

# On the viability of detrital biotite Rb-Sr geochronology

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**Abstract.** Re-examination of IODP sediment samples collected from the Bay of Bengal via laser-ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) Rb-Sr geochronology demonstrates the viability of the biotite Rb-Sr system for use  
10 as a detrital chronometer. The age population defined by the Rb-Sr dates essentially reproduces that previously published for  
detrital <sup>40</sup>Ar/<sup>39</sup>Ar dates. The effect of unknown/assumed initial <sup>87</sup>Sr/<sup>86</sup>Sr on the calculated population can be ameliorated by  
filtering for higher <sup>87</sup>Rb/<sup>86</sup>Sr ratios. Such filtering, however, could introduce bias toward more radiogenic populations,  
especially in younger material that has not had time to accumulate radiogenic product (e.g. limiting the effect of initial <sup>87</sup>Sr/<sup>86</sup>Sr  
15 to ~<5% requires filtering of <sup>87</sup>Rb/<sup>86</sup>Sr > 500 at 250 Ma and <sup>87</sup>Rb/<sup>86</sup>Sr > 50 at 2500 Ma). Finally, Ti-in-biotite temperatures  
calculated based on element concentration data collected during LA-ICP-MS overlap with those calculated for the same  
material based on electron probe microanalyzer data demonstrating the potential for *in situ* biotite petrochronology based on  
the Rb-Sr system.

## 1 Introduction

Detrital geochronology is a commonly used approach to assess a wide variety of geological questions. For example, detrital  
20 zircon can provide information about the maximum depositional age for a sedimentary unit, likely sediment sources, and  
timescales of exhumation and sediment transport (e.g. Gehrels, 2014; Thomas, 2011; Malusà and Fitzgerald, 2020). Moreover,  
detrital zircon chemistry can provide additional insight into the nature of the source rocks, such as crystallization depth and  
degree of fractional crystallization (e.g. Stevenson and Patchett, 1990; Iizuka et al., 2010; Howard et al., 2009; Mueller et al.,  
2008). Detrital geochronology (or thermochronology) of mineral phases dated via radio-decay systems that may record cooling  
25 rather than crystallization, such as <sup>40</sup>Ar/<sup>39</sup>Ar on mica, or fission track and U-Th-He dating of U bearing minerals, can be used  
to quantify rates of exhumation and/or burial in active orogenic systems (e.g. Ruiz et al., 2004; Najman et al., 1997, 2001;  
Reiners et al., 2005).

Critical to detrital geochronology, in all forms, is analysing sufficient material to characterise the statistical variation in the  
age data. Prior to the development of spot geochronology analytical techniques, analysing enough material (e.g. Vermeesch,  
30 2004) to characterise a specimen was time-consuming and expensive. The proliferation of laser ablation inductively coupled

plasma mass spectrometry-based geochronology has enabled the rapid acquisition of large datasets. While this is especially true for detrital U-Pb zircon geochronology, limitations still exist for other methods. For example,  $^{40}\text{Ar}/^{39}\text{Ar}$  LA-ICP-MS geochronology still requires irradiation of samples prior to analyses, which increases the expense and time it takes to generate data.

35 The development of *in situ* beta decay geochronology techniques (e.g. Zack and Hogmalm, 2016; Simpson et al., 2021) now allows additional detrital geochronometer options, with the critical caveat that these are typically isochron-based methods. As such, quantifying a initial isotope composition can be an integral part of calculating a date. In the absence of a measured cogenetic non-radiogenic phase to constrain that intercept, multiple radiogenic data points, which typically have varying initial isotopic reservoirs, can be regressed to define a date. Such isochrons (regressions) rely on all analyses comprising a single closed isotopic system, which is incompatible with detrital geochronology. It is possible, however, to assume an initial intercept for isochron-based data and calculate two-point regressions through that intercept and each datapoint to calculate a spot date. For both Rb-Sr and Lu-Hf, the possible/expected terrestrial initial ratios typically span a rather narrow range of values (e.g. 0.699-0.780 and 0.281-0.283; Rösel and Zack, 2022; Fisher and Vervoort, 2018). Moreover, the more radiogenic the spot analyses are, the less control the initial ratio exerts on the final two-point isochron date. This dependence is demonstrated in Larson et al. (2023a), who show that correcting Rb-Sr data for common  $^{87}\text{Sr}$  based on the current  $^{87}\text{Sr}/^{88}\text{Sr}$ , which effectively mimics the result of a two-point isochron through 0.71, effectively reproduces the isochron regression dates for samples with high Rb/Sr. The coincidence of the isochron and spot-dates derived independent of the measured initial  $^{87}\text{Sr}/^{86}\text{Sr}$  indicates that detrital Rb-Sr geochronology may be a viable alternative or addition to detrital  $^{40}\text{Ar}/^{39}\text{Ar}$  geochronology (e.g. Crossingham et al., 2024), eliminating the potential time-consuming step of irradiation and facilitating increased numbers of analyses.

50 This study presents the results of *in situ* Rb-Sr analysis of biotite mica grains picked from sand samples collected from the Bay of Bengal that have either previously been dated via  $^{40}\text{Ar}/^{39}\text{Ar}$  detrital geochronology or are directly adjacent to dated samples (see Najman et al., 2019). The potential viability of Rb-Sr as a detrital chronometer is compared against the published  $^{40}\text{Ar}/^{39}\text{Ar}$  data. Moreover, the derivation of additional information (i.e., Ti-in-biotite temperature) from the mica grain via LA-ICP-MS is also investigated to determine if it can be used to help further quantify source characteristics.

