Low $\delta^{18}O$ zircon grains in the Neoarchean Rum Jungle Complex, northern Australia: An indicator of emergent continental crust

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ABSTRACT

The timing of widespread continental emergence is generally considered to have had a dramatic effect on the hydrological cycle, atmospheric conditions, and climate. New secondary ion mass spectrometry (SIMS) oxygen and laser-ablation–multicollector–inductively coupled plasma–mass spectrometry (LA-MC-ICP-MS) Lu-Hf isotopic results from dated zircon grains in the granitic Neoarchean Rum Jungle Complex provide a minimum time constraint on the emergence of continental crust above sea level for the North Australian craton. A 2535 ± 7 Ma monzogranite is characterized by magmatic zircon with slightly elevated $\delta^{18}O$ (6.0‰–7.5‰ relative to Vienna standard mean ocean water [VSMOW]), consistent with some contribution to the magma from reworked supracrustal material. A supracrustal contribution to magma genesis is supported by the presence of metasedimentary rock enclaves, a large population of inherited zircon grains, and subchondritic zircon Hf (ε_{Hf} = –6.6 to –4.1). A separate, distinct crustal source to the same magma is indicated by inherited zircon grains that are dominated by low $\delta^{18}O$ values (2.5‰–4.8‰, *n* = 9 of 15) across a range of ages (3536–2598 Ma; ε_{Hf} = –18.2 to +0.4). The low $\delta^{18}O$ grains may be the product of one of two processes: (1) grain-scale diffusion of oxygen in zircon by exchange with a low $\delta^{18}O$ magma or (2) several episodes of magmatic reworking of a Mesoarchean or older low $\delta^{18}O$ source. Both scenarios require shallow crustal magmatism in emergent crust, to allow interaction with rocks altered by hydrothermal meteoric water in order to generate the low $\delta^{18}O$ zircon. In the first scenario, assimilation of these altered rocks during Neoarchean magmatism generated low $\delta^{18}O$ magma with which residual detrital zircons were able to exchange oxygen, while preserving their U-Pb systematics. In the second scenario, wholesale melting of the altered rocks occurred in several distinct events through the Mesoarchean, generating low $\delta^{18}O$ magma from which zircon crystallized. Ultimately, in either scenario, the low $\delta^{18}O$ zircons were entrained as inherited grains in a Neoarchean granite. The data suggest operation of a modern hydrological cycle by the Neoarchean and add to evidence for the increased emergence of continents by this time.

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INTRODUCTION

Ever since Logan (1857) identified a fundamental difference between Laurentian and Huronian series rocks in Canada, and Stockwell (1961) dated the age of the former at ca. 2.5 Ga, Archean Earth has widely been regarded as a significantly different planet to its younger equivalent. This difference is primarily manifest as a unique dome-and-keel map pattern of Archean granite-greenstone terranes (Macgregor, 1951; Hickman, 1984) with basaltic carapaces erupted largely under submarine conditions (Arndt, 1999; Kump and Barley, 2007). Lack of widespread subaerial exposure of crustal blocks until the end of the Neoarchean reflects hotter geotherms and more ductile continental lithosphere that was generally unable to support large mountain belts (Arndt, 1999; Rey et al., 2003; Cruden et al., 2006; Flament et al., 2011). Only when crustal radiogenic heat production decreased (e.g., Bodorkos and Sandiford, 2006), and geothermal gradients fell with secular cooling of the mantle (e.g., Labrosse and Jaupart, 2007; Brown, 2008), did the continental crust stiffen and emerge above sea level (Taylor and McLennan, 1985; Flament et al., 2008, 2011). Although there is considerable evidence for emergent continental crust at various times through the Archean (e.g., from the Paleoarchean to Neoarchean sedimentary basins of the Kaapvaal and Pilbara cratons; McLennan et al., 1983; Nocita and Lowe, 1990; Nelson et al., 1999; Van Kranendonk et al., 2002; Hessler and Lowe, 2006), the timing of widespread continental emergence in the latest Neoarchean into the early Paleoproterozoic is only loosely constrained by poorly dated supracrustal successions and by geochemical and isotopic proxies (e.g., Taylor and McLennan, 1985; Eriksson et al., 1999; Farquhar et al., 2000; Anbar et al., 2007).

Constraining the timing of continental emergence is important because newly exposed continental crust would have had a dramatic effect on the hydrological cycle, as well as on global atmospheric conditions, climate, and biological evolution. Newly exposed Neoarchean crust would have effected

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changes to the atmosphere and hydrosphere and climate through increased drawdown of CO_2 via continental weathering. In combination with a change from dominantly submarine to subaerial volcanism (e.g., Kump and Barley, 2007), a decrease in overall magmatism and associated volcanic degassing at the close of the Archean (Condie et al., 2009), and an increase in photosynthesis prompted by increased areas of continental shelves and erosive supply of nutrients to the oceans (e.g., Campbell and Allen, 2008), CO_2 drawdown via weathering of newly exposed crust contributed to oxygenation of the atmosphere (Farquhar et al., 2000; Anbar et al., 2007), which in turn supported the evolutionary development of eukaryotic life (e.g., Margulis et al., 1976; Martin and Müller, 1998; Van Kranendonk, 2012).

An important indicator of continental emergence is the interaction of continental crust and its derived weathering products with meteoric water, which can be recorded in the oxygen isotopic signature of zircon (e.g., Tang et al., 2008). Meteoric water is characterized by low $\delta^{18}O$ (typically 0‰ to −55‰; Valley et al., 2005) as a result of preferential evaporation of ^{16}O at Earth's surface. Oxygen diffusion in zircon is regarded to be prohibitively slow in most crustal environments, including during high-temperature metamorphism and anatexis (Peck et al., 2003; Moser et al., 2008). However, in a restricted range of environments involving high-temperature hydrothermal fluids (e.g., Bindeman et al., 2008), alteration of the primary oxygen isotopic character of zircon can indicate the interaction between zircon crystals and fluids.

In this paper, we present data from a Neoarchean granite from the Rum Jungle Complex of northern Australia that contains evidence of fractionated zircon oxygen isotopes indicative of an active hydrological cycle affecting emergent continental crust by ca. 2535 Ma. The data presented are interpreted as evidence of the minimum time of continental emergence of the North Australian craton. The data also highlight the potential for similar samples to yield valuable information on fluid-rock interaction and the development of continental emergence through the Archean-Proterozoic transition.

