1	Greater Kerguelen large igneous province reveals no role for mantle plume
2	in the continental breakup of eastern Gondwana
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16 Abstract

17 The link between mantle plumes and continental breakup remain a topic of debate. Here, a new 40 Ar/ 39 Ar age of 135.9 ± 1.2 Ma (2 σ) and previous ages from the Bunbury Basalt – lava flows that are 18 19 part of the Greater Kerguelen large igneous province (LIP) – reveal that >80% of magmatism in the 20 southern Perth Basin was concomitant with the continental breakup of eastern Gondwana at ca. 137-21 136 Ma. New and existing isotope geochemical data show that only lithospheric and depleted asthenospheric sources were melted to form the Bunbury Basalt and most other early, ca. 147-124 Ma 22 23 magmatic products part of the Greater Kerguelen LIP. All lines of evidence strongly point towards 24 passive continental breakup of eastern Gondwana, including the restriction of 147-124 Ma magmatism to continental rifts, the lack of excess oceanic magmatism in this period and the >1000 25 km distance between the Kerguelen plume underneath Greater Indian lithosphere and the breakup 26 nexus. It is not possible to reconcile the influence of a thermochemical plume with the observed 27 geochemical, spatial and geochronological information. Instead, we posit that eastern Gondwana 28 breakup occurred proximal to the former suture zone associated with the ca. 550-500 Ma Kuunga 29 Orogeny between Indo-Australia and Australo-Antarctica. Enrichment of the mantle with volatiles 30 associated with subduction during the Kuunga Orogeny permitted partial melting when the 31 continental crust was sufficiently attenuated in the Early Cretaceous. Therefore, repeated and 32 33 protracted rifting of Greater India from Australo-Antarctic since the mid-Paleozoic eventually led to 34 the rupture of the continental lithosphere and to mafic magmatism at ca. 137–136 Ma, approximately along the position of the former suture zone, without the influence of a mantle plume. 35

Keywords: large igneous province; Perth Basin; Bunbury Basalt; continental flood basalts; west
 Australian margin; ⁴⁰Ar/³⁹Ar geochronology

38 1 Introduction

The links between supercontinent breakup and plume-induced large igneous provinces (LIP) remain a 39 topic of debate (Buiter and Torsvik, 2014; Condie et al., 2015; Pirajno and Santosh, 2015). The 40 41 breakup of the most recent supercontinent Gondwana still remains hotly debated in spite of the fact that much more is known about the breakup of Gondwana than other supercontinents like Rodinia or 42 Nuna (Ma et al., 2018; Olierook et al., 2016; Veevers, 2004). Part of this debate has been fueled by an 43 44 uncertain relationship between the breakup of Greater India and Australo-Antarctica in eastern Gondwana and the Greater Kerguelen large igneous province, purported to have been derived from 45 46 the Kerguelen mantle plume. In particular, the early magmatic products that are potentially 47 synchronous with continental breakup remain poorly constrained.

Together with the Tethyan Himalaya igneous province, the Bunbury Basalt are the earliest recognized 48 49 volcanic products that may have been driven by the Kerguelen mantle plume (Frey et al., 1996; 50 Olierook et al., 2016; Zhu et al., 2009). Previous geochronological and 3D modelling work has 51 established that the Bunbury Basalt erupted in at least four pulses in two paleovalleys (Coffin et al., 52 2002; Olierook et al., 2016; Olierook et al., 2015b). The western Bunbury Paleovalley hosts three of these four flows that were dated at 136.96 \pm 0.43 Ma, 132.71 \pm 0.43 Ma and 130.45 \pm 0.82 Ma 53 (Olierook et al., 2016). The oldest of these at ca. 137 Ma are synchronous with the breakup of Greater 54 India and Australo-Antarctica at 137-136 Ma (Gibbons et al., 2012). The fourth lava flow event is 55 present in the eastern Donnybrook Paleovalley and accounts for >50% of the preserved volume of the 56 57 Bunbury Basalt (Olierook et al., 2015b). However, it has not yet been dated successfully. Thus, establishing a clear link between continental breakup and the Bunbury Basalt and, in turn, whether a 58 59 mantle plume was implicated requires a comprehensive understanding of the entire Bunbury Basalt, 60 not just the western portion. Isotope geochemical data paint an enigmatic picture in that most of the 61 Greater Kerguelen large igneous province, including the main Southern and Central Kerguelen Page 3

Plateau magmatic constructs, do not show any evidence of melt from the Kerguelen plume head but only comprise depleted asthenosphere and lithospheric products (Frey et al., 1996; Ingle et al., 2004; Liu et al., 2015; Ma et al., 2018; Olierook et al., 2016; Olierook et al., 2017). This picture is also significantly fragmented in that only two isotope geochemical analyses are from the older ca. 137 Ma flows in the Bunbury Basalt (Frey et al., 1996). There is little evidence for any evolution of mantle sources for the Bunbury Basalt, which again precludes a definitive link to whether a mantle plume was necessary.

69 To solve the conundrum on whether the Kerguelen mantle plume played a role in the production of 70 the Bunbury Basalt and the breakup of eastern Gondwana, we provide five new step-heated plagioclase ⁴⁰Ar/³⁹Ar ages from the eastern paleovalley of the Bunbury Basalt lava flows. 71 Geochronological data is complemented with new Sr-Nd-Pb isotopic geochemical data from all 72 known Bunbury Basalt lava flows to assess the evolution of mantle sources. Together, 73 74 geochronological and isotope geochemical data allow a robust evaluation of the drivers for magmatism in onshore Western Australia and, ultimately, whether a magma plume was involved in 75 triggering the breakup of eastern Gondwana. 76

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78 2 Geological Setting of the Bunbury Basalt