## 55 **2 Methods**

To test the viability of detrital biotite Rb-Sr geochronology, 4 samples (1450-24-25F; 1450-100-104-108F; 1451-47-49F, 1451-86F), collected from late Miocene to middle or late Pleistocene sediments during the Bengal Fan Ocean Discovery Program (IODP) Expedition 354, were examined. For simplicity, the sample names have been shortened here to 25F, 108F, 49F, and 86F, respectively. These same samples, or closely adjacent ones, have been previously investigated for detrital white mica  $^{40}\text{Ar}/^{39}\text{Ar}$ , zircon fission track, apatite U-Pb, and/or rutile U-Pb geochronology (Najman et al., 2019). Given the propensity for biotite (*sensu lato*) to be more radiogenic than white mica (e.g. Fournier et al., 2016), and the sensitivity of low radiogenic material to the initial value of an isochron, biotite were targeted solely in this study. The biotite grains were either

manually picked from sediment separates (see Najman et al., 2019) and mounted in epoxy, or the sediments were poured directly into an epoxy mount. Grains were not mounted with a preferred orientation resulting in a semi-random *c*-axis orientation (the natural shape of a mica lath favours a *c*-axis normal to the mounting surface). Mixed *c*-axis orientations may help avoid less optimal signal stability ablating parallel to the mica *c*-axis (Rösel and Zack, 2022) that may affect the final Rb-Sr date calculated (Lloyd et al., 2023). After polishing, targets were verified (i.e. the presence of K, Fe and lack of visible inclusions) via microXRF elemental mapping of each mount prior to analysis.

## 2.1 Rb-Sr geochronology

Rb-Sr geochronology was carried out via laser ablation inductively coupled plasma tandem mass spectrometry following the methods initially set out in Zack and Hogmalm (2016) and Hogmalm et al. (2017). Analyses were carried out in the Fipke Laboratory for Trace Element Research (FiLTER) at the University of British Columbia, Okanagan (UBCO) using an ESL NWR 193 laser with a TwoVol3 ablation cell paired with an Agilent 8900 triple quadruple (QQQ) ICP-MS as described in Larson et al. (2023a, b). A circular laser spot, with a diameter of 65  $\mu\text{m}$ , approximate pit depth of  $\sim 10\text{-}15\ \mu\text{m}$ , a fluence of  $\sim 3\ \text{J}/\text{cm}^2$  and repetition rate of 5 Hz was used for all materials. Analyses of the glass reference material NIST SRM610 (Jochum et al., 2011) were carried out using both 60 and 30  $\mu\text{m}$  diameter laser spots to ensure analyses measured in both pulse and analogue detector modes for cross calibration (e.g. Zack and Hogmalm, 2016; Glorie et al., 2024). Instrument drift was corrected, and down-hole fractionation was minimized (e.g. Glorie et al., 2023), based on analyses of NIST SRM610 using an in-house data reduction scheme (DRS; Larson, 2024) developed for Iolite v.4.X (Paton et al., 2011) based on the freely available ‘U-Pb Python Example.py’ DRS (Petrus, 2022), following the approach and error propagation detailed in Redaa et al. (2021), and using a covariance matrix to calculate error correlation. Matrix fractionation effects on  $^{87}\text{Rb}/^{86}\text{Sr}$  were corrected based on repeated analyses of Mica 1O (phlogopite -  $986 \pm 5\ \text{Ma}$ ; Camacho et al., 2020) and verified based on the analyses of GA-1550 (Mt. Dromedary biotite -  $98.7 \pm 1.9\ \text{Ma}$ , Li et al., 2008). During run 1 (108F and 47-49F), GA-1550 returned an isochron date of  $99.6 \pm 3.5\ \text{Ma}$  (mean squares weighted deviates (MSWD) = 1.9,  $n = 14/15$ , initial  $^{87}\text{Sr}/^{86}\text{Sr} = 0.7049 \pm 0.0017$ ). During run 2 (25F and 86F), GA-1550 returned an isochron date of  $98.1 \pm 2.3\ \text{Ma}$  (MSWD = 0.93,  $n = 17/17$ , initial  $^{87}\text{Sr}/^{86}\text{Sr} = 0.7049 \pm 0.0017$ ) while an additional in-house reference material, Mica 1B (phlogopite -  $990 \pm 6\ \text{Ma}$ ; Camacho et al., 2012), returned an isochron date of  $997 \pm 6\ \text{Ma}$  (MSWD = 3.7,  $n = 17/20$ , initial  $^{87}\text{Sr}/^{86}\text{Sr} = 0.7035 \pm 0.0005$ ). Full Rb-Sr data are provided in Tables S1 and S2.

## 2.2 Ti-in-biotite thermometry

In addition to Rb-Sr isotopes, Ti, Mg, and Fe concentrations were measured for each analytical spot via LA-ICP-MS with dwell times of 1 ms, 0.5 ms, and 1 ms, respectively. Concentrations were normalized to repeated measurements of the NIST SRM610 glass reference material (Jochum et al., 2011) assuming stoichiometric Si (16.36 weight (wt.) % Si or 35 wt.%  $\text{SiO}_2$ ), typical of metamorphic biotite from greenschist through granulite facies (e.g. Dyck et al., 2021). Titanium concentrations

(ppm) were converted to molar weight per oxide TiO<sub>2</sub> equivalents and then normalized to calculated Si content based on an assumed 5.4 atoms per formula unit (a.p.f.u.) Si per 22 O as detailed in Eq. (1):

$$(1) \text{ Ti a.p.f.u.} = \left( \frac{c\text{Ti}}{w\text{T}TiO_2} / \frac{10000}{M\text{TiO}_2} \right) * (5.4 / A\text{Si})$$

Where,  $c\text{Ti}$  = concentration of Ti in ppm,  $w\text{T}TiO_2$  = weight proportion of Ti in TiO<sub>2</sub>,  $M\text{TiO}_2$  = molar weight per oxide, and  $A\text{Si}$  is the molar weight per cation Si assuming 35 wt % SiO<sub>2</sub>. Ti-in-biotite temperatures were calculated using the equation of Henry et al. (2005), which requires Ti a.p.f.u and Mg# (=Mg/(Mg+Fe)) for each analysis. The values for Mg# were calculated using ppm concentrations of Fe and Mg. The 1 $\sigma$  uncertainty of the Ti-in-biotite temperatures is <24 °C at 480 °C decreasing to 12 °C at 800 °C (Henry et al., 2005).