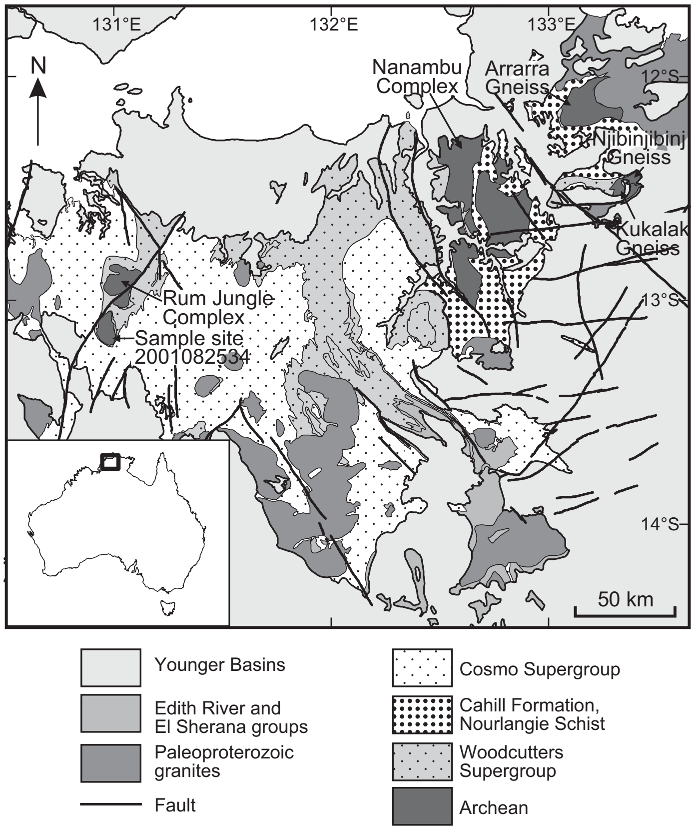

Figure 1. Location of the Rum Jungle Complex within the Pine Creek Orogen. Inset map shows the location within Australia.

GEOLOGICAL SETTING

Exposed Neoarchean basement of the North Australian craton lies within the Paleoproterozoic Pine Creek Orogen (Fig. 1). The basement is dominated by ca. 2545–2521 Ma granite and granitic gneiss of the Rum Jungle Complex (Cross et al., 2005), the ca. 2520 Ma Nanambu Complex, and the ca. 2527–2510 Ma Kukalak Gneiss (Page et al., 1980; Hollis et al., 2009; Carson et al., 2010; Kositcin et al., 2012), and ca. 2640 Ma Arrarra Gneiss and ca. 2670 Ma Njibinjibinj Gneiss (Hollis et al., 2009; Carson et al., 2010). Assuming that these form a continuous basement, largely under cover of younger rocks, this represents an extent of at least 22,000 km^2 of Neoarchean crust (see Hollis et al., 2009).

The Rum Jungle Complex outcrops in the Rum Jungle and Waterhouse domes and is separated from overlying sedimentary and volcanic rocks of the ca. 2020 Ma Woodcutters Supergroup by a major unconformity (Needham et al., 1988). The Rum Jungle Complex consists mainly of syenogranite, monzogranite, and quartz monzonite (Drüppel et al., 2009), intruded by pegmatites, dolerite, and quartz tourmaline veins (Lally, 2002). The Rum Jungle Complex includes enclaves of amphibolite-facies metasedimentary rocks of the Stanley Metamorphics, which include biotite gneiss, biotite-muscovite gneiss, biotite granofels, feldspathic gneiss, quartz-muscovite schist, chlorite schist, actinolite schist, and banded ironstone, which were deformed and metamorphosed to amphibolite facies prior to being incorporated into the Rum Jungle Complex, and which are variably retrogressed (Rhodes, 1965; Lally, 2002). There are no available constraints on the pressure-temperature conditions of metamorphism. The timing of deformation and metamorphism also remains unconstrained and could be as young as the emplacement age of the main population of late Neoarchean granites themselves.

Five granites and one diorite from the Rum Jungle Complex yield secondary ion mass spectrometry (SIMS; sensitive high-resolution ion microprobe [SHRIMP]) U-Pb zircon magmatic crystallization ages in the range 2545–2521 Ma (Cross et al., 2005). Here, we present data from one of these (sample 2001082534), a fine- to medium-grained, equigranular monzogranite (Fig. 1; GDA94 UTM zone 52, 710747mE, 8543205mN). This is the only one of the six dated samples of the Rum Jungle Complex that contains inherited zircon. It belongs to the felsic group of Drüppel et al. (2009; see also Northern Territory Geological Survey, 2012), which are rich in K and large ion lithophile elements (LILEs), are depleted in Sr, Eu, and high field strength elements (HFSEs, e.g., Nb and Ti), and have anomalously high Th and U. These rocks are thought to have formed by intracrustal melting (Drüppel et al., 2009). Analytical methodologies for sample preparation, zircon U-Th-Pb and O SIMS analysis, and zircon Lu-Hf laser-ablation–multicollector–inductively coupled plasma–mass spectrometry (LA-MC-ICP-MS) analysis are described in Appendix 1.[1]

ZIRCON ISOTOPIC RESULTS

Zircons are dominated by euhedral to subrounded prisms and their broken equivalents that range from clear and colorless to turbid and

[1]GSA Data Repository Item 2014079, Appendix 1: zircon analytical techniques, is available at www.geosociety.org/pubs/ft2014.htm, or on request from editing@geosociety.org, Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301-9140, USA.

