarities with the U deposits from the Otish Basin are the lack of Mg-rich minerals (sudoite, Mg-foitite) or alumino-phosphate-sulfate minerals, which are markers of unconformity-related U deposits. The Matoush and 666 Camie River deposits are spatially related to dykes (Beyer et al., 2012; Alexandre et al. 2015; Lesbros-Piat-667 668 Desvial et al. 2017), which played a role as the reductant of uranium, because it was a source of Cr and Fe at Matoush (Alexandre et al. 2015). An equivalent process was proposed for the Westmoreland area with the 669 implication of iron from the dykes as reductant (Polito et al. 2005a). The mafic activity in the Otish Basin and in 670 671 the southern McArthur Basin (Westmoreland area) could thus have favoured the formation of atypical U 672 deposits, although linked to the same types of fluids as unconformity-related U deposits.

673 6. Conclusions

The new data provided on U mineralised intercepts from three deposits of the Westmoreland area (Queensland 674 675 and Northern Territory, Australia) indicate that this region of the McArthur Basin experienced several episodes of fluid circulation between ca. 1680 Ma to Present, which document a protracted mobility of uranium over more 676 677 than 1.3 Ga. Based on published and new data, six different events of U mobility were active at ca. 1680 Ma, 1035 Ma, 850 Ma, 730 Ma, 550 Ma and 350 Ma. The ca. 1680 Ma mineralisation formed due to the circulation 678 679 and mixing of NaCl-rich, CaCl₂-rich basin-derived brines and low-salinity fluids. The timing and type of fluids 680 involved were comparable to those of the unconformity-related U deposits in the ARUF to the North of the 681 McArthur Basin. The physico-chemical conditions for the formation of these contemporaneous uranium mineralisations are different between the two regions, with higher chlorite temperature (> 300 °C) for the 682 683 Westmoreland area and absence of Mg- and B-metasomatism typical of unconformity-related U deposits. 684 Although the precise chemical composition of the uranium oxides formed at ca. 1680 Ma has not been measured, 685 all the other U generations found in the Westmoreland area are chemically different from the uranium oxides 686 found in unconformity-related U deposits. The Westmoreland area samples contain higher contents of numerous 687 minor and trace elements, like REEs, which supports a higher temperature of crystallisation. If the timing and the types of fluids between the two parts of the basin were similar at ca. 1680 Ma, fluids in the Westmoreland area 688 689 were at higher temperatures, driven by a high regional geothermal gradient controlled by volcanism (Seigal 690 Volcanics), or injection of fluids heated in the basement into the basin. This elevated thermal regime may have 691 triggered fluid circulation in the Westmoreland area, with the dykes providing the structural and chemical traps 692 that focused the uranium-bearing brines and favoured U deposition at ca. 1680 Ma. Such conditions appear 693 similar to those known for the formation of U deposits in the Proterozoic Otish Basin (Québec, Canada) at ca. 694 1725 Ma. The new data proposed in this study indicate that the geochemical signature of the successive uranium 695 oxides from the Westmorland area are poorly compatible with uranium oxides formed in unconformity-related U 696 deposits, as confirmed by the difference in mineral association. It is therefore considered as unlikely to find 697 unconformity-related U deposits in this area. The significant changes in the chemical compositions of the different generations of uranium oxides are in favour of changing conditions between 1680 and 350 Ma. 698 699 Although the precise physico-chemical are loosely defined, the signatures observed in the Westmorland area for 700 the first U stages are closer to those proposed for U deposits from the Otish Basin than those for the Athabasca 701 or Thelon basins.

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703 Acknowledgements

704 The Northern Territory Geological Survey provided logistical support for the sampling mission. Laramide 705 Resources Ltd. provided authorisation, logistical support and guidance for sampling on the Westmoreland 706 exploration camp (especially Adrian Buer and Evan Hughes). Alexandre Flammang and John Moine 707 (GeoRessources Lab) as well as Thin Section Lab company staff are acknowledged for careful thin section 708 preparation. Chantal Peiffert, Andreï Lecomte and Olivier Rouer (GeoRessources Lab) are acknowledged for 709 support in LA-ICP-MS, SEM and EPMA analyses respectively. This research was funded by (i) a French 710 Ministry of Higher Education and Research PhD salary Grant to J.G., (ii) a Région Lorraine-FEDER grant to 711 J.M: « Rôle des phases fluides dans la distribution spatiale des ressources métalliques dans les bassins 712 sédimentaires paléoproterozoiques australiens », (iii) OSU OTELo grants to J.M. : « Conditions de transport des 713 métaux dans un mégabassin protérozoïque » and A.R.: « Transferts de fluides et métaux dans le bassin de 714 McArthur (Australie) » and (iv) a CNRS-INSU-CESSUR grant to J.M. « Transferts des fluides et métaux dans