To verify the LA-ICP-MS derived Ti-in-biotite temperatures, the chemistries of the same mica grains were analysed using the CAMECA SXFive FE electron probe microanalyser (EPMA) also housed in the FiLTER facility. Quantitative spot analyses were carried out with an acceleration voltage of 15 kV, regulated beam current of 20nA and a spot size of 5  $\mu\text{m}$ . Elemental x-ray data were collected using a dwell time of 30 s on peak and 15 s on background and were calibrated to the known composition of synthetic and natural mineral reference standards from Micro-Analysis Consultants Ltd. The standards used for calibration were spessartine for Si and Mn, titanite for Ti, almandine for Al and Fe, wollastonite for Ca, albite for Na, diopside for Mg, orthoclase for K, and apatite for P. The crystals used were LTAP for Si and Al, LPET for Ti, Ca, K, P LLIIF for Fe, Mn, and TAP for Na, Mg. The EPMA data were converted to atoms per formula unit biotite on the basis of 22 oxygen whereby:

$$(2) \text{ cation (a.p.f.u.)} = \text{cation proportion} * \# \text{ oxygen (a.p.f.u.)} / \text{sum of oxygen proportions,}$$

$$(3) \text{ oxygen proportion} = \text{cation proportion} * (1/2) \text{ cation charge,}$$

$$(4) \text{ cation proportion} = \text{oxide wt. \%} / \text{oxide molecular wt.}$$

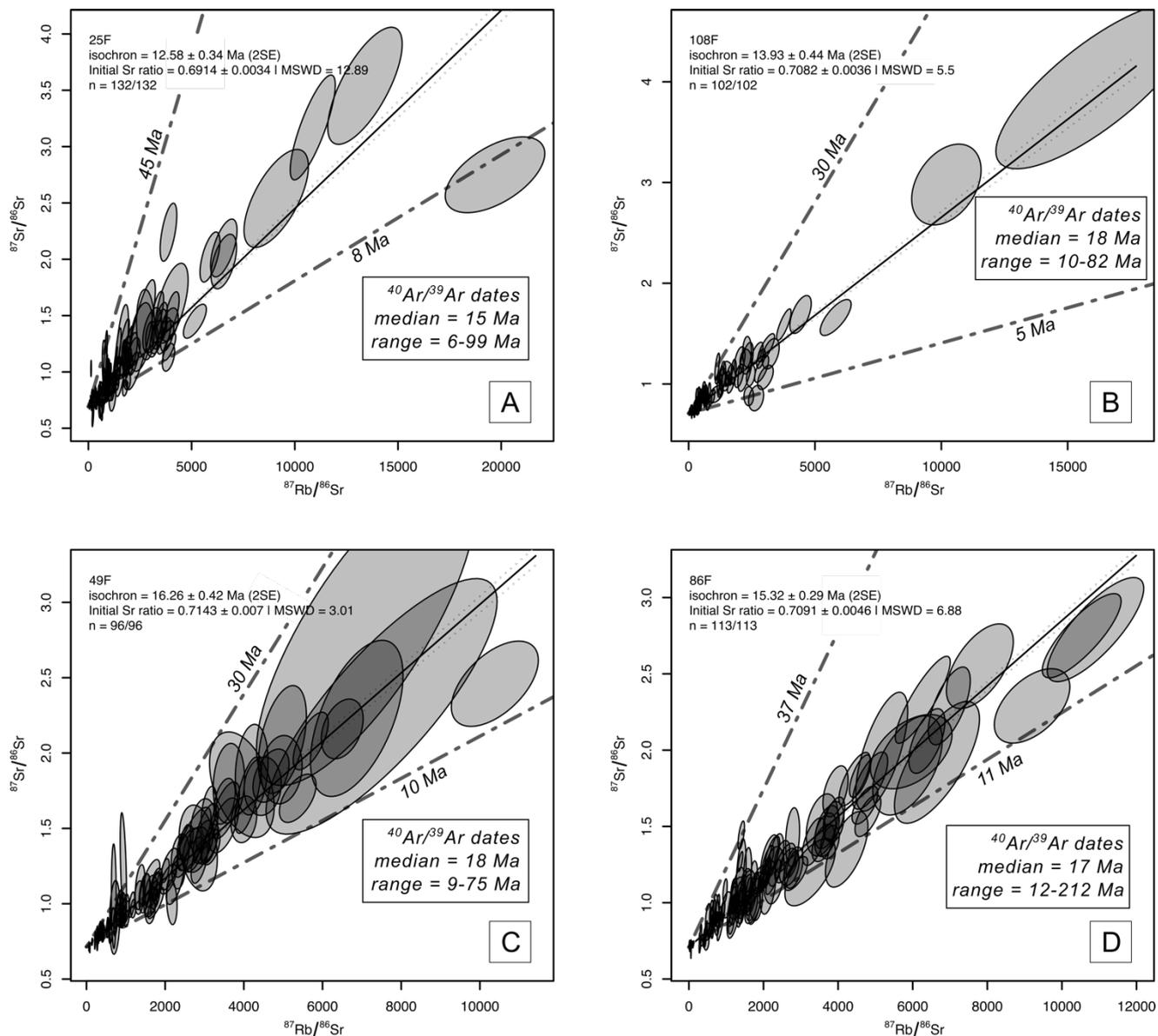
Given that the biotite analysed was collected as detritus from the Bengal Fan, and the tendency of biotite to weather (e.g. Wilson, 2004), alteration was expected. To avoid the most altered material, EPMA data were filtered for totals > 88 wt % and K concentrations > 1.1 a.p.f.u. Plotting K a.p.f.u against Mg# or calculated temperature shows that analyses with K a.p.f.u. > 1.1 rarely form outliers (Fig. 3). While it is recognized that an a.p.f.u of 1.1 K is significantly lower than that expected of unaltered biotite, K is expected to leave the mineral via hydrated cation exchange as part of the initial weathering stage/vermiculitization (Price and Velbel, 2014; Gilkes and Suddhiprakarn, 1979). In contrast, Fe, Mg and Ti are relatively immobile until more advanced alteration of biotite to goethite or kaolinite (Gilkes and Suddhiprakarn, 1979). Exchange of hydrated cations with interlayer K will result in lower oxide EPMA wt. % totals, which upon normalization (for a biotite with a  $X\text{Mg} = 0.44$ ), increases the temperature calculated using the Ti-in-biotite thermometer by ~2.2 °C per 1 wt. % oxide deficit. The sensitivity of Ti-in-biotite temperatures to EPMA oxide wt.% totals, using the Henry et al. (2005) calibration, is shown in Fig. S1. To illustrate the effect of weathering on our data we plot all analyses, coloured by oxide wt. % totals, in Fig. 3A, B. In general, filtering analyses with K concentrations > 1.1 a.p.f.u rather than filtering solely on oxide wt. % totals is the more robust approach to removing outliers with anomalously low Mg# (e.g., < 0.15) and Ti-in-biotite temperatures (e.g., < 400 °C). Full EPMA data are provided in Table S3.