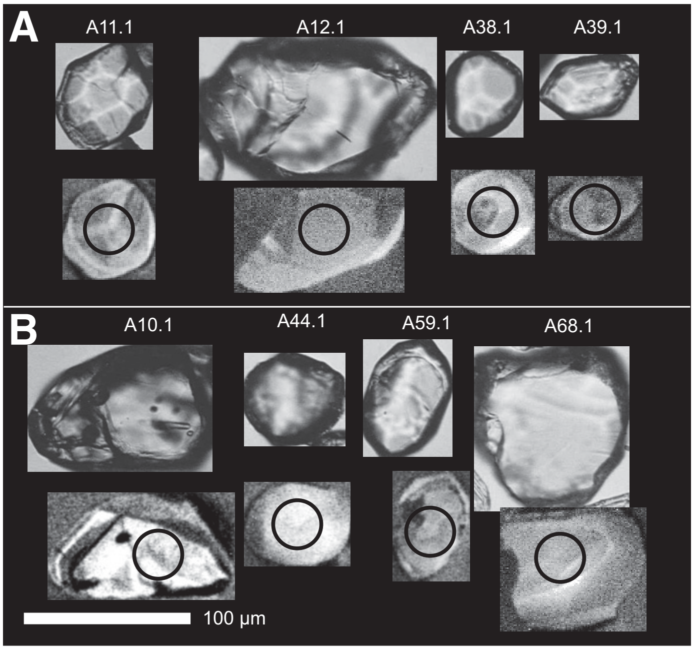

Figure 2. Representative transmitted light (top rows) and cathodoluminescence (bottom rows) images of analyzed zircon grains from sample 2001082534. (A) Neoarchean zircon grown during magmatic crystallization. (B) Paleoarchean to Mesoarchean inherited zircon. Analyzed sites are indicated by circles on the zircon images. O analyses were made on exactly the same spot locations as the U-Pb analyses of Cross et al. (2005). See Table 1 for analytical data for each labeled site.

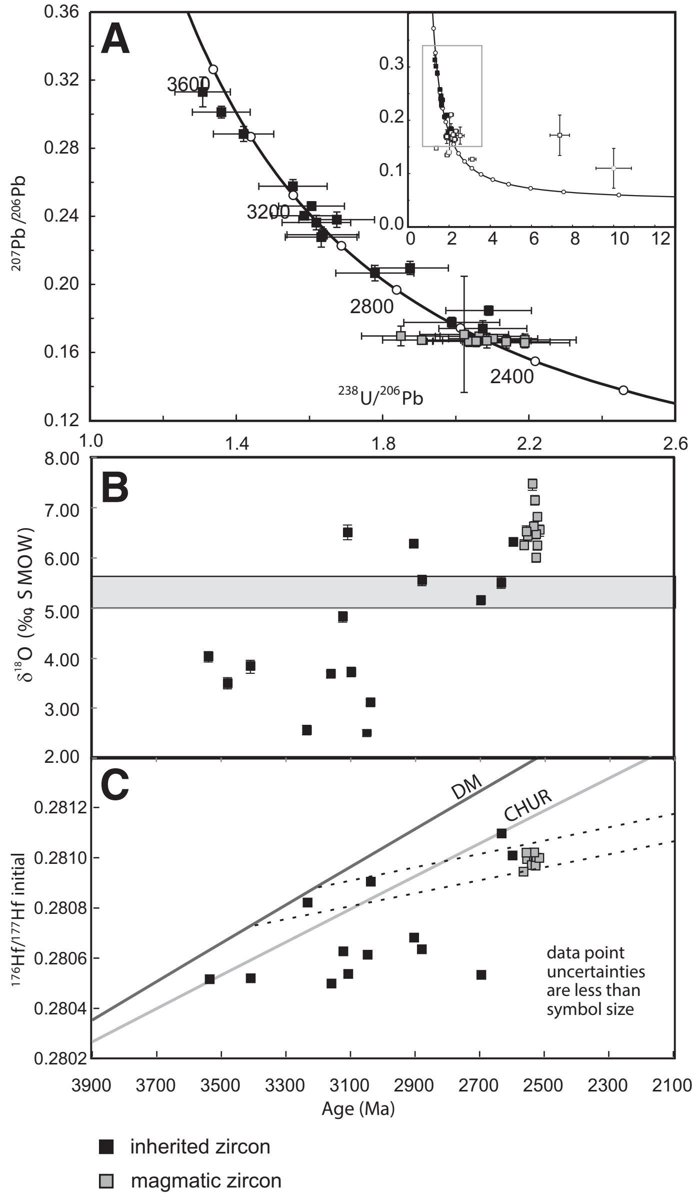

Figure 3. (A) Tera-Wasserburg plot of zircon U-Pb secondary ion mass spectrometry (SIMS) data for sample 2001082534. Magmatic zircon grown during granite crystallization is shown in gray, and inherited grains are shown in black. Error bars are 1σ. (B) Zircon $\delta^{18}O$ data for the same sample. Error bars are 2σ. SMOW—standard mean ocean water. (C) Zircon $^{176}Hf/^{177}Hf$ data for the same sample. Chondritic values are from Blichert-Toft and Albaréde (1997), and present-day depleted mantle values are from Vervoort and Blichert-Toft (1999). Dashed lines show Hf isotope evolution lines for the preferred initial Lu/Hf ratio of 0.015, encapsulating the Hf isotopic range for the magmatic zircon and indicating average crustal residence ages of 3.4–3.2 Ga.

brown. Grain size varies from 50 to 200 μm in length with aspect ratios of 1:1–1:3. Many grains show well-developed oscillatory or sector zoning, a few are homogeneous, and some display irregular embayment of oscillatory zones and development of oscillatory zoned or homogeneous rims (Fig. 2). Approximately 10% of the grains have optically distinct cores.

U-Pb Results

Fifty-one analyses were made on 46 zircon grains from sample 2001082534 (Cross et al., 2005). Fourteen analyses are discordant above an arbitrary threshold of 10% or have high common Pb contents of >8% $^{206}Pb_c$ and are thus not considered further. A further 10 analyses are interpreted as mixtures of different age components and are also not considered further. The remaining 27 analyses are characterized by a dominant population between 2695 Ma and 2515 Ma, and a range of discrete older grains that extend up to ca. 3535 Ma (Fig. 3A; Table 1). The 10 youngest grains are clear, colorless, euhedral to subrounded, prismatic grains with faint to well-developed oscillatory or sector zoning (Fig. 2). Twelve analyses of these 10 grains yield a weighted mean $^{207}Pb/^{206}Pb$ age of 2535 ± 7 Ma (95% confidence, mean square of weighted deviates [MSWD] = 0.92), interpreted as the age of magmatic crystallization. This age is consistent with other ages for the volumetrically dominant component of the Archean basement to the North Australian craton (2545–2510 Ma; Cross et al., 2005; Hollis et al., 2009). The range of older dates is interpreted to reflect a substantial component of zircon inheritance in this sample. These grains are morphologically diverse, ranging from clear to patchy brown, prismatic to rounded, and equant to elongate. They are usually oscillatory zoned, although a few are homogeneous or sector zoned (Fig. 2).

