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Tectonic settings of continental crust formation: Insights from Pb isotopes in feldspar inclusions in zircon

Hélène Delavault[1,2], Bruno Dhuime[2], Chris J. Hawkesworth[1,2], Peter A. Cawood[1], Horst Marschall[3,4], and Edinburgh Ion Microprobe Facility (EIMF)[5]

[1]*Department of Earth Sciences, University of St. Andrews, North Street, St. Andrews KY16 9AL, UK*

[2]*School of Earth Sciences, University of Bristol, Wills Memorial Building, Queens Road, Bristol BS8 1RJ, UK*

[3]*Department of Geology and Geophysics, Woods Hole Oceanographic Institution, Woods Hole, Massachusetts 02543, USA*

[4]*Goethe-Universität Frankfurt, Institut für Geowissenschaften, Altenhöferallee 1, 60438 Frankfurt am Main, Germany*

[5]*School of GeoSciences, University of Edinburgh, West Mains Road, Edinburgh EH9 3JW, UK*

ABSTRACT

Most crustal rocks derive from pre-existing crust, and so the composition of newly generated ('juvenile') continental crust, and hence the tectonic settings of its formation, have remained difficult to determine – especially over the first billion years of Earth's evolution. Modern primitive mantle-derived magmas have distinct U/Pb ratios, depending on whether they are generated in intraplate (mean U/Pb = 0.37) or in subduction settings (mean U/Pb = 0.10). The U/Pb ratio can therefore be used as a proxy for the tectonic settings in which juvenile continental crust is generated. This paper presents a new way to see back to the U/Pb ratios of juvenile continental crust that formed 100's to 1000's of millions years ago, based on ion probe analysis of Pb isotopes in alkali feldspar and plagioclase inclusions within well-dated zircons. Pb isotope data are used to calculate the time-integrated U/Pb ratios (i.e., $^{238}U/^{204}Pb = \mu$) for the period between the Hf model age and the U-Pb crystallization age of the zircons. These time-integrated ratios reflect the composition of the juvenile continental crust at the time it was extracted from the mantle, and so they can be used as a proxy for the tectonic setting of formation of that crust. Two test samples with Proterozoic Hf model ages and Paleozoic crystallization ages have feldspar inclusions with measured Pb isotope ratios that overlap within analytical error for each sample. Sample Z7.3.1 from Antarctica has Pb isotope ratios (mean $^{206}Pb/^{204}Pb = 16.88 \pm 0.08$, 1σ) that indicate it was derived from source rocks with low U/Pb ratios (~0.11), similar to those found in subduction-related settings. Sample TEMORA 2 from Australia has more radiogenic Pb isotope ratios (mean $^{206}Pb/^{204}Pb = 19.11 \pm 0.23$, 1σ) indicative of a source with higher U/Pb ratios (~0.36), similar to magmas generated in intraplate settings. Analysis of detrital populations with a range of Hf model ages (e.g., Hadean to Phanerozoic), and for which zircons and their inclusions represent the only archive of their parent magmas, should ultimately open new avenues to our understanding of the formation and the evolution of the continental crust through time.

INTRODUCTION

The continental crust provides the principal record of the Earth's evolution during the past 4.4 billion years. Despite recent advances in analytical techniques and the explosion in the number of high quality analyses of rocks, minerals and sediments of various ages and provenance, the question of how the continental crust formed and evolved through time remains controversial. It is widely accepted that most of the rocks of the continental crust (80%–90%) available for sampling were themselves derived from pre-existing crustal rocks (e.g., Belousova et al., 2010; Roberts and Spencer, 2014). Thus the key to understanding the early stages of evolution of the continental crust is to interrogate the geochemical record of the 'juvenile' crust, i.e. at the time of its extraction from the mantle.

Dhuime et al. (2015) recently used Sr isotopes in whole rocks with a range of Nd model ages of crust formation to evaluate the time-integrated Rb/Sr and silica contents of juvenile continental crust. This study goes one step further and introduces a new geochemical tool to distinguish subduction and intraplate settings of juvenile continental crust generation. Our approach is based on the calculation of the time-integrated U/Pb ratios from Pb isotope analyses of feldspar inclusions within zircon. The U-Pb system is used because (i) measured U/Pb ratios are different in subduction-related magmas, which have mean U/Pb = 0.10, and in intraplate-related magmas that have higher ratios and a mean U/Pb = 0.37 (Fig. 1); (ii) feldspar contains sufficient Pb (typically 40–100 ppm in alkali feldspar, Doe and Tilling, 1967) for small inclusions (typically 30–40 μm) to yield precise isotope ratios when measured *in situ* with an ion probe (Table DR1); (iii) feldspar has low U/Pb ratios such that the measured Pb isotope ratios can be taken to be the same as those at the time of crystallization (Doe and Tilling, 1967; Doe et al., 1965); and (iv) the time-integrated U/Pb ratio ($\mu = {}^{238}U/{}^{204}Pb$) can be determined from both ${}^{206}Pb/{}^{204}Pb$ and ${}^{207}Pb/{}^{204}Pb$ ratios (e.g., Oversby, 1974). Which of these two ratios is used depends on the geological age, given the change from relatively more rapid ${}^{207}Pb$ ingrowth in the