The Bunbury Basalt comprises a series of lava flows and possible scarce sheeted intrusive rocks in the southern Perth Basin of Western Australia (see Olierook and Timms, 2016; Olierook et al., 2015c and references therein for recent tectonic and stratigraphic reviews of the Perth Basin). During the Early Cretaceous, the Bunbury Basalt lava flows erupted from vent sites postulated to lie at intersections of minor faults with the basin-bounding Darling Fault (Fig. 1b; Olierook et al., 2015b). They were extruded as four distinct lava flow events into two deeply-incised paleovalleys that form the Valanginian breakup unconformity (Olierook et al., 2015b). Page 4

The western paleovalley, the Bunbury Paleovalley, host three of the four flows and preserves some 86 ~43 km³ of mafic lava. Two of these flows in the Bunbury Paleovalley – a lower and upper flow – 87 were dated at ca. 137 and 133 Ma, respectively, and comprise the bulk of the \sim 43 km³ (Olierook et al., 88 2016; Olierook et al., 2015b). Up to 5m of interflow siltstone and claystone separate the two flows in 89 90 the Bunbury Paleovalley (Backhouse, 1988). The upper flow only defines the top ~ 10 m of the western paleovalley (Olierook et al., 2015b). On the basis of an average thickness of ~30 m of mafic 91 lava in the western paleovalley (Olierook et al., 2015b), the upper flow comprises ~ 14 km³ and the 92 93 lower flow 29 km³ of mafic lava. The third lava flow in the Bunbury Paleovalley is situated at its southern end along the southern coast at a locality called Black Point (Olierook et al., 2015b). Black 94 Point was originally thought to record one of the two flows within the Bunbury Paleovalley (Olierook 95 et al., 2015b) but was subsequently dated at ca. 130.5 Ma, revealing a distinct age and geochemistry 96 97 (Olierook et al., 2016).

98 The eastern paleovalley, the Donnybrook Paleovalley, hosts ~47 km³ of mafic lava in a continuous
99 layer averaging 70 m in thickness, which is interpreted to be a single flow (Olierook et al., 2015b). No
100 interflow sediment is present.

101 Possibly comagmatic, deep-seated, discontinuous dolerite bodies intruded into Permian and Triassic sedimentary rocks at depths of 2700-4600 m below sea level (Olierook et al., 2015b; Poynton and 102 Hollams, 1980; Union Oil Development Corporation, 1972; Wharton, 1981; Williams and Nicholls, 103 1966). The intrusive bodies have been analyzed using plagioclase 40 Ar/ 39 Ar geochronology but failed 104 to yield a reliable age on all occasions (Olierook et al., 2016). The age spectra of the intrusive bodies 105 are compatible with scenarios involving (1) the presence of excess 40 Ar* affecting Early Cretaceous 106 sills, or (2) a Precambrian crystallization age, thus unrelated to the emplacement of the Greater 107 108 Kerguelen large igneous province.

109 The vast majority of age, isotopic and chemical data is taken from the western of the two channels, 110 the Bunbury Paleovalley (Coffin et al., 2002; Frey et al., 1996; Olierook et al., 2016). The Bunbury Paleovalley shows an overall negative magnetic anomaly with respect to the present-day magnetic 111 field, implying that at least the thicker lower flow erupted during a period of reversed magnetic 112 113 polarity (Olierook et al., 2015b). The eastern Donnybrook Paleovalley has a positive magnetic anomaly, indicating that the magmas were emplaced during a period of normal magnetic polarity; i.e., 114 indicating an eruption time that was temporally distinct to both lava flows in the Bunbury Paleovalley. 115 However, there were ~35 magnetic reversals in the Valanginian and Hauterivian (139.4–130.8 Ma), 116 with periods of normal or reversed polarity lasting as little as ~20 ka (Ogg, 2012). Our ${}^{40}\text{Ar}/{}^{39}\text{Ar}$ ages 117 - with analytical uncertainties of >0.4 Ma - will therefore not necessarily resolve age differences for 118 flows erupted during periods of opposing magnetic polarity. It is presently uncertain what the 119 120 temporal relationship between the two paleovalleys is, and how the paleodrainage system evolved 121 during continental breakup (Olierook et al., 2015b).

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123 **3** Sample selection

For ⁴⁰Ar/³⁹Ar geochronology, five ~20 cm-long, quarter-core, basaltic andesite samples (BN25-124 BN29) were collected from two drill cores that penetrate through a single ~70 m thick lava flow in the 125 Donnybrook Paleovalley (Fig. 1b, Table 1, Supplementary Table A). These two drill cores, DDB-7 126 and DDB-8, together with a few weathered outcrop localities, are the only rocks available for 127 128 sampling that unambiguously belong to the Donnybrook Paleovalley (Morant, 1988; Playford et al., 1976). Differentiating between the Bunbury and Donnybrook paleovalleys relies upon magnetic data 129 and drill hole constraints that were used to build the 3D model of the Bunbury Basalt (Olierook et al., 130 2015b). Both DDB-7 and DDB-8 clearly belong to the Donnybrook Paleovalley, in which the lava 131 flow has a normal magnetic polarity. Hand specimens indicate the Bunbury Basalt in DDB-7 and 132 Page 6

DDB-8 comprises dark grey, sparsely vesicular, porphyritic mafic rocks with fine-grained groundmass. All samples contain ~20% plagioclase phenocrysts up to 250 µm long with a groundmass of plagioclase, pyroxene and opaque minerals. The only noticeable variability in samples is the absence of pyrite towards the top, and progressively more pyrite towards the base of the flow.

For Sr–Nd–Pb isotope geochemistry, nine mafic lava samples from the Bunbury Basalt were analyzed, including four of the five newly collected samples (see Supplementary Table B). The other five samples were selected from sites where reliable 40 Ar/ 39 Ar ages were previously obtained (Olierook et al., 2016), including two samples from Donnelly River (ca. 137 Ma), one sample from each of the lower and upper flow of the Gelorup Quarry (ca. 137 Ma and 133 Ma, respectively) and one sample from Black Point (ca. 130 Ma, Fig. 1).