Oxygen Isotopic Data

Oxygen isotopic ratios were measured for 26 zircons across a range of ages (Fig. 3B; Table 1). Eleven grains of the 2535 ± 7 Ma magmatic population fall within a limited range of $\delta^{18}O$ = 6.0‰–7.5‰ (relative to Vienna standard mean ocean water [VSMOW]), indicative of a contribution from reworked, or assimilated, supracrustal rocks. Inherited grains

(3536–2632 Ma) span a broad range in $\delta^{18}O$ from 2.5‰ to 6.5‰. The majority of these grains (n = 9 of 15, 3536–3037 Ma) have $\delta^{18}O$ values (2.5‰–4.8‰) significantly below what would be expected for zircon grown in equilibrium with a mantle-derived melt (5.3‰ ± 0.6‰; Valley et al., 2005; Fig. 3B; Table 1).

Lu-Hf Results

Hf isotopic ratios were measured for 23 zircons. The 2535 ± 7 Ma magmatic population is characterized by a weak spread in subchondritic Hf values ($^{176}Hf/^{177}Hf$ = 0.280052–0.281064, ε_{Hf} = –4.1 to –6.6; Fig. 3C; Table 1), consistent with derivation from melting of significantly older (Paleoarchean) sources with crustal residence ages of older than ca. 3.3 Ga (assuming initial $^{176}Lu/^{177}Hf$ = 0.015). The 15 inherited grains analyzed (3536–2632 Ma) range from strongly radiogenic (close to depleted mantle) to highly unradiogenic ($^{176}Hf/^{177}Hf$ = 0.280581, ε_{Hf} = –18.2, at 2695 Ma). Three inherited grains have a two-stage model age similar to that of the 2535 Ma population (ca. 3.3 Ga; Fig. 3C) and may have been derived from comparable sources. The 10 other analyses have two-stage model ages ranging from 4.3 to 3.0 Ga (Table 1).

DISCUSSION

Low $\delta^{18}O$ zircon ($\delta^{18}O$ < 5.3‰ ± 0.6‰, i.e., less than that in equilibrium with mantle-derived melt) is rare, particularly in Archean rocks (e.g., Valley et al., 2005; data compiled in Van Kranendonk and Kirkland, 2013). The sample analyzed here (sample 2001082534) is unusual amongst dated samples of the Archean basement of the North Australian craton (e.g., Hollis et al., 2009), and unique amongst the dated samples of the Rum Jungle Complex (Cross et al., 2005), in that it contains inherited zircons. These inherited grains are also unusual in being dominated by low $\delta^{18}O$ values (n = 9/15, $\delta^{18}O$ = 2.5‰–4.8‰). Therefore, although the data set presented here is small, and restricted to a single sample, the data nonetheless provide a potentially significant insight into otherwise elusive environmental processes operating in the Archean.

The zircon U-Pb, Lu-Hf, and O isotopic systematics of zircon grains from sample 2001082534 indicate diverse crustal sources that contributed to the granitic magma. The zircon population can be divided into two broad components based on age and isotopic signature: (1) a ca. 2535 Ma magmatic zircon component (n = 12), grown within the magma, which is characterized by elevated $\delta^{18}O$ values of 6.0‰–7.5‰ and a subchondritic zircon Hf mixing trend (Fig. 3C; $^{176}Hf/^{177}Hf$ = 0.280052–0.281064, ε_{Hf} = –4.1 to –6.6); and (2) an inherited zircon component, identified by a large proportion (9 of 15) of xenocrystic grains that have low $\delta^{18}O$ values of 2.5‰–4.8‰. A further six inherited grains have slightly higher $\delta^{18}O$ values of 5.2‰–6.5‰ and may represent a distinct inherited component to the granite. Of these six grains, three that have mantle-like to slightly higher $\delta^{18}O$ values (5.2‰–6.5‰) have U-Pb zircon ages and $\delta^{18}O$ isotopic values consistent with the known older component of the Archean basement of this region (ca. 2640 Ma and 2670 Ma; Hollis et al., 2010; Beyer et al., 2012). This component is interpreted to have been derived from partial melting or assimilation of older granitic basement in the region. The other three inherited grains have significantly older concordant U-Pb ages (3107–2879 Ma) and $\delta^{18}O$ isotopic values of 5.5‰–6.5‰ and are derived from unknown sources.

The 2535 Ma magmatic zircon population, interpreted to have grown during granite crystallization, has a small spread in elevated $\delta^{18}O$ values of 6.0‰–7.5‰ and relatively clustered, evolved Hf isotopic compositions (ε_{Hf} = –4.1 to –6.6). Such values are consistent with assimilation of Paleoarchean or older sedimentary protoliths into the granitic magma.

Regarding the second distinct crustal source, nine of the 15 analyzed inherited grains (60%) have $\delta^{18}O$ values significantly lower than that expected for zircon in equilibrium with mantle-derived melt (2.5‰–4.8‰). These low $\delta^{18}O$ values indicate that the zircon grains either crystallized from, or diffusively exchanged oxygen with, a low $\delta^{18}O$ melt or fluid. As the Rum Jungle Complex granitic melt is interpreted to have had an elevated bulk $\delta^{18}O$ composition (source 1 listed earlier), the low $\delta^{18}O$ zircons indicate a distinct source or sources.

Two questions should be addressed in relation to this second source: (1) What was the nature of the fluid responsible for generating a low $\delta^{18}O$ source, or sources? (2) Did the zircons crystallize from, or did they diffusively exchange oxygen with, that low $\delta^{18}O$ source?

Nature of the Fluid that Generated a Low $\delta^{18}O$ Source

Low $\delta^{18}O$ zircon that is produced by crystallization from, or diffusive exchange with, a melt or fluid requires interaction with a source having $\delta^{18}O$ at least as low as, and probably much lower than, the minimum $\delta^{18}O$ value found in zircon (Bindeman, 2011). In the case examined here, this source must have been lower than 2.5‰, the lowest value analyzed herein (Table 1). One possibility is that the fluid involved was seawater (assuming seawater $\delta^{18}O$ ~ 0‰ during the Archean; e.g., Gregory and Taylor, 1981; Gregory, 1991). However, this is considered unlikely, because very large volumes of seawater, with continued replenishment, would be required to achieve the observed degree of isotopic exchange. Similarly low $\delta^{18}O$ zircons (2.4‰–4.4‰) are known from remelting of basaltic oceanic crust that has been hydrothermally altered by seawater (e.g., Peck, 2000). However, in the case of the inheritance in the granite studied here, a much more extensive isotopic exchange with seawater would be required (e.g., Eiler, 2001) to reduce the relatively $\delta^{18}O$ heavy metasedimentary rocks (possibly the Stanley Metamorphics) to the low $\delta^{18}O$ values implied by the range of inherited zircon ages.