Hadean and early Archean to more rapid ^{206}Pb ingrowth subsequently (e.g., Oversby, 1974; Stacey and Kramers, 1974).

Zircons preserve robust crystallization ages and their initial Hf isotope ratios constrain when juvenile magma was separated from ambient mantle. This time is commonly referred to as the Hf ‘model age’, because it is model-dependent (see a recent review in Vervoort and Kemp, 2016). Inclusions in zircon occur in rocks of all ages and provenance (Cavosie et al., 2004; Darling et al., 2009; Hopkins et al., 2008; Maas et al., 1992; Nutman and Hiess, 2009), and inclusions such as apatite and biotite preserve trace element contents that can be linked to those in the matrix, and those of the rocks from which zircons crystallized (Jennings et al., 2011; Bruand et al., 2016). Radiogenic isotope analyses in zircon and its mineral inclusions are now used here to explore the history of the source precursor of these rocks. Integration of both trace element and isotope data on mineral inclusions in the detrital zircon record will provide further insight into the complex history of crustal evolution, especially for the early Earth where the rock record is largely missing.

SAMPLES AND METHODOLOGY

This study was undertaken using two test samples, selected for a) the availability of suitable material in Bristol; b) the presence of a few large (>30–40 μm) feldspar inclusions within zircons (Figure S1 in the GSA Data Repository[1]); and c) a time period greater than 500 m.y. between the Hf model age and U-Pb crystallization age for each sample, to ensure reasonable resolution for the calculation of time-integrated parent/daughter trace element ratios in long-lived radioactive decay schemes (Dhuime et al., 2015). Z7.3.1 is from a ca. 492 m.y. monzonite intrusion from Dronning Maud Land, eastern Antarctica. The sample and the inclusions within zircons have been described by Jennings et al. (2011), and zircons have a Paleoproterozoic (ca. 1.9 Ga) depleted mantle Hf model age (Table DR2 in the GSA Data Repository[1]). The second sample is the TEMORA 2 zircon standard from the ca. 417 m.y. Middledale gabbroic diorite in the Paleozoic Lachlan Orogen, eastern Australia (Black et al., 2004). It has a younger, Meso/Neoproterozoic Hf model age of ca. 1.0 Ga (Table DR2 and Woodhead and Hergt (2005)).

Heavy mineral fractions were concentrated using heavy liquids, and inclusion-rich zircons were handpicked, mounted in epoxy, polished and examined by cathodoluminescence, backscattered electron imaging, and energy-dispersive spectrometry to identify and characterize feldspar inclusions. Pb isotope analyses on the inclusions were carried out using a Cameca ims 1270 secondary ion mass spectrometer (SIMS) at the Ion Microprobe Facility (EIMF), University of Edinburgh. Inclusions were analyzed using a 4 nA O_2^- primary ion source with a 22 keV net impact energy. The beam was focused using Köhler illumination to ensure a uniform beam density. The primary beam alignment resulted in ellipsoidal analysis pits with a maximum major axis length of ~15 μm. In order to limit peripheral contamination the spatial resolution of the analyzed area was limited further by the use of a field aperture, which restricts the secondary ion signal to the center of the analysis pit. A fixed 2700 μm field aperture was used throughout and the blank was <0.003% of the signal measured on a zircon crystal. The instrument was operated in ‘rectangular mode’ in order to maintain optimum conditions for flat topped peaks. Pb isotopes were analyzed at a mass resolution of >4000R using a peak switching routine. The $BaSiO_2$ peak was used for energy centering on each analysis and a 100eV energy window was used throughout. The Zr_2O peak at mass 196 was used to monitor potential overlap onto the zircon host. Secondary ion intensities were measured using an electron multiplier in ion counting mode. A 5 μm^2 raster was applied to the primary beam for 120 seconds in order to remove any surface contamination (e.g., ^{204}Pb) around the sputter pit. Forty cycles of analysis were then made over the following masses of interest; 196 (Zr_2O), 198($BaSiO_2$), ^{204}Pb, ^{206}Pb, ^{207}Pb, ^{208}Pb. Each analysis lasted 30 min in total. A mass fractionation correction of 2‰/amu was applied based on previous measurements of natural glass and equal atom Pb standards. The repeat measurements of $^{207}Pb/^{206}Pb$ and $^{208}Pb/^{206}Pb$ ratios of Shap Granite K-feldspar compared to values of Tyrrell et al. (2006) similarly averaged 2‰/amu.