- 715 les méga-bassins paléoprotérozoïques ». The French National Agency through the national program
- 716 «Investissements d'avenir» of the Labex Ressources 21 (reference ANR-10-LABX-21-RESSOURCES21)
- 717 partly funded the last developments of the SIMS lab. The comments of David Quirt and an anonymous reviewer
- 718 greatly helped improving the manuscript. Mostafa Fayek is warmly thanked for careful editorial handling.
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 SIMS U-Pb dating, in-situ Hf-O isotopes, and tectonic implications. Journal of Asian Earth Sciences
 151:101–111
- 1000
- 1001
- 1002 Fig. 1 Schematic geological map of the study area, Northern Territory and Queensland, Australia. a Location of the
- 1003 McArthur and South Nicholson basins and Murphy Inlier in Australia. **b** Zoom indicating the location of the Alligator Rivers
- 1004 Uranium Field (ARUF) and the Westmoreland area (black rectangle). c Simplified geological map around the studied
- 1005 deposits of the Westmoreland area. After Ahmad et al. (2013).

Fig. 2 Conceptual cross-section showing the position of the Junnagunna, Redtree, Huarabagoo and Long Pocket deposits in
the stratigraphic sequence of the McArthur Basin, Australia (compilation after Fuchs and Schindlmayer 1981; Schindlmayr
and Beerbaum 1986; International Atomic Energy Agency 2018a, b) and the approximate locations of the studied samples.

1010

Fig. 3 Paragenesis of the Redtree, Junnagunna and Huarabagoo deposits (Westmoreland area, Australia); compiled from Polito et al. (2005a) and this study. Ages are based on zircon U-Pb ages (Page et al. 2000) for the detrital minerals, U-Pb, $^{207}Pb/^{204}Pb$ and $^{40}Ar/^{39}Ar$ ages from Polito et al. (2005a) and U-Pb and Pb/Pb ages on apatite and uraninite from this study. Italic: data from the paragenesis presented by Polito et al. (2005a); regular: mineral sequence observed in this study; bold: mineral sequence observed in this study and by Polito et al. (2005a). The temperatures estimates are from chlorite and illite thermometry (Polito et al., 2005a; this study) and from the Σ REE (this study).

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Fig. 4 Photomicrographs and backscattered electron image (d) illustrating the diagenetic sequence for mineralized intercepts
of the Redtree, Junnagunna and Huarabagoo U deposits (Westmoreland area, Australia). a Q0 detrital quartz grains coated by
hematite (H1), and quartz overgrowths Q1, sample WDD10-151 27.9B. b Quartz partial dissolution (Q⁻) after Q1, sample
JDD08-003 74.4A. c Rosette of chlorite C1 after partial dissolution of Q0 and Q1, sample WDD07-2 51.7A. d Diagenetic
rutile R1 in a matrix formed by uraninite U4, sample WDD07-2 73.5B. e Illite alteration on chlorite C1, sample WDD9-128
53.3B. f Quartz Q2 after quartz Q1, sample WDD07-2 55.9B.

1024

Fig. 5 Backscattered electron image images of uraninite mineral assemblages and location of the analyses from the Redtree,
Junnagunna and Huarabagoo U deposits (Westmoreland area, Australia). For the location of the analyses: purple circle = LAICP-MS; blue cross = SIMS, dark green triangle = EPMA. a Uraninite U2 as matrix around apatite grains (Ap), sample
JDD08-003 74.4B. b Uraninite U4 with altered zones, sample WDD07-2 55.9A. c Uraninite U4 in vein associated with
chalcopyrite and pyrite, sample WDD10-151 29.4. d Uraninite U6 associated with clay minerals, sample WDD07-2 73.5B.