Rather, we consider that hydrothermal exchange of much lower $\delta^{18}O$ meteoric fluids ($\delta^{18}O$ = 0‰ to –55‰; Valley et al., 2005) with supracrustal rocks in a near-surface environment presents the most viable method of producing the observed low $\delta^{18}O$ values in the source. This conclusion is supported by previous studies that have deduced a meteoric origin of fluids that generated low $\delta^{18}O$ sources, which in turn yielded low $\delta^{18}O$ zircon (e.g., 2‰–4‰ for Eo- to Neoarchean Greenland gneisses—Hiess et al., 2011; 2‰–3‰ for Neoproterozoic Dabie-Sulu granites—Wang et al., 2011; 2‰–4‰ for Tertiary granitic rocks, Scotland—Gilliam and Valley, 1997).

How and When Did the Zircons Obtain Their Low $\delta^{18}O$ Signature?

Low $\delta^{18}O$ values in zircon are usually explained as the result of crystallization from magmatic rocks produced by melting of crust that was previously altered by low $\delta^{18}O$ fluids (e.g., Gregory and Taylor, 1981; Gilliam and Valley, 1997; King et al., 2000; Valley et al., 2005; Bindeman, 2008; Hiess et al., 2011). Alternatively, hydrothermal zircon can grow directly from solutions with a low $\delta^{18}O$ component (Kirkland et al., 2009). A third alternative is that intracrystalline diffusion of oxygen in zircon can occur under hydrothermal supersolidus conditions, as indicated by the experimental data of Watson and Cherniak (1997; see also Cherniak and Watson, 2003), although evidence for oxygen diffusion has not been observed in natural samples, which instead more commonly show evidence of dissolution and reprecipitation (e.g., Bindeman et al., 2008). In the case of the Rum Jungle Complex, the timing of alteration to form the low $\delta^{18}O$ source is important in understanding the process by which the zircon obtained low $\delta^{18}O$ values. Next, we assess whether alteration could have occurred after, during, or before granite emplacement.

(1) Interaction of meteoric fluids with the granite *after* granite emplacement and crystallization is considered unlikely, because the oxygen isotopic systematics of the 2535 Ma magmatic zircon population form a coherent population and are undisturbed. Alteration via this mechanism would require these magmatic grains to be more resilient to alteration, whereas fracturing and metamictization of the older, inherited grains (resulting from alpha recoil damage) preferentially would have resulted in oxygen diffusion in these grains. In this scenario, one would expect there to be a correlation between alpha dose and $\delta^{18}O$. However, as shown in Figure 4, there is no such correlation. Alpha doses and grain densities (Table 1) were calculated using the method of Murakami et al. (1991), who compared radiation doses to transmission electron microscopy (TEM) diffraction patterns and outlined three stages of zircon structure, ranging from crystalline to completely amorphous. Each stage is defined by characteristic radiation doses. Calculations indicate that the majority of grains analyzed from the sample studied here have radiation doses in the range that is consistent with a highly crystalline structure (i.e., $<3 \times 10^{15}$ alpha events/mg). Also, the inherited grains with low $\delta^{18}O$ all show a high degree of U-Pb concordance (>95%), consistent with low, or no, alteration. Furthermore, there is no correlation between age and uranium content (Table 1), which implies that radiogenic-Pb loss and disturbance have not demonstrably affected the zircon population. Finally, calculated ε_{Hf} values are sensitive to disturbance of U-Pb systematics (which might also indicate disturbance of the O isotopic system), resulting in erratic or scattered $^{177}Hf/^{176}Hf$ arrays. This is not the case for this sample (i.e., clustered ε_{Hf} of –4.1 to –6.6), which is inconsistent with the generation of low $\delta^{18}O$ values by postmagmatic alteration of zircon.

(2) Alteration of the supracrustal rocks to low $\delta^{18}O$ values by meteoric water *before* granite emplacement could either have occurred by (a) generation of a low $\delta^{18}O$ source prior to ca. 3.5 Ga (the age of the oldest low $\delta^{18}O$ grain), from which all of the low $\delta^{18}O$ inherited zircons were then grown during subsequent remelting events, or (b) diffusive exchange of oxygen in the inherited zircons with a low $\delta^{18}O$ melt into which they were entrained after ca. 3.0 Ga (the age of the youngest low $\delta^{18}O$ grain), but before emplacement of the 2535 Ma granite. The latter possibility is considered along with item 3 next.

A key observation regarding this scenario is that the low $\delta^{18}O$ zircon values occur in inherited grains with a wide range of ages (3536–3037 Ma). Therefore, if these grains crystallized directly from either a low $\delta^{18}O$ melt or from a hydrothermal fluid, then this must have occurred at different times, over the course of ~500 m.y. (between 3.5 and 3.0 Ga, i.e., the ages of the low $\delta^{18}O$ inherited zircons). Particularly given the rarity of low $\delta^{18}O$ zircon in Archean rocks in general (e.g., data compiled in Van Kranendonk and Kirkland, 2013), this is most likely to have occurred by magmatic reworking of the same older than 3.5 Ga low $\delta^{18}O$ source. This source must have been originally formed by interaction with and alteration by meteoric water, and it was then melted and remelted at various times through the Paleoarchean to Mesoarchean. This reworking of the same source suggests an isolated geological system over at least 500 m.y. The resulting low $\delta^{18}O$ zircons, crystallized from low $\delta^{18}O$ magma during several reworking events, were then delivered as inherited zircons into the Rum Jungle Complex through a combination of erosion, deposition in a sedimentary environment, and assimilation into the granite.