RESULTS

Pb isotope ratios were analyzed in feldspar inclusions in six zircons: four K-feldspars and one plagioclase in sample Z7.3.1 (Antarctica), and two K-feldspars in TEMORA 2 (Australia). One K-feldspar inclusion in zircon #75 from sample Z7.3.1 was large enough to allow two analyses within the same inclusion (see light green triangles on Figure 2 and SEM picture on Figure DR1 in the GSA Data Repository[1]). All data are plotted in Figure 2 with their associated analytical precision (2σ level), and reported in Table DR1[1]. SEM pictures of the analyzed samples, before and after SIMS analyses, are provided in Figure DR1.

The feldspars in the two samples have very different Pb isotope ratios. K-feldspars of sample Z7.3.1 (Antarctica) define a narrow range of Pb isotope compositions with $^{206}Pb/^{204}Pb$ = 16.79–16.93, $^{207}Pb/^{204}Pb$ = 15.24–15.32 and $^{208}Pb/^{204}Pb$ = 36.36–36.85, with 2 s.e. ~1.3% on average for these three isotope ratios (Table DR1). The analysis of the single plagioclase inclusion is within error of those of the K-feldspar inclusions (Fig. 2), but the uncertainty on Pb isotopes ratios for the plagioclase inclusion is about twice as large as for K-feldspar (2 $s.e._{plagioclase}$ ~2.8%), because of its lower lead content (Doe and Tilling, 1967).

The abundance of K-feldspar inclusions within TEMORA 2 zircons is low, most of the inclusions are very small (i.e., <<20 μm), and only two were large enough to be analyzed (Fig. DR1). These inclusions have more radiogenic Pb isotope compositions with $^{206}Pb/^{204}Pb$ = 19.27–19.95, $^{207}Pb/^{204}Pb$ = 15.75–15.86 and $^{208}Pb/^{204}Pb$ = 39.46–40.06, with 2 s.e. errors of 1.3% (zircon #25) and 0.3% (zircon #27) on $^{206}Pb/^{204}Pb$.

DISCUSSION

A 2-Stage Model to Calculate the U/Pb Ratio of the Juvenile Continental Crust

Our approach uses a two-stage model to calculate the $\mu = {}^{238}U/^{204}Pb$ ratio of the juvenile continental crust (Fig. 3A). Stage 1 is the period of Pb isotope evolution of the juvenile continental crust, from its extraction from a depleted mantle reservoir until the formation of a derivative crustal melt from which the zircons and their inclusions subsequently crystallized. Stage 2 is the period from the crystallization of those melts to the present day. Our model therefore relies on knowing the crystallization age of the zircon, and its model age that constrains when the source of the magma from which the zircon crystallized was derived from the mantle. The measured Pb isotope ratios of the feldspar inclusions are used to back-calculate the U/Pb ratios of the juvenile continental crust (JCC) in the periods between the Hf model ages and the U-Pb crystallization age (i.e., Stage 1), following Equations 1 and 2 below:

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$$(\mu)_{JCC} = \frac{\frac{^{206}Pb}{^{204}Pb}^{Fsp}_{meas} - \frac{^{206}Pb}{^{204}Pb}^{DM}_{at\ T_{DM}}}{e^{(\lambda \times T_{DM})} - e^{(\lambda \times T_{cryst\ zrn})}} \quad (1)$$