### 3 Results

#### 3.1 Rb-Sr geochronology

130 All Rb-Sr results were filtered for Rb/Sr ( $>3$ ), Fe + Mg (between 15 and 26 wt. %), and Ti ( $>0.1$  a.p.f.u) to avoid spurious analyses. One hundred and thirty-two of 137 spot analyses of biotite from specimen 25F remained after filtering. The data spread between isochrons at 45 and 8 Ma and define an over-dispersed (MSWD = 12.9) ca. 13 Ma isochron (Fig. 1A). Similarly, One-hundred and thirty-eight spots in biotite from 108F yield 102 viable analyses that spread between 5 and 30 Ma isochrons. The data define an over-dispersed regression (MSWD = 5.5) at ca. 14 Ma (Fig. 1B). Ninety-six of 104 analyses in  
135 biotite separated from 49F spread between isochrons at 10 and 30 Ma, defining an over-dispersed (MSWD = 3.0) isochron at ca. 16 Ma (Fig. 1C). Finally, 113 of 117 biotite analyses of material from 86F spread between isochrons at 37 and 11 Ma and comprise an over-dispersed (MSWD = 6.88) regression at ca. 15 Ma (Fig. 1D).

For each specimen, spot dates were calculated as two-point dates based on a regression between each analysis and a specified initial  $^{87}\text{Sr}/^{86}\text{Sr}$  value (see Tables S4-S7). The initial  $^{87}\text{Sr}/^{86}\text{Sr}$  values used (0.71, 0.72, 0.73, 0.74, 0.75 and 0.760 cover the  
140 range of expected values for most plutonic and metamorphic sources (Rösel and Zack, 2022). The dates calculated for each specimen are and depicted in Fig. 2 as kernel density estimations (KDE; bandwidth = 3). In general, the KDEs have a single peak for  $> 0$  Ma dates (if a spot analysis has an  $^{87}\text{Sr}/^{86}\text{Sr} <$  than the initial  $^{87}\text{Sr}/^{86}\text{Sr}$ , a negative date will be calculated, a situation exacerbated in young material that has not had time to accumulate radiogenic  $^{87}\text{Sr}$  - see Tables S1-4). The KDE peaks for each specimen generally increase in width, decrease in height, and move to slightly younger positions with two-point dates  
145 calculated with progressively higher initial  $^{87}\text{Sr}/^{86}\text{Sr}$  (Fig. 2).