(3) Alteration of metasedimentary rocks by meteoric water *during* intrusion of granitic magmas could have occurred by magma intrusion resulting in fracturing of the host rocks and associated infiltration and circulation of meteoric groundwater heated by the intruding granite. Metasedimentary supracrustal rocks (perhaps the Stanley Metamorphics) were altered to low $\delta^{18}O$ bulk compositions by the circulating, heated meteoric water through a process of repeated intrusion, alteration, and wholesale melting and assimilation of altered rocks (e.g., Bindeman, 2008). Generation of low $\delta^{18}O$ magmas by this process has been suggested for a range of modern and ancient felsic magmatic rocks in varied environments, including near-surface magmatic-hydrothermal systems (e.g., Yellowstone Plateau volcanic field—Hildreth et al., 1984; Bindeman and Valley, 2000, 2001; Bindeman et al., 2001, 2008; Heise volcanic field, Idaho—Bindeman et al., 2007; Timber Mountain/Oasis Valley caldera complex—Bindeman and Valley, 2003; Dabie-Sulu orogen—Zheng et al., 2004; Tang et al., 2008; West Greenland gneisses—Hiess et al., 2011).

These processes of alteration and crustal cannibalization may have occurred at any time between ca. 3.0 Ga (the age of the youngest low $\delta^{18}O$ inherited grain) and 2535 Ma (the age of the Rum Jungle Complex granite), but they are most likely to have occurred during emplacement of the Neoarchean magmas, because, locally, this is the only known magmatic heating event in the period 3.0–2.5 Ga.

In this scenario, detrital zircon captured from assimilated metasedimentary rocks diffusively exchanged oxygen with a low $\delta^{18}O$ magma on a time scale of <1 m.y. while not affecting the U-Pb system of the magma and magmatic zircons, which requires much higher temperatures and longer time scales (Cherniak and Watson, 2001, 2003). Recharge of the system with granitic magmas having more elevated $\delta^{18}O$ bulk compositions (generated by deeper-level melting in the mid- to upper crust) resulted in assimilation of existing low $\delta^{18}O$ magmas and their altered, low $\delta^{18}O$ inherited zircon component, while accounting for crystallization of the distinct 2535 Ma elevated $\delta^{18}O$ magmatic zircon population.

Supporting this model is new evidence of low $\delta^{18}O$ zircon from the Billabong Complex, Tanami region, located ~800 km to the south of the Rum Jungle Complex (Whelan et al., 2013). SIMS oxygen data for the Billabong Complex reveal a significant proportion (~30%) of low $\delta^{18}O$ magmatic and inherited zircon in the range ca. 2550–2510 Ma ($\delta^{18}O$ = 2.0‰–4.7‰; Whelan et al., 2013). These data indicate that high-crustal-level hydrothermal-meteoric systems may have been widespread during Neoarchean magmatism in the North Australian craton.

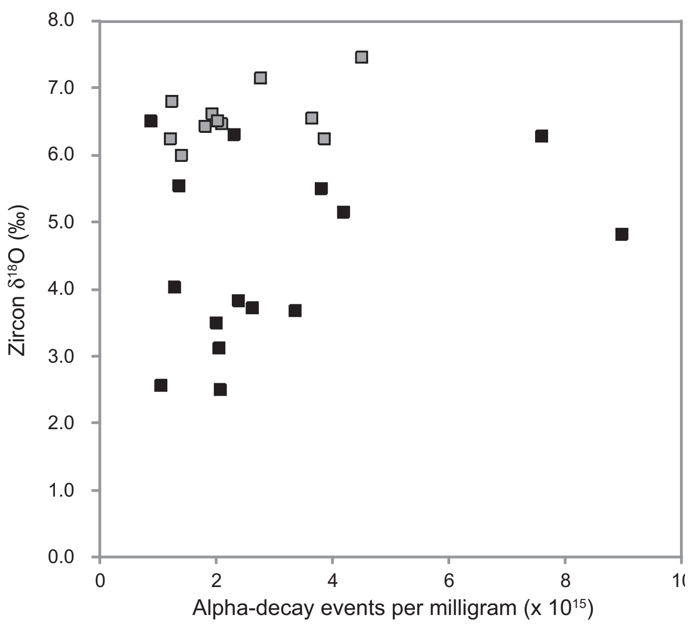

Figure 4. Alpha recoil events (dose) vs. $\delta^{18}O$ for zircon from sample 2001082534. See Table 1 for tabulated alpha recoil data. Magmatic zircon grown during granite crystallization is shown in gray, and inherited grains are shown in black.