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Fig 6 EPMA maps of distribution of U, Pb, Ca, Fe and Si in uraninite for samples WDD07-2 55.9A (top) and WDD07-2
73.5B (bottom, Westmoreland area, Australia). The U- and Pb-rich zones correlate with Ca- and Si-poor zones. The element
concentration values (in wt%) are estimated from *in-situ* spot EPMA analyses.

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Fig. 7 Chemical compositions of uraninite generations from the Redtree, Junnagunna and Huarabagoo U deposits
(Westmoreland area, Australia). a Pb vs Si + Ca + Fe. b Binary diagram of As vs Nb (in ppm). c Binary diagram of As vs W

1037 (in ppm). d Binary diagram of V vs Ba (in ppm). e Rare Earth Element abundance (ΣREE) vs. REE fractionation
1038 ((ΣLREE/ΣHREE)_N) relative to published values from Mercadier et al. (2011b), Alexandre et al. (2016) and Martz et al.
1039 (2019). REE fractionation normalised to chondrite values of Anders and Grevesse (1989).

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1041 Fig. 8 Chondrite-normalised REE patterns of the different uraninite generations from the Redtree, Junnagunna and 1042 Huarabagoo U deposits (Westmoreland area, Australia). Each generation exhibits a unique REE pattern indicating different 1043 chemical environments, as previously observed for the other minor and trace elements (Fig. 7). The C1 Chondrite 1044 normalization is after Sun and McDonough (1989). a U2 generation. Sample JDD08-003 74.4B. b U4 generation. The LREE 1045 enrichment indicated by the arrow corresponds to alteration indicated by Pb loss. The numbers indicated to the left of the 1046 diagram corresponds to the LA-ICP-MS spots presented in Fig. 5. Sample WDD07-2 55.9A. c U4 generation. Sample 1047 WDD10-151 29.4. d U5 generation. Sample WDD08-106 10.6. e U6 generation. Sample WDD07-2 73.5B. f Uraninite data 1048 from unconformity-related basement-hosted deposit in the ARUF (Mercadier et al. 2011b) and the Camie River deposit, 1049 Otish Basin, Canada (Lesbros-Piat-Desvial et al. 2017).

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Fig. 9 ²³⁸U/²⁰⁶Pb and ²⁰⁷Pb/²⁰⁶Pb ratios measured and related age on diagenetic apatite from the sample JDD08-003 74.4B
 (Junnagunna deposit, Westmoreland area, Australia). a Detrital quartz Q0 and diagenetic apatite in a uraninite U2 matrix
 (BSE image). b Tera–Wasserburg Concordia diagram.

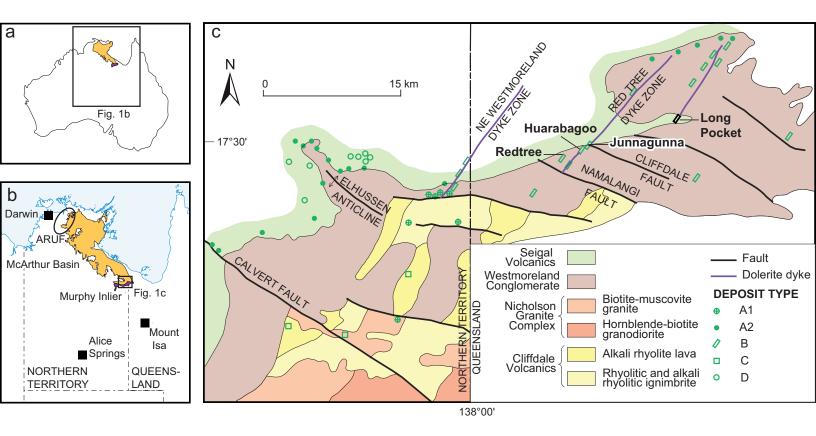
1054

Fig. 10 U-Pb Concordia diagrams from in-situ isotopic analysis by SIMS of uranium oxides from the Redtree, Junnagunna and Huarabagoo U deposits (Westmoreland area, Australia). Plots were constructed using ISOPLOT (Ludwig 2007) from the isotope ratios presented in Table SM11. **a** Sample JDD08-003 74.4B. **b** The black ellipses are the data obtained on a single uraninite and the grey ellipses are additional data from the same depth in the same drillcore. Samples WDD07-2 55.9A and WDD07-2 55.9B. **c** Sample WDD10-151 29.4 anchored at 0 ±5 Ma. **d** Sample WDD08-106 10.6 anchored at 0 ±5 Ma. **e** Sample WDD07-2 73.5B anchored at 0 ±5 Ma. **f** All data from this study (open circles, anchored at 0 ±5 Ma) and from Polito et al. (2005a, pink).