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144 **4** Analytical techniques

145 4.1 ${}^{40}Ar/{}^{39}Ar$ geochronology

All five samples from both drill cores were dated using ⁴⁰Ar/³⁹Ar geochronology. Samples were 146 crushed and minerals were separated using a Frantz magnetic separator. The non-magnetic fraction 147 was carefully hand-picked under a binocular microscope to select unaltered, optically transparent, 148 125-212 µm-size plagioclase grains. In all cases, picked plagioclase crystals were optically 149 transparent when viewed under a binocular microscope and free from inclusions. The selected 150 151 plagioclase grains were leached in diluted (5N) HF for one minute and then thoroughly rinsed with distilled water in an ultrasonic cleaner. Samples were loaded into several large wells of 1.9 cm 152 153 diameter and 0.3 cm depth aluminum discs. The discs were Cd-shielded (to minimize undesirable 154 nuclear interference reactions) and irradiated for 40 hours in the TRIGA nuclear reactor (Colorado, USA) in central position. 155

The ⁴⁰Ar/³⁹Ar analyses were performed on an ARGUS VI at the Western Australian Argon Isotope 156 Facility at Curtin University. The ages were calculated relative to GA1550 biotite neutron flux 157 monitor, for which an age of 99.738 \pm 0.104 Ma was used (Renne et al., 2011). The mean J-values 158 computed from standard grains are $0.01085900 \pm 0.00000923$ (0.08%), determined as the average and 159 160 standard deviation of J-values of the small wells for the irradiation disc. Mass discrimination was calculated using a power law and monitored using an automated air pipette, providing a mean value of 161 0.992403 ± 0.000397 (0.04%) per dalton. The correction factors for interfering isotopes were 162 $({}^{39}\text{Ar}/{}^{37}\text{Ar})_{Ca} = 6.95 \times 10^{-4} (\pm 1.3\%), ({}^{36}\text{Ar}/{}^{37}\text{Ar})_{Ca} = 2.65 \times 10^{-4} (\pm 0.8\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%), ({}^{36}\text{Ar}/{}^{37}\text{Ar})_{Ca} = 2.65 \times 10^{-4} (\pm 0.8\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%), ({}^{36}\text{Ar}/{}^{37}\text{Ar})_{Ca} = 2.65 \times 10^{-4} (\pm 0.8\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%), ({}^{36}\text{Ar}/{}^{37}\text{Ar})_{Ca} = 2.65 \times 10^{-4} (\pm 0.8\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%), ({}^{36}\text{Ar}/{}^{37}\text{Ar})_{Ca} = 2.65 \times 10^{-4} (\pm 0.8\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%), ({}^{36}\text{Ar}/{}^{37}\text{Ar})_{Ca} = 2.65 \times 10^{-4} (\pm 0.8\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%) \text{ and } ({}^{40}\text{Ar}/{}^{39}\text{Ar})_{K} = 7.30 \times 10^{-4} (\pm 1.3\%) \text{ and } ({}^{40}$ 163 12%). Modern atmospheric argon was considered to be 298.56 ± 0.31 (Lee et al., 2006). Additional 164 constants, ratios and all ⁴⁰Ar/³⁹Ar data may be found in Supplementary Table A. 165

166 Our criteria for the determination of plateaus are as follows: (i) plateaus must include at least 70% of ³⁹Ar, and (ii) the plateaus should be distributed over a minimum of three consecutive steps agreeing at 168 95% confidence level and satisfying a probability of fit (*p*) of at least 0.05. Plateau ages are given at 169 the 2σ level and are calculated using the mean of all the plateau steps, each weighted by the inverse 170 variance of their individual analytical error. All sources of uncertainties are included in the 171 calculation, including the decay constants.

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4.2 Sr–Nd–Pb isotope geochemistry

Radiogenic isotope ratios of Sr (⁸⁷Sr/⁸⁶Sr), Nd (¹⁴³Nd/¹⁴⁴Nd) and Pb (²⁰⁶Pb/²⁰⁴Pb, ²⁰⁷Pb/²⁰⁴Pb,
²⁰⁸Pb/²⁰⁴Pb) were measured at the University of Geneva, Switzerland. The method is described in
detail in Béguelin et al. (2015). Between 100 and 120 mg of whole rock powder were dissolved for 7
days in Savillex® Teflon vials using 4 ml of concentrated HF and 1 ml of HNO₃ 14M, at a
temperature of 140 °C and with the help of ultrasonication for 30 minutes twice a day. Subsequently,
samples were dried and re-dissolved for 3 days (also with 30 minutes ultrasonication twice a day) in 3
Page 8

180 ml of HNO₃ 14M and dried again. Sr, Nd and Pb were then separated using cascade columns with Sr-181 Spec, TRU-Spec and Ln-Spec resins according to a protocol modified from Pin et al. (1994). Finally, the material was redissolved in 2% HNO₃ solutions and ratios were measured using a Thermo 182 Neptune PLUS Multi-Collector ICP-MS in static mode. Ratios used to monitor internal fractionation 183 were: 88 Sr/ 86 Sr = 8.375209 for the 87 Sr/ 86 Sr ratio, 146 Nd/ 144 Nd = 0.7219 for the 143 Nd/ 144 Nd ratio and 184 203 Tl/ 205 Tl = 0.418922 for the three Pb ratios (a Tl standard was added to the solution). External 185 standards used were SRM987 (87 Sr/ 86 Sr = 0.710248, long-term external reproducibility: 10 ppm, 1 σ ; 186 McArthur et al., 2001), JNdi-1 (143 Nd/ 144 Nd = 0.512115; long-term external reproducibility: 10 ppm, 187 1σ; Tanaka et al., 2000), and SRM981 for Pb (long-term 1σ external reproducibility of 0.0082% for 188 ²⁰⁶Pb/²⁰⁴Pb, 0.0064% for ²⁰⁷Pb/²⁰⁴Pb and 0.0094% for ²⁰⁸Pb/²⁰⁴Pb; Baker et al., 2004). ⁸⁷Sr/⁸⁶Sr, 189 ¹⁴³Nd/¹⁴⁴Nd and Pb isotope ratios were further corrected for external fractionation (due to a systematic 190 difference between measured and accepted standard ratios) by a value of -0.021‰, +0.051‰ and 191 +0.36‰ amu respectively. Interferences at masses 84 (⁸⁴Kr), 86 (⁸⁶Kr) and 87 (⁸⁷Rb) were corrected 192 by monitoring ⁸³Kr and ⁸⁵Rb, ¹⁴⁴Sm interference on ¹⁴⁴Nd was monitored on the mass ¹⁴⁷Sm and 193 corrected by using a ¹⁴⁴Sm/¹⁴⁷Sm value of 0.206700 and ²⁰⁴Hg interference on ²⁰⁴Pb was corrected by 194 monitoring ²⁰²Hg. Total procedural blanks were <500 pg for Pb and <100 pg for Sr and Nd which are 195 196 insignificant compared to the amounts of these elements purified from the investigated whole rock 197 samples.