Where $T_{cryst\ zrn}$ is the U-Pb crystallization age of the zircon, T_{DM} is the depleted mantle (DM) Hf model age of the zircon (calculated assuming a linear evolution of the DM from $\varepsilon_{Hf} = 0$ at 4560 Ma to $\varepsilon_{Hf} = 17$ at present, and an assumed $^{176}Lu/^{177}Hf = 0.015$ for the crustal source from which the zircon crystallized), λ is the decay constant of ^{238}U (1.55125×10^{-10} y^{-1}), $(^{206}Pb/^{204}Pb^{Fsp})_{meas}$ is the measured $^{206}Pb/^{204}Pb$ ratio in the feldspar inclusion, and $(^{206}Pb/^{204}Pb^{DM})_{at\ TDM}$ is the Pb isotope composition of the depleted mantle at T_{DM}. The Pb isotope evolution of the mantle was calculated using the composition of the Canyon Diablo troilite (CDT) at t_0 = 4560 Ma and a 2-stage model similar to that of Stacey and Kramers (1975), in which $\mu_{1(4560\text{-}3700Ma)} = 7.13$ and $\mu_{2(3700\text{-}0Ma)} = 9.33$ where chosen to match the Gale et al. (2013) present-day MORB average. This mantle Pb isotope evolution curve is similar to those previously published by Kramer and Tolstikhin (1997) and Kamber (2015).

$$\left(\frac{U}{Pb}\right)_{JCC} = \frac{(\mu)_{JCC}}{\left(^{M_{Pb}}/_{M_U}\right) \times \left(^{ab^{238}U}/_{ab^{204}Pb}\right)} \quad (2)$$

Where M_U and M_{Pb} are the standard atomic weights of U and Pb, and $ab^{238}U$ and $ab^{204}Pb$ are the relative abundances of ^{238}U as percent of total U and ^{204}Pb as percent of total Pb, respectively.

In principle, our 2-stage model can also be applied to the $^{207}Pb/^{204}Pb$ isotopic system. However given that the half-life of the decay of ^{235}U to ^{207}Pb is ~6 times lower than that of ^{238}U to ^{206}Pb, the evaluation of $^{235}U/^{207}Pb$ with this method appears better suited to samples with Hadean/early Archean model ages.

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Application of the Method

The Antarctica sample Z7.3.1 is particularly well suited for this study, because of the presence of large and abundant K-feldspar inclusions within the zircons (see Fig. DR1), and because there is ca. 1.4 billion years between its Hf model age and crystallization age. This period is often referred to as the ‘residence time’, and it is long enough to offer reasonable resolution on the back-calculation of the U/Pb ratios of the juvenile continental crust ($(U/Pb)_{JCC}$). The $(U/Pb)_{JCC}$ ratios calculated for five K-feldspar inclusions in this sample range from 0.11 to 0.12 (Fig. 3B), and these ratios are similar to the values observed in recent subduction-related magmas (see Fig. 1). Thus, although the host monzonite magma for the zircons (and their inclusions) crystallized in the early Paleozoic during the later stages of the collisional assembly of East and West Gondwana (Jacobs and Thomas, 2004), the monzonite magma was derived from crust that originally separated from the mantle around 1.9 billion years ago in a subduction-related environment. Flowerdew et al. (2012) analyzed K-feldspar separates from two ca. 520 m.y. granitoids from Dronning Maud Land, and they reported $^{206}Pb/^{204}Pb$ ratios of 16.45–16.54. Calculated $(U/Pb)_{JCC}$ ratios for these feldspars (ca. 0.09) also fall in the range of subduction-related magmas (Fig. 1), and so the tectonic setting of the continental crust formation indicated by both feldspars from bulk rocks and from inclusions within zircons is similar.

TEMORA 2 has a residence time of ca. 0.6 billion years that is also taken to be large enough to estimate U/Pb ratios in its source (Fig. 3A). The results are presented in Figure 3B, and the calculated $(U/Pb)_{JCC}$ ratios range from 0.34 to 0.39. These ratios are higher than the U/Pb ratios in most subduction-related magmas, but they are similar to the U/Pb ratios in intraplate magmas (Fig. 1). We conclude that the crustal source of the TEMORA magma was generated in an intraplate setting at ~1.0 Ga, consistent with paleogeographic reconstructions that place eastern Australia adjacent to western Laurentia within the Rodinian supercontinent (e.g., Hoffman, 1991).

Implications of the New Method

This study demonstrates that back-calculation of the U/Pb ratios of juvenile continental crust can be used to constrain whether new crustal material was generated in a subduction (Z7.3.1) or an intraplate (TEMORA 2) setting. Our approach highlights the potential of interrogating the isotope record of mineral inclusions encapsulated within zircons. Mineral inclusions offer a richer record of the evolution of the magmatic rocks than does the mineral zircon, which tends to crystallize at a relatively late stage in the evolution of the parent magma. Pb isotope analysis of feldspar inclusions within zircons therefore represents a new addition to the geochemical toolbox for unraveling the evolution of the continental crust through time. The use of this method on magmatic and detrital zircons with a range of Hf model ages covering Earth's history from the Hadean to Phanerozoic has the potential to open new avenues into our understanding of the large-scale evolution of the continental crust.