1062

Fig. 11 Chlorite composition and nomenclature for the Redtree, Junnagunna and Huarabagoo U deposits (Westmoreland area, Australia). **a** Fe-Mg diagram indicating that all Fe atoms are in the Fe²⁺ state in the ferromagnesian chlorite. **b** Classification diagram after Plissart et al. (2009). **c** Al^{VI}-Al^{IV} diagram for analysed chlorites. Structural formulae are calculated on the basis of 14 oxygen. **d** Classification diagram for rock-forming chlorites according to Zane and Weiss (1998). Compositions of chlorite from the Westmoreland area (this study; Polito et al., 2005a) are plotted together with 1068 chlorite from deposits of the ARUF (Polito et al. 2004, 2005b; Beaufort et al. 2005; Gigon et al. 2019), the Athabasca Basin
1069 (Kotzer and Kyser 1995; Billault et al. 2002; Martz 2017) and the Thelon Basin (Grare et al. 2020) for comparison. For data
1070 from the ARUF and the Canadian basins, the plotted data corresponds to the mean compositions for each sample or
1071 generation.

- 1073 Fig. 12 Synthesis of the alteration and mineralisation ages obtained in the ARUF and in the Westmoreland area of the
- 1074 McArthur Basin (Australia). 1: Hills and Richards (1976). 2: Clauer et al. (2015). 3: Ludwig et al. (1987). 4: Skirrow et al.
- 1075 (2016). 5: Polito et al. (2004b). 6: Polito et al. (2005b). 7: Maas (1989). 8: Polito et al. (2005a). 9: this study.



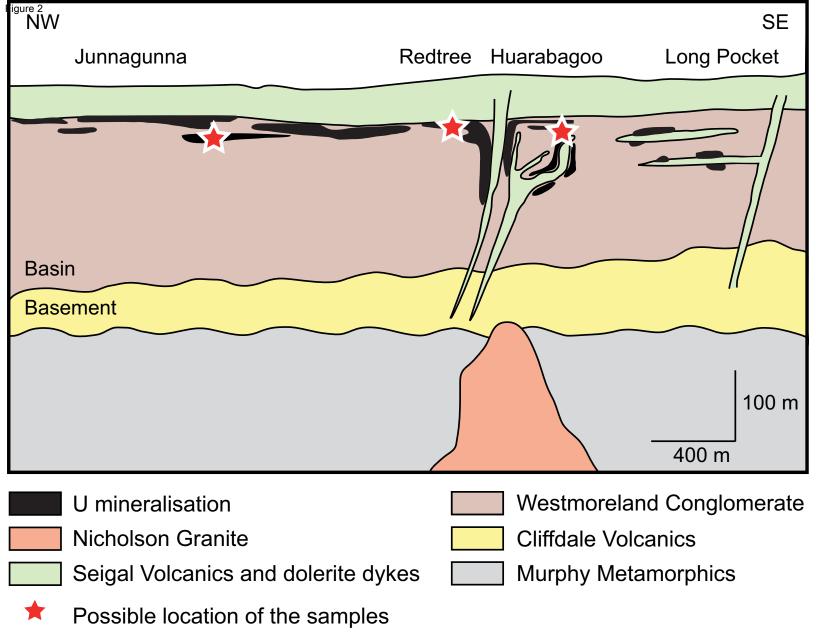
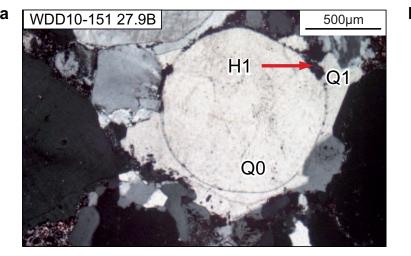
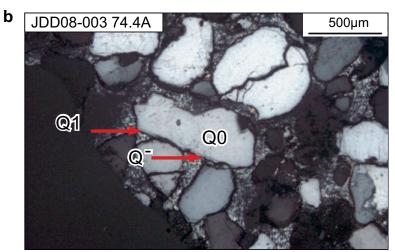
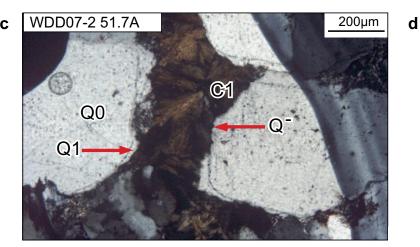