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199 5 Results
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200 5.1 ${}^{40}Ar/{}^{39}Ar$ age results of the Donnybrook Paleovalley, Bunbury Basalt

Five plateau ages were obtained from six analyses, with ages ranging between 138.7 ± 2.4 and 133.7

 ± 2.8 (2 σ , Table 2, Fig. 2). All ages are indistinguishable within error. Given that these were from the

same flow, a weighted mean of the plateau steps (n = 92) was calculated, yielding an age of 135.9 ± Page 9

1.2 (MSWD = 0.63; p = 1.00; Fig. 3). Ratios of K/Ca (0.0017–0.0018; Ca/K = 560–590) are indicative of fresh plagioclase rather than alteration products (Fig. 2; Verati and Jourdan, 2014). Such low K/Ca values explain why the individual plateau ages have relatively low precision compared to more 'conventional' LIP plagioclase compositions despite using similar analytical conditions (e.g., 181.4 \pm 0.4 Ma for plagioclase dolerites from the Ferrar LIP; Ware and Jourdan, 2018). The age of 135.9 \pm 1.2 Ma (2 σ) is considered the crystallization age of the eruption that extruded into the Donnybrook Paleovalley.

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212 5.2 Sr–Nd–Pb isotope geochemical results from the Bunbury Basalt

213 The ca. 136 Ma Donnybrook Paleovalley (BN25, 26, 28, 29) shows Sr-Nd-Pb isotopic ratios that are very similar to Black Point (ca. 130 Ma, Figs. 1, 4) and are thus considered part of the 'Gosselin-type' 214 geochemical group (Gosselin is an alternative name for Black Point; Frey et al., 1996). Both the upper 215 (BN3, ca. 133 Ma) and lower (BN4, ca. 137 Ma) flows in Gelorup Quarry are similar to the 216 217 'Casuarina-type' magmas, named after Casuarina Point (Fig. 1; Frey et al., 1996). The Gosselin-type analyses have higher 87 Sr/ 86 Sr (>0.706), lower 143 Nd/ 144 Nd (<0.5125) and higher 207 Pb/ 204 Pb ratios 218 (>15.6) compared to the Casuarina-type analyses (Fig. 4). Sample BN22 and BN10 from the Donnelly 219 220 River and Black Point, respectively, are very similar in Sr-Nd-Pb composition to previously acquired samples from these sites (Frey et al., 1996). Sample BN21 shows anomalously high measured 221 87 Sr/ 86 Sr (~0.716), 206 Pb/ 204 Pb (~18.7) and 208 Pb/ 204 Pb (~40.1) isotopic ratios. Given BN21 is from the 222 same unit as BN22 and that BN22 yielded comparable isotopic ratios to previous studies (Frey et al., 223 1996), we consider the analyses from BN21 unreliable. 224

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226 **6 Discussion**

227 6.1 Age and distribution of the Bunbury Basalt

Prior to this study, the Bunbury Basalt had been characterized by reliable ⁴⁰Ar/³⁹Ar ages from four 228 localities, yielding three different populations at ca. 137 Ma, 133 Ma and 130.5 Ma (Fig. 1; Coffin et 229 al., 2002; Olierook et al., 2016). The oldest age population was obtained from the lower of the two 230 231 flows of the western Bunbury Paleovalley at both its southern extremity and near its northern margin (Olierook et al., 2015b). Here, we add a robust age of 135.9 ± 1.2 Ma for the Donnybrook Paleovallev 232 (Fig. 2). Although this new age is indistinguishable from the oldest age of 136.96 ± 0.43 Ma obtained 233 234 on the lower flow in the Bunbury Paleovalley (Olierook et al., 2016), magnetic polarity differences 235 indicate that these represent separate lava flows. Nevertheless, most Bunbury Basalt magmatism occurred at ca. 137–136 Ma and was spatially ubiquitous across the southern Perth Basin, both within 236 the Donnybrook Paleovalley and as part of the lower flow of the Bunbury Paleovalley. A first order 237 volume calculation using the model of Olierook et al. (2015b) shows that the preserved volume of 238 239 magmatism erupted at 137–136 Ma is >80% of the total volume of the Bunbury Basalt (73 km³). A significant temporal gap separates the ca. 137–136 Ma lava flows from the younger ca. 133 and 130.5 240 241 Ma eruptive products. The younger magmatic events were also spatially restricted. The ca. 133 Ma 242 younger lava flows were confined to the western of the two paleovalleys, which can be traced via aeromagnetic and drillhole data from north to south. The youngest eruptions of ca. 130.5 Ma were 243 244 from an isolated locality, Black Point, which probably represents the final burst of volcanism in onshore Australia. 245