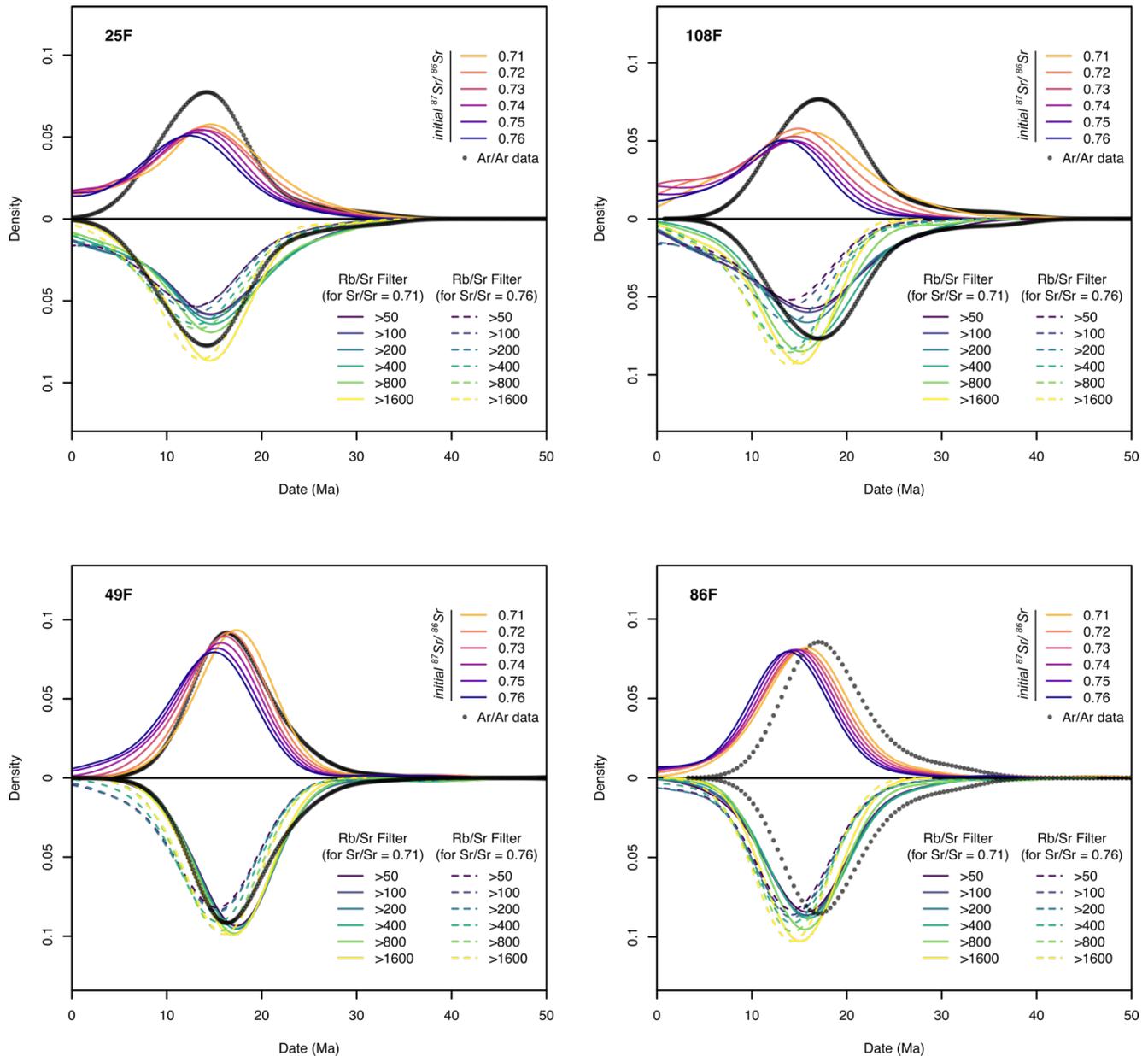
**Figure 1:**  $^{87}\text{Rb}/^{86}\text{Sr}$  versus  $^{87}\text{Sr}/^{86}\text{Sr}$  isochron plots of data collected from each sample investigated (A-D). Dashed lines denote the approximate envelope of each dataset with corresponding dates as marked.  $^{40}\text{Ar}/^{39}\text{Ar}$  dates quoted are from Najman et al. (2019) and correspond to data from samples 1450-38-40F, 1450-98F, 1451-31-33-37F, and 1451-86F for 25F, 108F, 49F and 86F, respectively.

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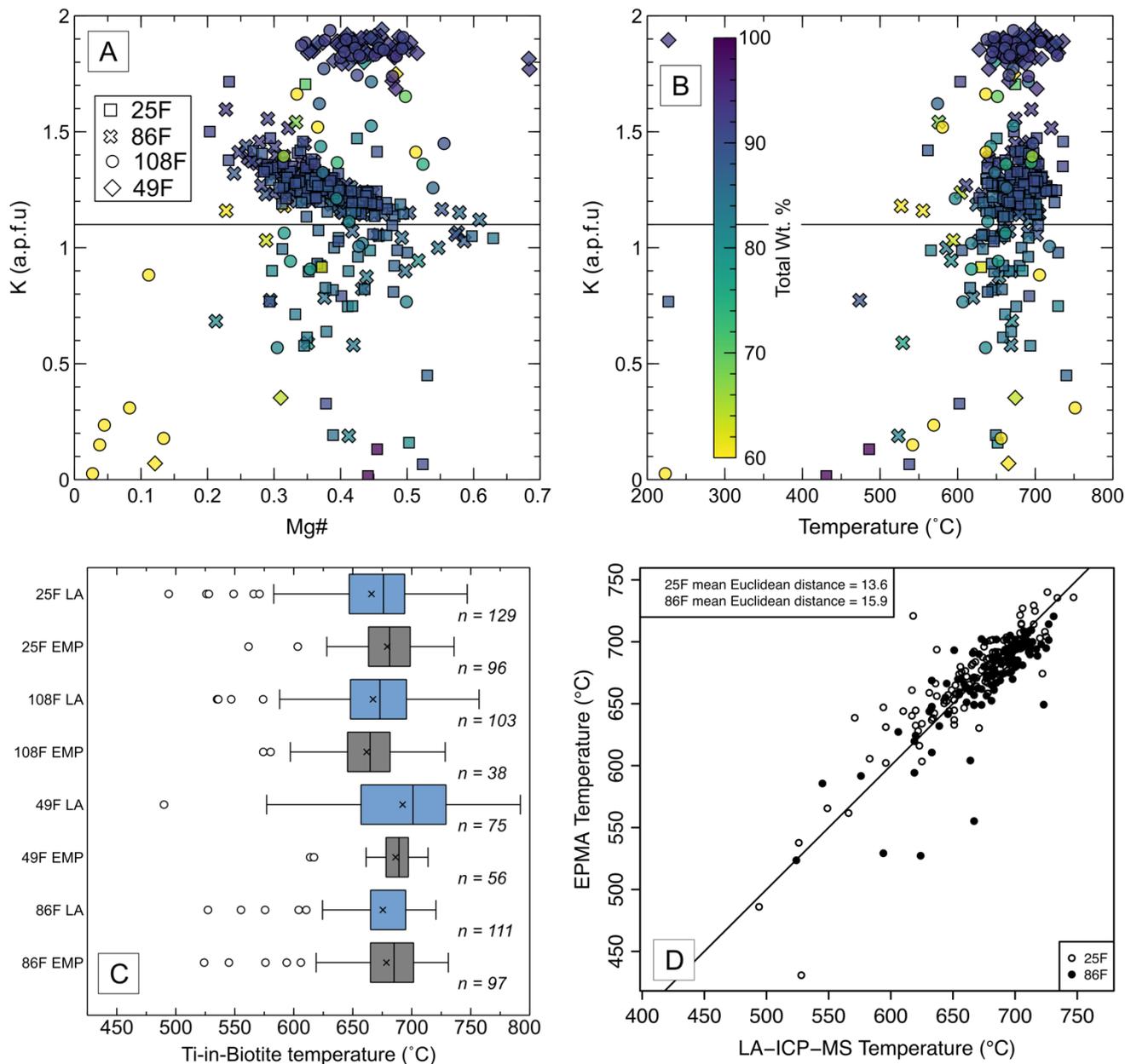
### 3.2 Ti-in-biotite thermometry