TABLE 1. TABULATED U-Pb-Th AND O SIMS AND Lu-Hf LA-ICP-MS DATA FOR SAMPLE 2001082534, RUM JUNGLE COMPLEX

Analysis no.	U (ppm)	Th/U_{calc}	% common ^{206}Pb	$^{207}Pb/^{206}Pb$	± σ %	$^{206}Pb/^{238}U$	± σ %	$^{207}Pb/^{206}Pb$ age (Ma)	± σ	Disc %	Hf^{176}/Hf^{177}	1 s.e.	ε_{Hf}	T_{DM} (Ma)	$\delta^{18}O$ (‰)	1 σ	No. alpha events/mg (×10^{15})	Metamict stage	Calc. density (gcm^{-3})
Accepted analyses																			
A52.1	268	1.28	1.00	0.1657	0.9	0.457	2.8	2515	15	4	0.281041	0.000020	–5.8	3333	6.55	0.12	3.65	II: intermediate	4.44
A69.1	278	1.49	3.26	0.1663	1.1	0.468	2.8	2521	19	2							3.92	II: intermediate	4.42
A06.2	320	0.51	1.43	0.1666	0.9	0.491	2.8	2524	16	–2					6.24	0.08	3.87	II: intermediate	4.42
A38.1	93	1.13	0.45	0.1666	1.0	0.486	2.9	2524	16	–1	0.281032	0.000028	–5.5	3325	6.81	0.05	1.24	I: crystalline	4.68
A11.1	108	0.95	0.36	0.1668	0.8	0.486	2.9	2526	13	–1	0.281014	0.000020	–6.2	3368	6.00	0.08	1.41	I: crystalline	4.66
A39.1	146	1.61	0.93	0.1669	1.3	0.480	2.9	2527	21	0	0.281016	0.000015	–6.6	3395	6.46	0.08	2.10	I: crystalline	4.60
A56.1	215	0.87	1.03	0.1673	0.7	0.524	2.8	2531	12	–7	0.281064	0.000019	–4.7	3274	7.14	0.09	2.77	I: crystalline	4.53
A05.2	142	1.23	0.55	0.1674	1.0	0.457	3.2	2532	17	4	0.281029	0.000026	–5.5	3328	6.62	0.10	1.94	I: crystalline	4.61
A06.1	370	0.51	0.09	0.16807	0.4	0.476	2.9	2539	6.3	1	0.281011	0.000022	–6.2	3380	7.46	0.11	4.51	II: intermediate	4.36
A05.1	126	1.55	0.46	0.1695	0.7	0.484	2.9	2553	11	0	0.281032	0.000019	–5.1	3317	6.43	0.09	1.82	I: crystalline	4.63
A58.1	144	1.35	3.51	0.1697	1.7	0.541	2.9	2555	29	–9	0.281052	0.000021	–4.1	3258	6.52	0.11	2.02	I: crystalline	4.61
A12.1	91	0.99	0.31	0.1706	10.0	0.494	3	2564	17	–1	0.280969	0.000013	–6.6	3423	6.24	0.11	1.22	I: crystalline	4.68
Inherited																			
A51.1	170	0.99	1.28	0.1741	1.3	0.482	2.9	2598	21	2	0.281040	0.000036	–3.5	3253	6.31	0.08	2.31	I: crystalline	4.58
A37.3	313	0.22	2.00	0.1777	0.8	0.503	3.3	2632	14	0	0.281143	0.000015	0.4	3030	5.50	0.11	3.81	II: intermediate	4.43
A13.2	335	0.17	0.56	0.1846	0.7	0.478	2.8	2695	12	6	0.280581	0.000013	–18.2	4252	5.15	0.09	4.18	II: intermediate	4.39
A46.1	94	0.53	0.50	0.2066	1.1	0.562	3	2879	18	0	0.280682	0.000019	–10.3	3897	5.54	0.11	1.37	I: crystalline	4.67
A09.1	525	0.39	0.95	0.2096	0.9	0.533	2.8	2903	15	5	0.280731	0.000017	–8.1	3776	6.28	0.06	7.59	II: intermediate	4.15
A53.1	130	0.52	3.93	0.2278	1.3	0.613	3	3037	21	–1	0.280977	0.000020	3.0	3172	3.12	0.06	2.06	I: crystalline	4.60
A03.1	142	0.01	2.07	0.2292	0.8	0.611	3	3047	12	–1	0.280681	0.000014	–7.2	3827	2.50	0.06	2.07	I: crystalline	4.60
A44.1	158	0.65	0.29	0.2363	0.9	0.618	2.9	3095	14	0					3.72	0.09	2.63	I: crystalline	4.55
A40.1	52	0.71	0.52	0.238	1.0	0.597	3.1	3107	15	3	0.280588	0.000016	–8.5	3955	6.50	0.14	0.88	I: crystalline	4.71
A57.1	555	0.38	1.30	0.2403	0.4	0.631	2.8	3122	6.9	–1	0.280703	0.000017	–4.9	3742	4.83	0.10	8.96	III: highly metamict	4.10
A10.1	212	0.18	0.76	0.246	0.5	0.623	2.8	3159	8.1	1	0.280552	0.000014	–8.6	4005	3.68	0.06	3.37	II: intermediate	4.47
A41.1	61	0.52	0.38	0.2577	0.7	0.643	3	3233	12	1	0.280908	0.000024	4.6	3218	2.57	0.10	1.06	I: crystalline	4.69
A59.1	123	0.65	0.32	0.2884	0.8	0.704	2.9	3409	12	–1	0.280602	0.000021	–2.0	3775	3.83	0.12	2.39	I: crystalline	4.57
A01.1	101	0.61	<0.01	0.3012	0.6	0.736	2.9	3476	8.8	–2					3.50	0.11	2.01	I: crystalline	4.61
A68.1	64	0.47	0.08	0.3131	1.4	0.765	2.9	3536	22	–4	0.280574	0.000016	0.8	3688	4.02	0.10	1.29	I: crystalline	4.67

Note: Data use scheme of Murakami et al. (1991). SIMS—secondary ion mass spectrometry; LA-ICP-MS—laser-ablation–inductively coupled plasma–mass spectrometry; T_{DM}—depleted mantle model age.

The proposed scenario requires that the Rum Jungle Complex was emplaced at a shallow crustal level in order to access low $\delta^{18}O$ meteoric waters. Although there are no existing pressure-temperature constraints on the depth of emplacement, the geochemistry of the granites indicates they were formed by melting at pressures lower than the garnet stability field (<10 kbar; Drüppel et al., 2009). Therefore, they may have been emplaced at the shallow depths required to drive a meteoric-hydrothermal system (e.g., Bindeman, 2011). It may be possible to obtain a maximum constraint on emplacement depth by metamorphic studies of metasedimentary xenoliths of the Stanley Metamorphics within the Rum Jungle Complex. This model could be tested further by analyzing the whole-rock oxygen isotopic composition of the Stanley Metamorphics, which form xenoliths within, and wall rocks to, the Rum Jungle Complex.

Diffusive Exchange of Oxygen in Zircon in a Hydrothermal-Magmatic Environment

Diffusive exchange of oxygen in zircon with a low $\delta^{18}O$ melt is one of the two proposed models that could explain the occurrence of variably low $\delta^{18}O$ inherited zircon from a range of ages in the Rum Jungle Complex. However, oxygen diffusion profiles in zircon have, to date, not been demonstrated in natural samples. In order for this to be a viable mechanism, a long-lived, high-temperature (supersolidus) hydrothermal system within several kilometers of the surface is required, as oxygen diffusion in zircon is slow, even under prolonged exposure to high-temperature conditions, but particularly in the absence of fluid water (Page et al., 2007; Lancaster et al., 2009; Bowman et al., 2011). Examination of oxygen zoning profiles in low $\delta^{18}O$ zircon would be a useful further test.