The configuration of the paleodrainage system into which the Bunbury Basalt flowed also provides evidence of the migration of continental breakup. The paleodrainage system was initially widely laterally distributed across the southern Perth Basin at ca. 137–136 Ma (Fig. 5a). At ca. 133 Ma, magmatism was confined to the western paleovalley. By ca. 130.5 Ma, magmatism was restricted to Black Point. The implication here is that the drainage system preferentially avulsed towards the west

and south. This drainage avulsion probably occurred as fault blocks tilted westwards and southwards
towards the breakup nexus (Figs. 1 and 5; Olierook et al., 2019). Recent age data from the Naturaliste
Plateau ranging from ca. 132 to ≥128 Ma further support the hypothesis that magmatism proceeded
from east (onshore southwestern Australia) to west (offshore western Australia, Fig. 5b; Direen et al.,
2017; Olierook et al., 2017). Despite the spatial and volumetric progression of magmatism from ca.
137–130 Ma, there is no systematic evolution of mantle sources with time (Fig. 4). Therefore, the
proportion of melt sources is independent of the spatial and volumetric magmatic progression.

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259 6.2 Was the Kerguelen plume required to produce the Bunbury Basalt?

260 The link between the Kerguelen LIP and Bunbury Basalt was originally made on near-synchronicity, similar geochemical signatures and relatively close spatial association (Frey et al., 1996). However, 261 recent ${}^{40}\text{Ar}/{}^{39}\text{Ar}$ age data (Olierook et al., 2016; this study) have since been refined to reveal a ~10 262 Myr age gap between the last igneous activity associated with the Bunbury Basalt (ca. 130 Ma) and 263 264 the earliest eruption dated for the Southern Kerguelen Plateau (ca. 120 Ma; Fig. 6; Duncan, 2002). Moreover, there is also a paucity of age data in other eastern Gondwanan provinces during 130–120 265 Ma, with the sole exception of the Wallaby Plateau (ca. 124 Ma, Fig. 6; Olierook et al., 2015a). 266 267 Geochemically, it is well established that the Bunbury Basalt shares isotopic similarities with Early 268 Cretaceous magmatism from the Kerguelen Plateau, Broken Ridge and other circum-Indian Ocean magmatic products (Fig. 7; Olierook et al., 2017). However, the involvement of any plume melt has 269 yet to be demonstrated for most magmatic products associated with the Kerguelen LIP prior to ca. 100 270 Ma. The exception is the ca. 147–115 Ma Tethyan Himalaya igneous province, where a mixture of 271 272 Greater Indian continental crust and Kerguelen plume melt is likely (Fig. 7; Ghatak and Basu, 2011; 273 Liu et al., 2015; Olierook et al., 2017).

274 The Tethyan Himalaya igneous province holds the key for ascertaining the composition of the 275 Kerguelen plume head given that independently constrained plate-plume reconstructions place the plume head underneath northern Greater India, directly below the Tethyan Himalaya (Doubrovine et 276 al., 2012; Gibbons et al., 2013; Watson et al., 2016; Whittaker et al., 2013). The simplest method to 277 278 explain the Sr-Nd-Pb isotopic composition for the ca. 147-115 Ma Tethyan Himalaya is a mixture between Greater Indian continental crust (Ghatak and Basu, 2011) and the Kerguelen plume, 279 specifically the composition attributed to the Kerguelen plume tail (see discussion in Olierook et al., 280 2017; Weis et al., 1993). It follows that the Kerguelen plume head and plume tail have the same 281 composition, and that significant proportions of plume melt are only present in the post-100 Ma 282 Greater Kerguelen LIP (except the Tethyan Himalaya). Alternative methods of invoking a plume head 283 284 composition between a lithospheric and asthenospheric end-member are possible (Ingle et al., 2003) but are not necessary to explain the Sr-Nd-Pb isotopic compositions for the pre-100 Ma Greater 285 Kerguelen LIP. Moreover, these models were made prior to and fail to account for the composition of 286 the Tethyan Himalaya igneous province. Therefore, the simplest and most plausible explanation is 287 288 that all other pre-100 Ma products can be explained by a mixture of asthenospheric and lithospheric 289 (mantle and/or crust) sources without any thermochemical plume involvement (Fig. 7; Olierook et al., 290 2017).

One other crucial aspect that is poorly explained by a mantle plume model is the distribution of the early (pre-120 Ma) magmatic products. If a mantle plume is invoked, the magmatic products in the Tethyan Himalaya, the eastern Indian Ocean and onshore Western Australia require a ~2000 km diameter plume head (Fig. 8). Such distances are not impossible for mantle plumes but are at odds with where magmatism occurs and, just as importantly, where magmatism is absent (Figs. 1 and 8).

All early magmatic products of the Greater Kerguelen LIP are exclusively associated with continental
 rifts, including the ca. 147–115 Ma Comei province (Chen et al., 2018), the 137–130 Ma Bunbury
 Page 13

Basalt (Olierook et al., 2016), the ca. 130 Ma Naturaliste Plateau (Direen et al., 2017; Olierook et al.,
2017) and the ca. 124 Ma Wallaby Plateau (Olierook et al., 2015a). All of these regions eventually
formed into continental margins or microcontinents, attesting to the importance of continental and
subcontinental lithosphere in magmatic production (Olierook et al., 2017).