Ti-in-biotite temperatures calculated via LA-ICP-MS data generally range between  $\sim 650$  and  $725$  °C for most samples (Figs. 3B, C). The  $\text{SiO}_2$  content used in the calculations has little effect on temperature. Increasing the  $\text{SiO}_2$  from 35 to 40 wt. % results in only a  $\sim 5$  °C temperature change (Fig. S2), well within the expected uncertainty of the method (Henry et al., 2005).

155 While the precision of Ti data generated by LA-ICP-MS is lower than EPMA data, the second and third quartile temperatures overlap (within uncertainty) with those calculated using EPMA data for each specimen (Fig. 3C).



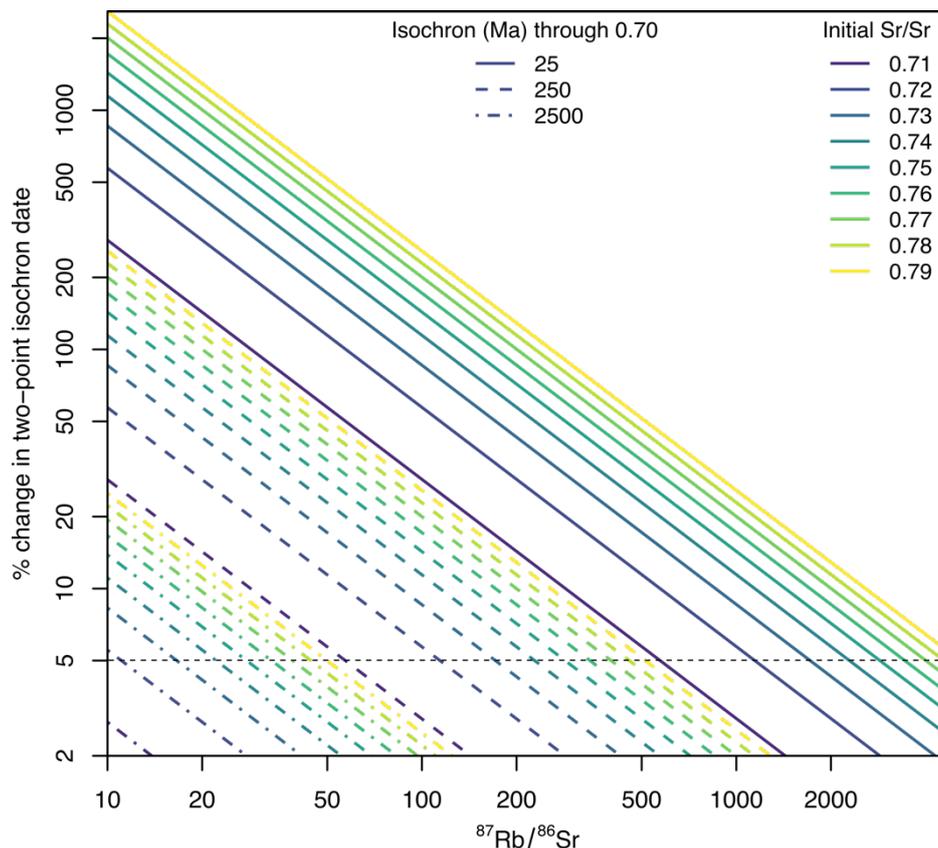
160 **Figure 2: Kernel density estimation (KDE) plots of Rb-Sr data from samples analysed and  $^{40}\text{Ar}/^{39}\text{Ar}$  of the same, or spatially adjacent, samples. The kernel bandwidth was 3 Ma for all plots. Each diagram shows two different datasets. The data plotted above the median line includes  $^{40}\text{Ar}/^{39}\text{Ar}$  white mica dates and unfiltered Rb-Sr two-point isochron dates based on initial  $^{87}\text{Sr}/^{86}\text{Sr}$  as marked. Dates were calculated using IsoplotR (Vermeesch, 2018). The data plotted below the median line represent the results of applying different  $^{87}\text{Rb}/^{86}\text{Sr}$  filters, as noted.**



165 **Figure 3:** A) Plot of K atoms per formula unit (a.p.f.u; based on 22O) as calculated from electron probe microanalyser (EPMA) data versus Mg# (Mg# = Mg/(Fe + Mg)). A horizontal line is drawn at K a.p.f.u = 1.1. B) Plot of K a.p.f.u. versus derived Ti-in-biotite temperature. A horizontal line is drawn at K a.p.f.u = 1.1. C) Box and whisker plots of calculated Ti-in-biotite temperatures calculated for each sample via EPMA (grey fill) and laser ablation inductively coupled plasma mass spectrometer (LA-ICP-MS) data (blue fill). D) Plot of EPMA versus LA-ICP-MS temperatures for adjacent spot analyses on the same grains in samples 25F and 86F. Calculated mean Euclidean distances between the EPMA versus LA-ICP-MS data points for each sample are shown.