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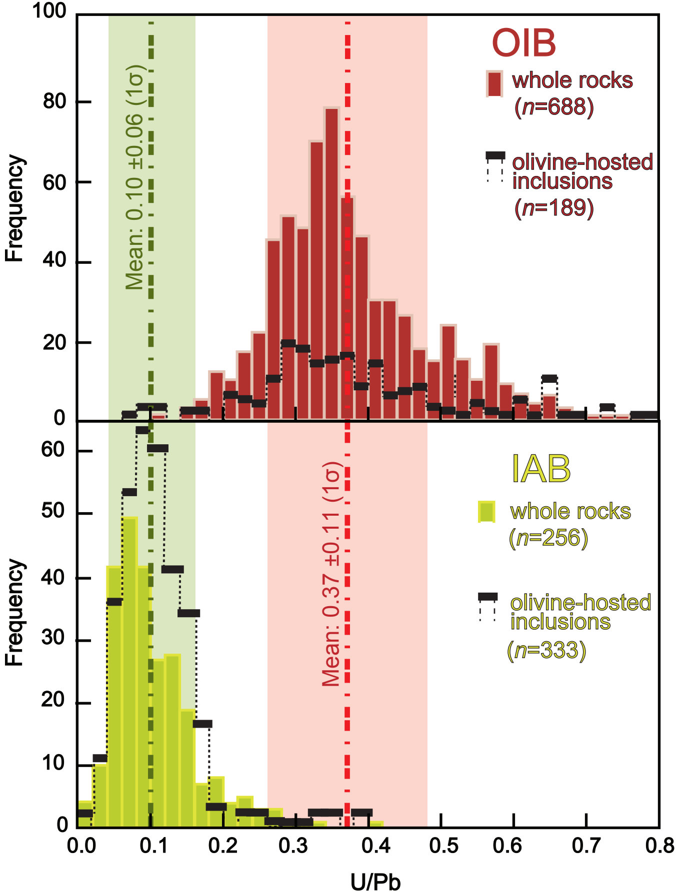
FIGURE CAPTIONS

Figure 1. Present-day distribution of U/Pb ratios in mafic rocks (filled color histograms) and olivine-hosted melt inclusions (hollow black histograms) in subduction and intraplate (OIB) settings. Data are from the GEOROC database (http://georoc.mpch-mainz.gwdg.de/georoc/), with analyses selected from primitive fresh basaltic whole rocks (i.e. SiO_2 = 45–53%, MgO = 7–25%, LOI < 5% and sum of major elements > 95%) of volcanic origin.

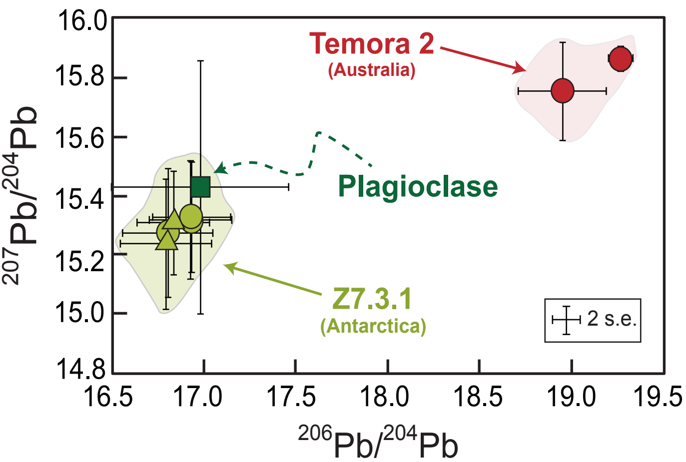
Figure 2. $^{207}Pb/^{204}Pb$ versus $^{206}Pb/^{204}Pb$ plot for mineral inclusions in sample Z7.3.1 (green) and TEMORA 2 (red) analyzed by SIMS. All but one (dark green square, plagioclase) analyses were in K-feldspars. Light green triangles show two analyses within a same inclusion in sample Z7.3.1 (zircon #75, see Figure DR1).