302 In contrast, the nascent eastern Indian Ocean that separated all these distinct (micro)continental provinces exhibits a bathymetry that is typical for mid-oceanic crustal production without excess 303 magmatism caused by a mantle plume (White et al., 1992). Using the $1^{\circ} \times 1^{\circ}$ crustal thickness model 304 of Laske et al. (2013) (CRUST 1.0: https://igppweb.ucsd.edu/~gabi/crust1.html), we compute an 305 oceanic crustal thickness of 7.1 ± 0.8 km (2σ) for the Perth Abyssal Plain (i.e., the earliest portions of 306 oceanic crust following the breakup of eastern Gondwana), negating areas covered by any 307 308 microcontinents. Such oceanic crustal thickness is compatible with standard 7 ± 1 km oceanic crustal 309 thickness formed at medium to fast-spreading mid-ocean ridges (Laske et al., 2013; Meier et al., 2007; White et al., 1992; Young et al., 2018). The implication is that excessive magmatism in the oceanic 310 realm did not occur during the breakup of Gondwana, such as would be caused by the interaction of a 311 312 mantle plume and oceanic crust (Bown and White, 1994). Although it is likely that a mantle plume was present underneath Greater India as far back as 147 Ma (Figs. 7, 8; Shi et al., 2018), it is difficult 313 314 to envisage how magmatism was produced in southwest Australia and at microcontinents in the 315 eastern Indian Ocean without additional magmatism in the nascent ocean basin. As the oceanic crust 316 is significantly thinner than the continental crust, any thermal anomaly underneath Greater India would not be able to produce magmatism in southwest Australia as it would first have to impinge and 317 318 decompress in the nascent Indian Ocean lithosphere. Therefore, if the plume-plate models are correct 319 in placing the Kerguelen mantle plume under Greater India until at least ca. 100 Ma (e.g., Doubrovine 320 et al., 2012), then the 137–130 Ma Bunbury Basalt, the ca. 130 Ma Naturaliste Plateau and the ca. 124 321 Ma Wallaby Plateau could not have had a mantle plume implicated in their geneses.

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323 *6.3*

Causes of the breakup of eastern Gondwana

If a thermochemical plume was not responsible for the magmatic products in southwestern Australia and in the eastern Indian Ocean, it follows that a mantle plume did not trigger the breakup of eastern Gondwana at 137–136 Ma. Yet, the synchronicity of the earliest magmatism in southwestern Australia and continental breakup of Greater India and Australo–Antarctica at ca. 137–136 Ma, and migration of paleodrainage systems towards the breakup nexus, implies that the cause of magmatism was also the cause of continental breakup (Fig. 8).

Rifting in the Perth Basin first occurred in the mid-Paleozoic (Hocking, 1991; Markwitz et al., 2017). 330 331 Since then, repeated and protracted rifting in the Perth Basin has meant that the lithosphere between 332 Greater India and Australo-Antarctica had already been weakened well before the breakup of these continental bodies in the Early Cretaceous (Harris, 1994; Olierook et al., 2015c; Song and Cawood, 333 2000). The earliest magmatic products in southwestern Australia could instead be derived solely from 334 335 passive rifting, leading to decompression of the asthenosphere and subsequent emplacement of tholeiitic Bunbury Basalt. However, the present-day thickness of the continental crust in southwestern 336 Australia is insufficiently attenuated ($\beta \approx 1.4$) to facilitate decompression melting of anhydrous, upper 337 338 mantle peridotite (Chappell and Kusznir, 2008; Olierook et al., 2016). Therefore, either an additional 339 heat source or a fertile mantle is necessary to produce the Bunbury Basalt and trigger the breakup of 340 eastern Gondwana.

Other than mantle plumes, one plausible mechanism to elevate mantle temperatures is via mantle warming under supercontinents, which can elevate upper mantle temperatures by >100 °C (Coltice et al., 2007) and may produce large igneous provinces as extensive as the Central Atlantic magmatic province (Marzoli et al., 2018). Eastern Gondwana had formed by ca. 500 Ma during the Kuunga Orogeny (Daczko et al., 2018; Halpin et al., 2017), so enough time had elapsed to elevate mantle Page 15 temperatures underneath the Gondwanan supercontinent. However, mantle warming under
supercontinents only occurs distal (>1000 km) from all oceanic crust, so that the mantle is effectively
insulated (Coltice et al., 2007). Greater India's and Australia's northern margins were adjacent to the
Neotethys Ocean and <700 km from the site of continental breakup in the Perth Abyssal Plain (Fig.
8). Thus, mantle warming is probably not a plausible mechanism to explain the production of
Bunbury Basalt and other magmatic provinces part of the Greater Kerguelen LIP.

Rather than an additional heat source, we propose that the mantle underneath eastern Gondwana was 352 353 hydrated, facilitating decompression melting at significantly lower temperatures to produce the 354 Bunbury Basalt and aid in promoting continental breakup (Olierook et al., 2016). Geochemical evidence such as Nb and Ta depletions, enrichment in large ion lithophile elements and high 355 ²⁰⁷Pb/²⁰⁶Pb for ²⁰⁶Pb/²⁰⁴Pb isotopic ratios are all compatible with an ancient subcontinental lithospheric 356 357 mantle that has been modified and made fertile through subduction processes (Murphy et al., 2002; 358 Olierook et al., 2017). The key to the position of continental breakup is the location of the original suture of Indo-Antarctica with Australo-Antarctica along the Pinjarra Orogen during the Kuunga 359 Orogeny (Aitken et al., 2016; Fitzsimons, 2003; Halpin et al., 2017). The inherent lithospheric 360 361 structural discontinuity between these two terranes was probably the primary reason why continental breakup occurred here (Buiter and Torsvik, 2014; Mole et al., 2014; Will and Frimmel, 2018). The 362 363 Pinjarra Orogen suture was a zone of lithospheric weakness that was easier to exploit than any other parts of the far thicker Greater Indian, Australian or Antarctic lithosphere. Continental breakup 364 thereby employed approximately the same zone as where it was originally sutured together. 365 366 Additionally, either collision associated with the Kuunga Orogeny (Halpin et al., 2017) or the subduction along the entire southern margin of Gondwana (Veevers, 2004) enriched the lithospheric 367 mantle in volatiles, as evidenced by significant lithospheric geochemical components in the Greater 368 Kerguelen LIP magmatic products (Fig. 7; Olierook et al., 2017). We advocate that this enrichment 369 370 was sufficient in promoting partial melting of the mantle without necessitating an additional heat Page 16

source to induce melting. Successive rifting events from the mid-Paleozoic through to the Early
Cretaceous eventually attenuated the continental lithosphere sufficiently to permit partial melting of
this enriched lithospheric mantle, all of which eventually led to the passive continental breakup of
eastern Gondwana at ca. 137–136 Ma without a thermochemical plume.