**4.1 Effect of initial  $^{87}\text{Sr}/^{86}\text{Sr}$** 

As with any isochron method, the value of the intercept can have a significant impact on the results when calculating two-point isochron Rb-Sr dates. That effect, however, is less significant with older material (higher radiogenic Sr) and more radiogenic analyses. As shown in Fig. 4, the effect of initial intercept selection can be demonstrated by generating three artificial datasets with 1 analysis every 500  $^{87}\text{Rb}/^{86}\text{Sr}$  increment between 10 and 4510. One dataset defines a 25 Ma isochron, one defines a 250 Ma isochron, and one defines a 2500 Ma isochron, each with an initial  $^{87}\text{Sr}/^{86}\text{Sr}$  of 0.70. Two-point spot Rb-Sr dates were calculated for each datapoint within each model for different initial  $^{87}\text{Sr}/^{86}\text{Sr}$  ranging from 0.71 to 0.79, and the % absolute deviation in two-point isochron date for each modelled point from the 'known' date (i.e. 25, 250 or 2500 Ma) was quantified (Fig. 4). Plotting the % change in two-point isochron date versus  $^{87}\text{Rb}/^{86}\text{Sr}$  demonstrates that low  $^{87}\text{Rb}/^{86}\text{Sr}$  and young material is most affected by changing the initial  $^{87}\text{Sr}/^{86}\text{Sr}$ . Moreover, the plot further demonstrates that for high  $^{87}\text{Rb}/^{86}\text{Sr}$  ( $\sim > 2500$ ), even in young material, the typical difference in initial  $^{87}\text{Sr}/^{86}\text{Sr}$  encountered in most crustal rocks (i.e. 0.70 - 0.76; Rösler and Zack, 2022), will have  $\sim < 5\%$  effect on the date calculated (Fig. 4). For older material, the  $^{87}\text{Rb}/^{86}\text{Sr}$  cut-offs to minimize the effect of initial  $^{87}\text{Sr}/^{86}\text{Sr}$  on dates (i.e.  $< 5\%$ ) drops significantly (i.e.  $\sim > ^{87}\text{Rb}/^{86}\text{Sr} = 500$  at 250 Ma and  $\sim > ^{87}\text{Rb}/^{86}\text{Sr} = 50$  at 2500 Ma; Fig. 4).



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**Figure 4: Plot of modelled data demonstrating the related effects of initial  $^{87}\text{Sr}/^{86}\text{Sr}$  on calculated two-point isochron dates as a function of  $^{87}\text{Rb}/^{86}\text{Sr}$ . See text for discussion.**

The effect of initial  $^{87}\text{Sr}/^{86}\text{Sr}$  relative to  $^{87}\text{Rb}/^{86}\text{Sr}$  can be further investigated using the real-world data presented herein. Filtering the two-point isochron dates calculated based on  $^{87}\text{Rb}/^{86}\text{Sr}$  demonstrates that the position of the main population of the data remains relatively invariant for different  $^{87}\text{Rb}/^{86}\text{Sr}$  but the half-width decreases and the density of the main population increases with higher  $^{87}\text{Rb}/^{86}\text{Sr}$  cut-offs (bottom curves Fig. 2). These results indicate that filtering detrital data by  $^{87}\text{Rb}/^{86}\text{Sr}$  may help ameliorate the complication of unknown initial  $^{87}\text{Sr}/^{86}\text{Sr}$  values, though it should be noted that doing would introduce bias toward more radiogenic populations and, as such, should not be done in isolation.

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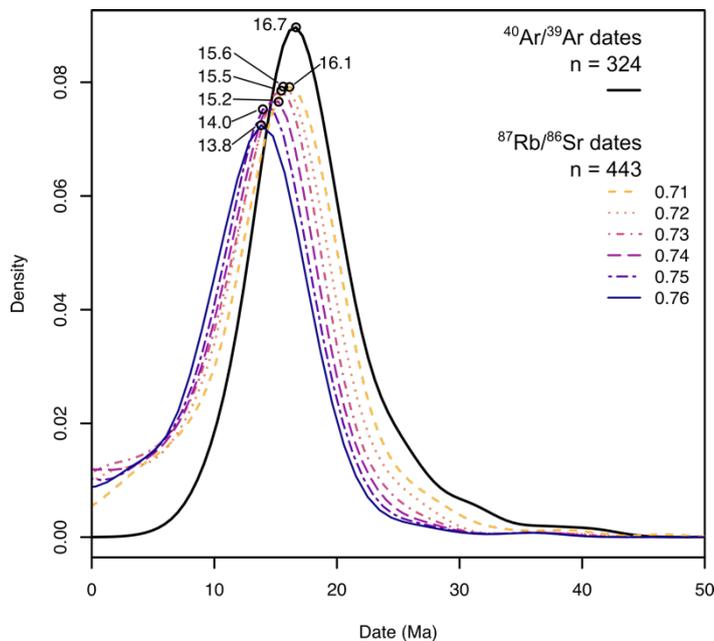
#### 4.2 Comparison with detrital $^{40}\text{Ar}/^{39}\text{Ar}$ geochronology

Najman et al. (2019) presented the results of various detrital geochronology methods employed on the samples examined in the current study and/or other proximal samples. These methods include  $^{40}\text{Ar}/^{39}\text{Ar}$  geochronology on white mica. White mica was targeted in that study both because of the common problem of excess Ar associated with biotite (Stübner et al., 2017; Larson et al., 2023a) in the Himalayan system from which these samples were collected (i.e. Himalayan detritus shed into the Bay of Bengal) and the resilience of white mica, relative to biotite, to weathering (Wilson, 2004). Given the differences in