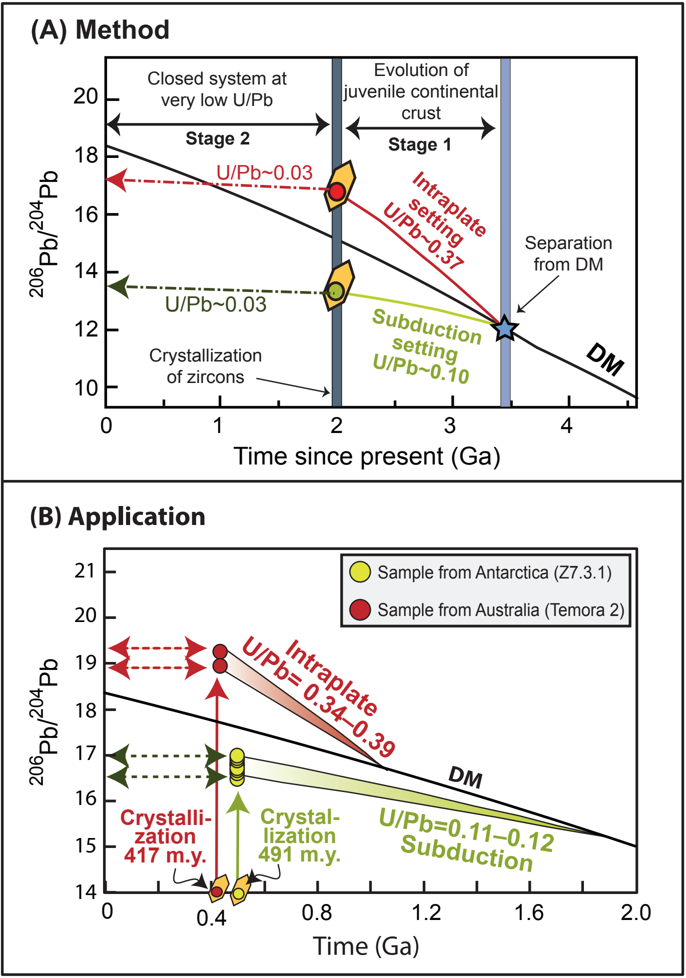
Figure 3. Evolution of the $^{206}Pb/^{204}Pb$ ratios through time. (A) Schematic representation of the calculation of the 'Stage 1' time-integrated U/Pb of juvenile continental crust, between the time of its separation from the depleted mantle (blue star) and the crystallization of the zircon host of the feldspar (Fsp) inclusions (red and green dots). Typical evolution paths for new crust generated in intraplate setting (red curve) and in subduction setting (green curve) are presented. The depleted mantle evolution is the black DM line. (B) Time-integrated U/Pb ratios calculated for two test samples Z7.3.1 (green dots) and TEMORA 2 (red dots), from the combination of U-Pb and Hf data in zircon and Pb isotope data in feldspar inclusions.

[1]GSA Data Repository item 201Xxxx, **Supplementary Figure DR1; Supplementary methods and references,** is available online at www.geosociety.org/pubs/ft20XX.htm, or on request from editing@geosociety.org.



OIB
whole rocks
(n=688)
olivine-hosted inclusions
(n=189)
Mean: 0.10 ±0.06 (1σ)
IAB
whole rocks
(n=256)
olivine-hosted inclusions
(n=333)
Mean: 0.37 ±0.11 (1σ)
Frequency
Frequency
U/Pb
100
80
60
40
20
0
60
50
40
30
20
10
0
0.0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8


Temora 2
(Australia)
Plagioclase
Z7.3.1
(Antarctica)
2 s.e.
16.0
15.8
15.6
15.4
15.2
15.0
14.8
16.5
17.0
17.5
18.0
18.5
19.0
19.5
$^{207}Pb/^{204}Pb$
$^{206}Pb/^{204}Pb$



(A) Method
Closed system at very low U/Pb
Stage 2
Evolution of juvenile continental crust
Stage 1
U/Pb~0.03
Intraplate setting U/Pb~0.37
U/Pb~0.03
Separation from DM
Subduction setting U/Pb~0.10
DM
Crystallization of zircons
20
18
16
14
12
10
0
1
2
3
4
Time since present (Ga)
$^{206}Pb/^{204}Pb$
(B) Application
Sample from Antarctica (Z7.3.1)
Sample from Australia (Temora 2)
Intraplate U/Pb= 0.34–0.39
DM
U/Pb=0.11–0.12 Subduction
Crystalli-zation 417 m.y.
Crystal-lization 491 m.y.
21
20
19
18
17
16
15
14
0
0.4
0.8
1.2
1.6
2.0
Time (Ga)
$^{206}Pb/^{204}Pb$