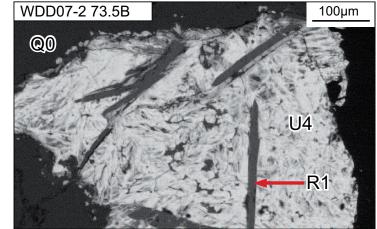
Figure 3

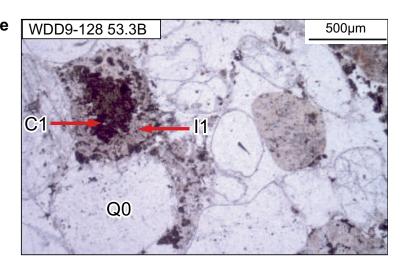
Characteristics	Detrital minerals	Early diagenesis	Diag	enesis	First stage uraninite	Other U-forming stages					
Age / timing	ca. 1800 <i>-</i> ca. 1750 Ma	ca. 1750 <i>-</i> ca. 1680 Ma	ca. 1680 Ma		1655 Ma	ca. 1030 Ma	ca. 870 Ma	ca. 730 Ma	ca. 550 Ma	ca. 350 Ma	
Est. temperature			> 300 °C	150- 250 °C	150- 250 °C	> 350 °C	< 350 °C	< 350 °C	< 350 °C	< 350 °C	
Quartz Q0											
Feldspar											
Fe-, Mg-silicates											
Fe-, Ti-oxides											
Hematite H1											
Quartz Q1											
Quartz dissolution			-								
Chlorite C1											
Rutile R1											
Dickite D1											
Illite I1											
Quartz Q2											
Apatite											
Pyrite P1											
Uraninite U1											
Hematite H2											
Illite I2											
Rutile R2											
Uraninite U2 JDD08-003 74.4B											
Uraninite U3											
Pyrite P2											
Galena											
Uraninite U4 WDD07-2 55.9											
WDD07-2 55.9 WDD10-151 29.4											
Pyrite P3											
Chalcopyrite											
Quartz Q3											
Uraninite U5 WDD08-106 10.6											
Uraninite U6 WDD07-2 73.5B											

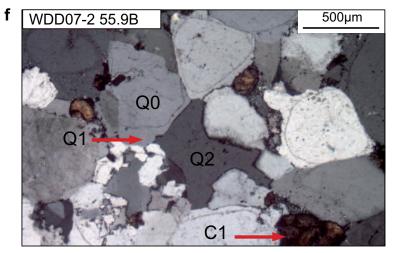


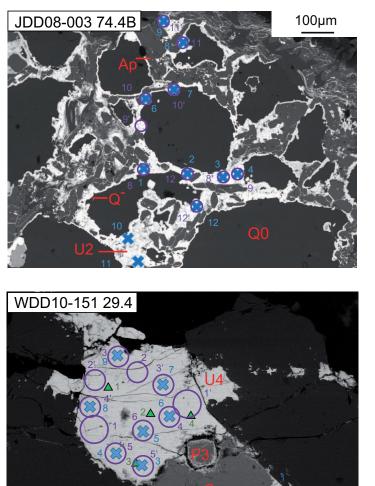


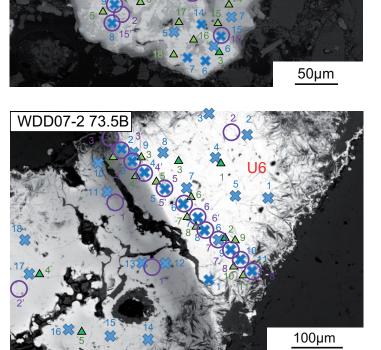












WDD07-2 55.9A

Δ

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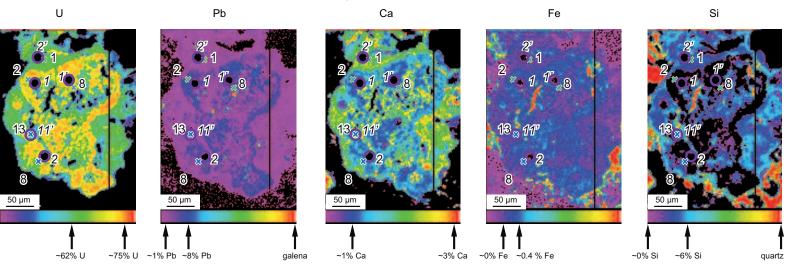
С