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200 child product retention between white mica  $^{40}\text{Ar}/^{39}\text{Ar}$  geochronology and biotite Rb-Sr geochronology it may not be expected that the data from the two systems would overlap. White mica has an estimated nominal closure temperature of  $\sim 425\text{-}400\text{ }^{\circ}\text{C}$  for Ar diffusion (100 micron radius grain,  $10^{\circ}\text{C}/\text{Ma}$  cooling rate, 5-10 kbar; Harrison et al., 2009), whereas closure to diffusion of Sr out of biotite is more varied with estimates ranging from  $\sim 300\text{ }^{\circ}\text{C}$  (Jager et al., 1967; Armstrong et al., 1966) to  $\sim 400\text{ }^{\circ}\text{C}$  (Verschure et al., 1980; Del Moro et al., 1982) or even in excess of  $400\text{ }^{\circ}\text{C}$  depending on the specifics of the mineralogy and chemistries of the samples (Jenkin et al., 1995, 2001). This variability in chronometer ‘closure temperatures’ (e.g. Dodson, 205 1973) may be reflected in variable offsets between the Rb-Sr and  $^{40}\text{Ar}/^{39}\text{Ar}$  dates (Fig. 2).

The various two-point isochron Rb-Sr dates calculated for 108F and 86F define density peaks with a slightly younger ( $\sim 1\text{ Ma}$ ) offset than the  $^{40}\text{Ar}/^{39}\text{Ar}$  dates (initial  $^{87}\text{Sr}/^{86}\text{Sr} = 0.71$ ), whereas for 49F and 25F two-point isochron dates essentially reproduce the density peak of the  $^{40}\text{Ar}/^{39}\text{Ar}$  dates (Fig. 2). Overall, the total Rb-Sr dataset, regardless of initial  $^{87}\text{Sr}/^{86}\text{Sr}$  used, defines a similar dominant, Early-Middle Miocene age population (Fig. 5) to that noted by Najman et al. (2019). That peak was interpreted to demonstrate rapid exhumation of the Eastern and Central Himalayan with a  $\sim 4\text{ m.y.}$  lag time between exhumation through mica closure to child product diffusion and sedimentation (Najman et al., 2019). Given the similarities between the datasets, the detrital Rb-Sr geochronology would have led to the same conclusions as the  $^{40}\text{Ar}/^{39}\text{Ar}$  data made by Najman et al. (2019).



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**Figure 5: Kernel density estimations (bandwidth = 2) of all  $^{40}\text{Ar}/^{39}\text{Ar}$  data reported by Najman et al. (2019) and Rb-Sr spot dates calculated in the present study assuming various initial  $^{87}\text{Sr}/^{86}\text{Sr}$  (as denoted). The date (Ma) corresponding to each KDE peak is shown.**

### 4.3 Ti-in-biotite thermometry

220 Comparison of EPMA and LA-ICP-MS thermometry results demonstrate that LA-ICP-MS data can yield results comparable  
with traditional methods (Fig. 3). The temperatures calculated for the spot data are consistent with derivation from amphibolite-  
facies metamorphic rocks or associated leucogranites (e.g. Waters, 2019), which dominate the inferred exhuming Himalayan  
midcrustal source (Najman et al., 2019). The temperatures also broadly overlap with Zr-in-rutile temperature derived from  
225 detrital rutile from the same samples (~650-780 °C; Najman et al., 2019). Direct comparison of LA-ICP-MS and EPMA-  
derived temperatures for the same grains from 25F and 86F define mean Euclidean distances of 13.6 and 15.9 °C, respectively  
(Fig. 3D). Because this approximately 15 °C uncertainty likely reflects the analytical precision of LA-ICP-MS rather than  
solely due to the uncertainties stemming from natural variations in biotite chemistry or the thermometer calibration, it should  
be considered additive to the  $\pm 12\text{--}24$  °C  $1\sigma$  uncertainty of Henry et al., (2005). The precision on the Ti-in-biotite temperature  
estimates, whether EPMA or LA-ICP-MS derived, does not facilitate quantifying a change across the sampled strata as data  
230 from all samples overlap (Fig. 3C).

The viability of quantifying Ti-in-biotite temperatures via LA-ICP-MS allows each Rb-Sr spot analysed to be associated with  
a unique temperature. This kind of petrochronologic association opens the possibility of many different types of studies, not  
unlike the linking of chemistry to spot dates did for U(-Th)-Pb geochronology in the early 2000s (e.g. Foster et al., 2004;  
Gibson et al., 2004; Rubatto, 2002).

### 235 5 Conclusions

LA-ICP-MS Rb-Sr geochronology appears to be a viable method to efficiently generate detrital biotite geochronological  
datasets. The effect of unknown initial  $^{87}\text{Sr}/^{86}\text{Sr}$  values can be mitigated for young (Cenozoic) material by filtering for  
significantly radiogenic analyses ( $^{87}\text{Rb}/^{86}\text{Sr} > 2500$ ), though such filtering may introduce bias. Filtering can be less aggressive  
for older material (i.e.  $> 500$  at 250 Ma) in which significant radiogenic child product has accumulated. Finally, the benefits  
240 of biotite Rb-Sr geochronology can be further extended by calculating a Ti-in-biotite temperature for each spot. Such  
information may allow for identification of multiple sources in detrital samples with complex provenance.

#### Data availability

All data related to this study are available on the Open Science Framework website:  
[https://osf.io/vgjh6/?view\\_only=52b27e0d000c4f2ea6b9ab665744e43c](https://osf.io/vgjh6/?view_only=52b27e0d000c4f2ea6b9ab665744e43c)

## 245 **Author contributions**

KPL – conceptualization; funding acquisition, methodology, investigation, visualization, writing – original draft preparation; BD – methodology, investigation, writing – review and editing; SS – investigation, writing – review and editing; MB – investigation, writing – review and editing; YN – resources, writing – review and editing.

## **Competing interests**

250 None of the authors have any competing interests.

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