Experimental data indicate that with water present, temperatures of ~700 °C are required to induce grain-scale diffusion for 120-μm-diameter grains on a time scale of 10^5 to 10^6 yr (Watson and Cherniak, 1997; Cherniak and Watson, 2003). A large, long-lived, high-crustal-level plutonic system would be required to sustain a hydrothermal-magmatic system capable of achieving this. Although the temperature-time history of the Rum Jungle Complex is impossible to analyze in the same detail as modern subvolcanic systems, the available data suggest that the Rum Jungle Complex may have fulfilled these constraints, as dated granites from the Rum Jungle Complex span a 24 m.y. age range (2545 ± 4 Ma, 2535 ± 7 Ma, 2534 ± 6 Ma, 2531 ± 3 Ma, 2525 ± 5 Ma, 2521 ± 4 Ma; Cross et al., 2005), thus recording a long history of high heat flow, and, potentially, meteoric water circulation, in the region.

Implications for Studies of Fluid-Rock Interaction in the Archean

The data presented here have implications for investigating the antiquity of emergence of the continents. In this study, the data provide a minimum constraint on the timing of emergence of Neoarchean crust of the North Australian craton at ca. 2535 Ma, the age of granite emplacement and inferred hydrothermal circulation of meteoric water under shallow crustal conditions. This constitutes the earliest evidence for emergence of continental crust of the North Australian craton.

However, this is not the earliest evidence for emergence of continental crust on a global scale, which has been established for rocks at least as old as 3.5 Ga (Buick et al., 1995). Elevated freeboard existed at various times throughout the Archean, as evidenced by Archean continental and shallow-marine successions (e.g., Burke et al., 1986; Nocita and Lowe, 1990; Hessler and Lowe, 2006), evaporitic sediments (e.g., Buick and Dunlop, 1990; Lowe and Fisher-Worrell, 1999; Sugitani et al., 2003), subaerial volcanism (Blake, 1993; Li et al., 2013), and paleosols (e.g., MacFarlane et al., 1994; Murikami et al., 2001). Much of this evidence is from Paleoarchean to Neoarchean rocks of the Kaapvaal and Pilbara cratons. However, continental emergence probably occurred only locally, and diachronously, and over a long time period, with emergent crust constituting perhaps only a small proportion of Earth's surface area by the end of the Archean (Taylor and McLennan, 1985; Stevenson and Patchett, 1990; Arndt, 1999; Vlaar, 2000; Flament et al., 2008).

An understanding of how and when continental emergence progressed is important, as widespread emergence of continents provided a means to draw down atmospheric CO_2, with concomitant increases in atmospheric oxygen (e.g., at ca. 2.5–2.3 Ga; Holland, 2002; Bekker et al., 2004; Anbar et al., 2007). However, these types of data are only available for a few cratons, such that the full picture of continental emergence across the Neoarchean-Paleoproterozoic transition remains uncertain.

Targeting the oxygen isotopic composition of zircons in Archean–Paleoproterozoic granites may provide a fruitful direction for further investigation of the antiquity and progress of continental emergence in different cratonic blocks. Assuming that postmagmatic alteration can be discounted, recognition of low $\delta^{18}O$ of inherited zircon having a range of ages necessitates the interpretation of hydrothermal alteration of the protolith. Several well-studied Phanerozoic examples indicate that extensional, rift-related silicic magmatism is conducive to extensive alteration of magmatic and associated sedimentary rocks by hydrothermal meteoric fluids and that these processes can be reflected in depleted ^{18}O isotopic compositions of the magmatic rocks and their zircon cargo (Eiler, 2001; e.g., the British Tertiary igneous province—Gilliam and Valley, 1997; Monani and Valley, 2001; Mesozoic granitoids of eastern China—Wei et al., 2002; Yellowstone intracaldera volcanic rocks—Bindeman and Valley, 2001; Bindeman et al., 2008; Timber Mount caldera complex, Nevada—Bindeman et al., 2006). In such environments, magmatism provides the heat source for circulation of meteoric hydrothermal fluids, and a shallow crustal level of emplacement allows water recharge. Low $\delta^{18}O$ detrital or xenocrystic zircon can be expected to be captured by melts in such environments. The same may be true of Archean examples. However, these may be only rarely recorded in the rock record, given the poor preservation potential of such high-level magmatic systems.

High-crustal-level granites generated in extensional tectonic settings are known from several Archean cratons (e.g., Witwatersrand Basin—Moore et al., 1993; Pilbara craton, Australia—Brauhart et al., 1998, 2000; Yilgarn craton, Australia—Hallberg, 1986). Targeting of the oxygen isotopic composition of inherited zircon in granites within these extensional geological domains, where assimilation of supracrustal material has occurred, may provide more insight into the prevalence of emergent crust through the Archean.

CONCLUSIONS

Low $\delta^{18}O$ zircons are rare, particularly in Archean rocks, but are present as 3.54–3.03 Ga inherited grains within a 2535 ± 7 Ma monzogranite of the Rum Jungle Complex, northern Australia. The zircon U-Pb, Lu-Hf, and O isotopic compositions of magmatic and inherited grains indicate that at least two distinct sources contributed to the granitic magma of the Rum Jungle Complex. The first source is interpreted to be a granitic magma, which was generated by melting of older than 3.3 Ga supracrustal rocks, as gleaned from elevated $\delta^{18}O$ (6.0‰–7.5‰) and evolved ε_{Hf} (–4.1 to –6.6) of the 2535 Ma magmatic zircon population, and consistent with the presence of metasedimentary enclaves and inherited older zircons in the granite. A second source is interpreted to be a low $\delta^{18}O$ magma formed by melting of hydrothermally altered supracrustal rocks, which were then assimilated by the intrusive Rum Jungle Complex monzogranite, based on the low $\delta^{18}O$ composition of most of the inherited zircon grains in the granite.

We propose that magmatism at shallow crustal levels provided the mechanism for generating low $\delta^{18}O$ magma by interaction with hydrothermal-meteoric water. This low $\delta^{18}O$ magma was either generated prior to 3536 Ma and then periodically remelted through the Paleoarchean to Mesoarchean, producing low $\delta^{18}O$ zircon by crystallization from melt, or low $\delta^{18}O$ zircons were generated by diffusive re-equilibration with a low $\delta^{18}O$ magma during the Neoarchean at ca. 2535 Ma. These low $\delta^{18}O$ zircons were ultimately entrained as inherited zircon in the Rum Jungle Complex monzogranite.

These data provide a minimum timing constraint for the emergence of continental crust of the North Australian craton by the end of the Archean (at 2535 Ma). Searching for the signatures of hydrothermal alteration in inherited zircon from other high-crustal-level Archean granites may yield further insights into fluid-rock interactions and the emergence of continental crust through time.

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