375

376 **7** Conclusions

377 New and previously published geochronological data from the Bunbury Basalt - lava flows that are 378 part of the Greater Kerguelen large igneous province - reveal that >80% of magmatism in the southern Perth Basin was concomitant with the continental breakup of eastern Gondwana at ca. 137-379 136 Ma. Radiogenic isotope data show that only lithospheric and depleted asthenospheric sources 380 381 were melted to form the Bunbury Basalt and other early circum-eastern Gondwana magmatic products 382 part of the Greater Kerguelen LIP (with the Tethyan Himalaya as an exception). The >1000 km 383 distance between the Kerguelen plume underneath Greater Indian lithosphere and the breakup nexus, 384 the restriction of 147-124 Ma magmatism to continental rifts, and the lack of excess oceanic magmatism in this period all strongly point towards a passive continental breakup of eastern 385 Gondwana. It is not possible to reconcile the influence of a thermochemical plume with the observed 386 geochemical, spatial and geochronological information. We propose that the position of eastern 387 Gondwana breakup is strongly linked to the former location of the suture zone associated with the ca. 388 550-500 Ma Kuunga Orogeny between Indo-Australia and Australo-Antarctica. Enrichment of the 389 390 mantle with volatiles associated with subduction during the Kuunga Orogeny permitted partial melting when attenuation of the continental crust was sufficient in the Early Cretaceous. Ultimately, 391 repeated and protracted rifting of Greater India from Australo-Antarctic since the mid-Paleozoic 392 393 eventually led to the rupture of the continental lithosphere and mafic magmatism at ca. 137-136 Ma,

approximately along the position of the former suture zone, without the influence of the Kerguelenmantle plume.

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607

608 Figure captions

Fig. 1: (a) Bathymetric map of the Indian Ocean in the vicinity of the margin of Western Australia, after Amante & Eakins (2009). Major bathymetric features of the Indian Ocean, and locations and ages of dated extrusive rocks recovered from DSDP, ODP, IODP and industry volcanic basement sites are indicated after Olierook et al. (2017). (b) Map of onshore southwestern Australia showing the

position and ages of the Bunbury Basalt, modified from Olierook et al. (2015b) with age data fromOlierook et al. (2016) and this study.

615

616 Fig. 2: 40 Ar/ 39 Ar apparent age and related K/Ca ratio spectra of the plagioclase separates versus the 617 cumulative percentage of 39 Ar released. Errors on plateau ages are quoted at 2σ. See Supplementary 618 Table A for full data.

619

Fig. 3: Weighted mean of individual ⁴⁰Ar/³⁹Ar plateau steps from the Donnybrook Paleovalley.
621

622 Fig. 4: Sr–Nd–Pb isotopic data for the Bunbury Basalt, including previously published data from

623 Storey et al. (1992), Colwell et al. (1994) and Frey et al. (1996). Assigned ages are from Coffin et al.

624 (2002), Olierook et al. (2016) and this study. Where samples have not been dated (i.e., age

625 uncertainty), extrapolation of lava flows using a 3D model of the Bunbury Basalt was employed

626 (Olierook et al., 2015b). Analytical uncertainties are smaller than the symbol sizes. NHRL = Northern

627 Hemisphere Reference Line (Hart, 1984). Axes dimensions are equivalent to the entire Greater

628 Kerguelen LIP dataset in Fig. 7.

629

Fig. 5: Age progression of Greater Kerguelen LIP magmatism with longitude. (a) Southwestern
Australia, showing the evolution of basin-wide magmatism at ca. 137–136 Ma, to restriction at ca.
133 Ma and localization at ca. 130.5 Ma (Coffin et al., 2002; Olierook et al., 2016; this study). (b)
Southwestern Australia and Naturaliste Plateau, showing further younging of magmatism offshore
(Direen et al., 2017; Olierook et al., 2017).

Fig. 6: Age data comparison from pre-95 Ma volcanism attributed to the Greater Kerguelen large

637 igneous province, the timing of continental breakup, magnetic anomalies and stratigraphic data

638 constraints, modified from Whittaker et al. (2016), and recalculated and filtered for reliable data. Our

639 criteria for reliability are: (a) all data have p > 0.05, (b) 40 Ar/ 39 Ar plateau ages have >70% 39 Ar and at

640 least three consecutive steps, (c) U–Pb analyses are concordant (Spencer et al., 2016), (d) all

641 uncertainties are less than 5 Ma at 2σ (~4–5%). See Supplementary Table C for data compilation and 642 references.

643

644 Fig. 7: Compilation of Sr-Nd-Pb isotopic data from the Greater Kerguelen large igneous province, modified from Olierook et al. (2017). Mantle sources: asthenospheric depleted mantle source from 645 Southeast Indian Ridge (cf. Olierook et al., 2017), Kerguelen mantle plume from Kerguelen 646 Archipelago (Weis et al., 1993), Greater Indian continental crust from northeastern India (Ghatak and 647 648 Basu, 2011) and lithospheric source from ultrapotassic rocks in Antarctica (Sushchevskaya et al., 649 2014). For detailed rationale for mantle sources, see section 4.4 in Olierook et al. (2017). In this study, analyses were filtered for loss-on-ignition (LOI < 2%). Where LOI was not reported, symbols are 650 651 semi-transparent. For a full list of sources, see Supplementary Table D. Analytical uncertainties are 652 smaller than the symbol sizes. NHRL = Northern Hemisphere Reference Line (Hart, 1984).