50µm

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Pb

Sample WDD07-2 55.9A

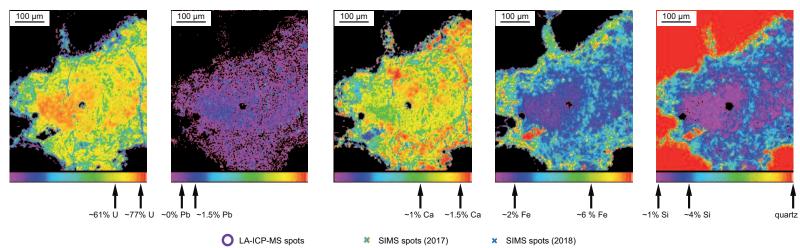


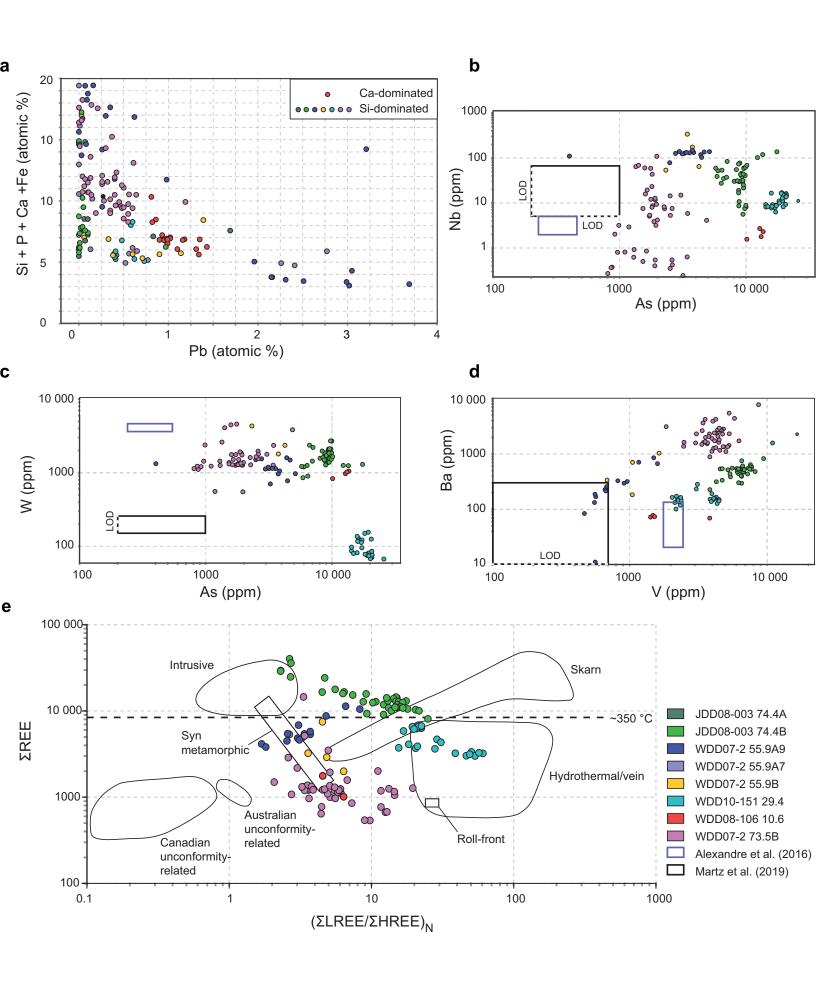
Sample WDD07-2 73.5B

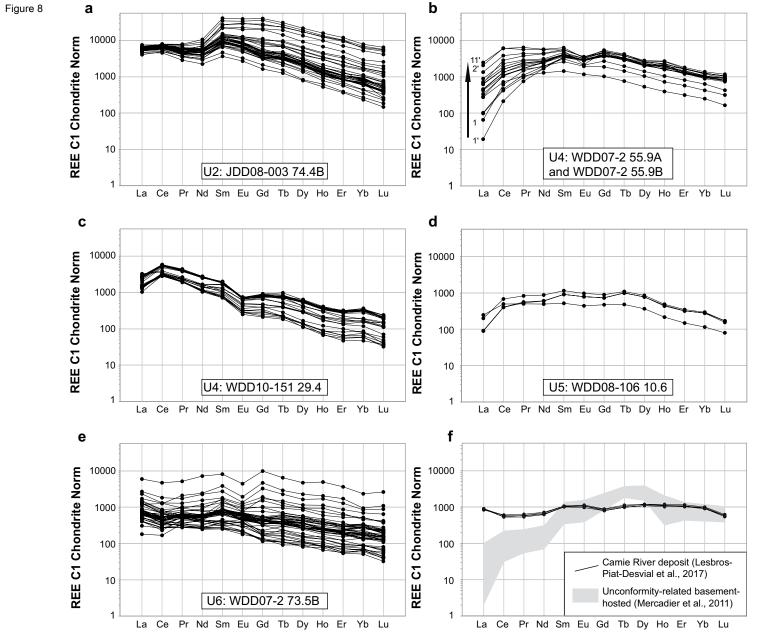
Ca

Fe

Si









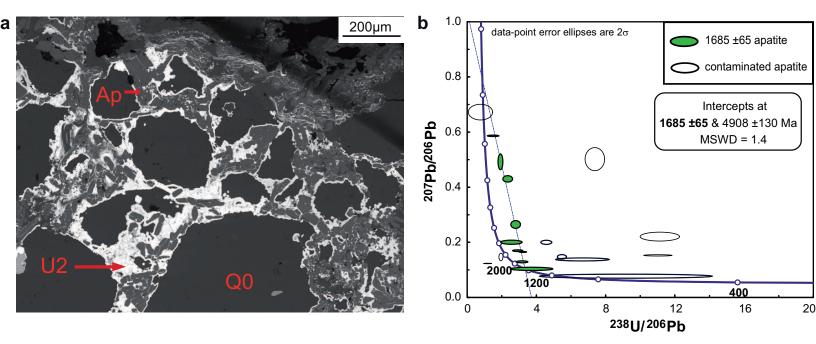
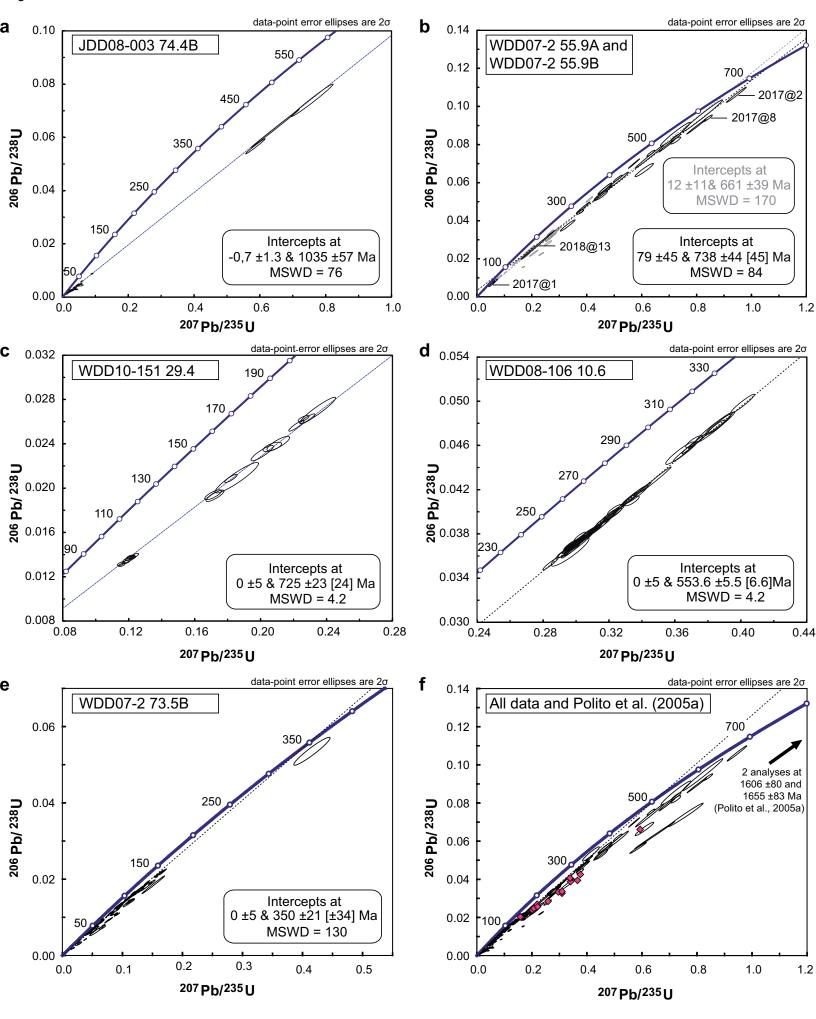
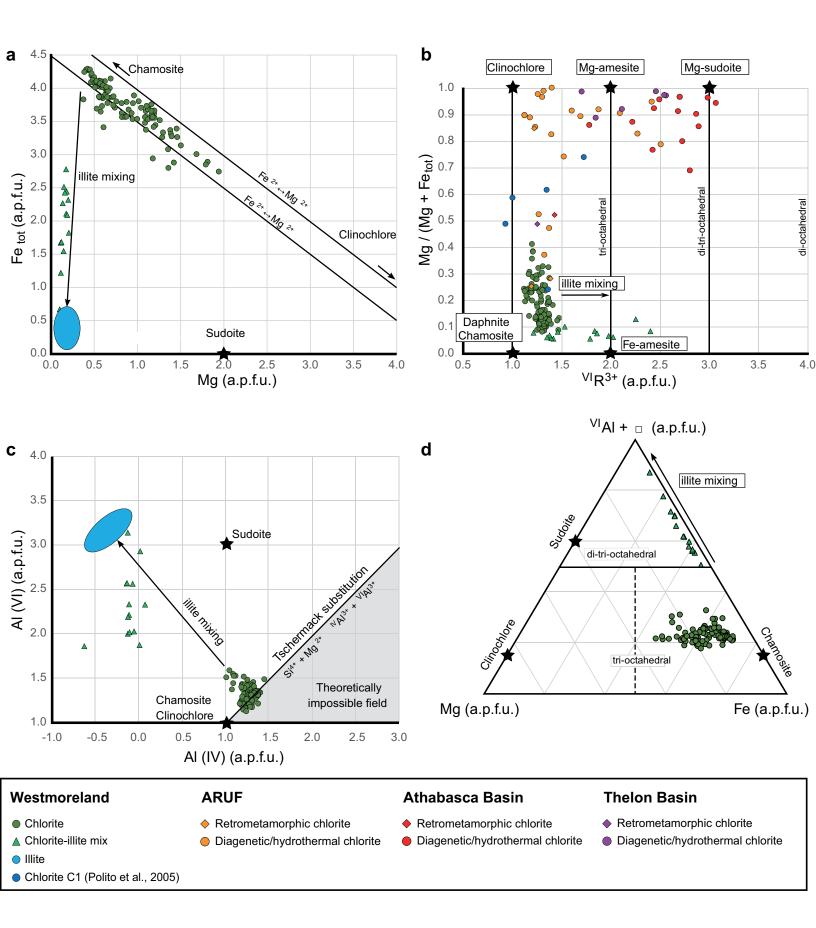


Figure 10





	1700 Ma	1650) Ma 1600	Ма			Deposit	Ref.
1880 1700				800-900	400-500	Several deposits	1	
1774 (basement) 1656		16				Nabarlek, Jabiluka	2	
1737				1554 1425 1355 —			Jabiluka,	3
	1720-1680			1437 1400		475	Ranger Ranger	4
U ARUF			1640	640 750			Nabarlek	5
	~1700 16	680	~1640	1 <u>30</u> 2	802		Jabiluka	6
	168	83			880		Nabarlek, Jabiluka,	7
			1650-1600				Koongarra	
U-Cu Westmoreland	1	680	1650 - 1606		870		Junnagunr	na. 8
				1	030 730	550 350	Redtree	
	168	85					Redtree	, .
U-Pb. ²⁰⁷ Pb	⊳/ ²⁰⁶ Pb or Sm-Nd on urai	ninites	Pb mod	el age from galena		⁴⁰ Ar/	³⁹ Ar on illite	
	Itered ore and white-mica			⁰⁶ Pb tuffaceous, volcanic input contemporary to sediment dep	oosition		o on apatite	

Figure 12

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ESM1

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