653

Fig. 8: Plate and plume reconstructions and schematic cross-sections showing the breakup of eastern
Gondwana, the arrival and impingement of the Kerguelen mantle plume, and magmatic production in
eastern Gondwana at key time periods, modified from Olierook et al. (2017). (a) 147 Ma: earliest
magmatic products in eastern Gondwana in the Tethyan Himalaya (Shi et al., 2018). (b) 137 Ma:

658	rupture of the continental lithosphere and onset of the Bunbury Basalt (Gibbons et al., 2013; Olierook
659	et al., 2016). (c) 130 Ma: end of magmatism in onshore southwestern Australia (Olierook et al., 2016).
660	(d) 124 Ma: magmatism on Wallaby Plateau immediately prior to onset of magmatism on the
661	Kerguelen Plateau (Olierook et al., 2015a). (e) 100 Ma: towards end of global plate reorganization
662	(Matthews et al., 2012) and . AR = Argoland, B = Batavia Knoll, BB = Bunbury Basalt, CKP =
663	Central Kerguelen Plateau, EB = Elan Bank, EP = Exmouth Plateau, G = Gulden Draak Knoll, GB =
664	Gascoyne Block, N = Naturaliste Plateau, PCM = Prince Charles Mountains, R = Rajmahal–Bengal–
665	Sylhet Traps, SKP = Southern Kerguelen Plateau, TH = Tethyan Himalaya, W = Wallaby Plateau, Z =
666	Zenith Plateau.
667	
668	Table 1: Location of newly-collected samples for the Bunbury Basalt lava flows. Flow context from
669	Olierook et al. (2015b). Pv. = Paleovalley.
670	
671	Table 2: Summary of plateau and inverse isochrons ages for concordant ⁴⁰ Ar/ ³⁹ Ar lava flow analyses.
672	
673	Supplementary Table A: Full ⁴⁰ Ar/ ³⁹ Ar data for new analyses from the Donnybrook Paleovalley.
674	
675	Supplementary Table B: New major and trace element data, and Sr–Nd–Pb isotopic data. Previously
676	published major and trace element data for BN3, BN4, BN10, BN21 and BN22 from Olierook et al.
677	(2016).
678	

- 679 Supplementary Table C: Compilation of statistically-reliable geochronological data from the Greater
- 680 Kerguelen Plateau.
- 681
- 682 Supplementary Table D: Compilation of Sr–Nd–Pb isotopic analyses from the Greater Kerguelen
- 683 large igneous province.

Figure 1 Click here to download Figure: Figure 1_Indian Ocean + SPB map.pdf



Olierook et al., Figure 1



Olierook et al., Figure 2





Olierook et al., Figure 3





Olierook et al., Figure 4



Olierook et al., Figure 5





Olierook et al., Figure 6

Figure 7 Click here to download Figure: Figure 7_Geochemistry isotopes _v2.pdf



Olierook et al., Figure 7

Figure 8 Click here to download Figure: Figure 8_Gplates reconstruction_v2.pdf



Olierook et al., Figure 8

Sample	Coordinates ^a		Well/Area name	Flow context	Depth (m)		
no.	Latitude	Longitude			Start	End	
BN25	-33.59299	115.81622	Donnybrook 7, DDB7	Donnybrook Paleovalley	173.1	173.3	
BN26	-33.59299	115.81622	Donnybrook 7, DDB7	Donnybrook Paleovalley	191.1	191.3	
BN27	-33.59299	115.81622	Donnybrook 7, DDB7	Donnybrook Paleovalley	218.5	218.7	
BN28	-33.77329	115.81208	Donnybrook 8, DDB8	Donnybrook Paleovalley	177.5	177.7	
BN29	-33.77329	115.81208	Donnybrook 8, DDB8	Donnybrook Paleovalley	203.5	203.7	

^aAll coordinates used GCS WGS84, and were determined by GPS (± 5m)

Olierook et al., Table 1

General characteristics		Plateau characteristics				Isochron characteristics					
Sample no	K/Ca	Plateau Age (Ma ± 2σ)	Total ³⁹ Ar Released (%)	MSWD	p	lnv. Isochron Age (Ma ± 2σ)	n	⁴⁰ Ar/ ³⁶ Ar Intercept (± 2σ)	MSWD	p	Spreading factor (%)
BN 25A	0.0017	138.7 ± 2.4	74	0.52	0.91	139 ± 4	13	298 ± 5	0.55	0.87	43
BN 26	0.0017	133.7 ± 2.8	97	0.53	0.96	135 ± 5	21	296 ± 10	0.54	0.94	58
BN 26A	0.0018	135.4 ± 2.3	84	0.37	0.98	135 ± 4	15	299 ± 6	0.4	0.98	45
BN 27	0.0019	135.3 ± 5.5	100	0.03	1.00	137 ± 10	22	293 ± 35	0.03	1.00	67
BN 28	0.0018	135.5 ± 3.5	97	0.29	1.00	138 ± 6	21	289 ± 21	0.27	1.00	62
BN 29A	0.0022	No plateau age									
ighted mean of plateau steps:		135.9 ± 1.2	<i>n</i> = 92	0.63	1.00						

^aMSWD and probability of fit (p) for plateau and isochron, percentage of ³⁹Ar degassed used in the plateau calculation, number of analyses included in the isochron, and ⁴⁰Ar/³⁶Ar intercept are indicated. Analytical uncertainties on the ages are quoted at 2 sigma (2σ) confidence levels.

Olierook et al., Table 2

Supplementary Table A Click here to download Supplementary material for online publication only: Supplementary Table A_Full Ar-Ar data.xlsx Supplementary Table B Click here to download Supplementary material for online publication only: Supplementary Table B_Geochemistry.xlsx Supplementary Table C Click here to download Supplementary material for online publication only: Supplementary Table C_ Age compilation_v2.xlsx Supplementary Table D Click here to download Supplementary material for online publication only: Supplementary Table D_Geochem compilation_v2.x