

Appendix

Methods

EPMA: A JEOL JXA 8200 superprobe in WDS mode, with 15 kV acceleration voltage and a 5 mm beam with 20 nA beam current, housed at the Advanced Analytical Centre, James Cook University (JCU) was used for quantitative major element analysis of all the major silicates phases and apatite. A defocused beam was used to avoid potential diffusion of Na and Cl by the beam. Counting times were 20 seconds on peak and 10 seconds on background for each element. The data were processed using $\rho\rho z$ corrections (Armstrong, 1991) and standardised against a set of well-characterized, in-house oxide (hematite) and silicate (albite, almandine, orthoclase, wollastonite, olivine), sulfate (barite), tugtupite (chlorine), and Sr-fluorite (fluorine) standards. Accurate analysis of apatite can be challenging due to Cl and F migration during analysis (McCubbin et al. 2010). We applied an offline calculation that corrects signal intensities for potential F and Cl migration in a similar way to that described in McCubbin et al. (2010). Allanite analyses were performed using single element phosphates for REE standardisation and standardisation was cross-checked against in-house allanite standards.

XRF: Major and selected trace elements analyses of bulk rock samples were done at the Advanced Analytical Centre, JCU by conventional X-ray fluorescence (XRF) using a Bruker-AXS S4 Pioneer XRF Spectrometer on fused beads and pressed powder pellets, respectively.

LA-ICP-MS: Trace element analysis on bulk rock samples were carried out at the Advanced Analytical Centre at JCU, following a similar methodology to that of Holm et al. (2013). Rock powders were mixed with 12:22 borate flux at a sample to flux ratio of 1:6 and fused to glass after heating to 1050 °C. The fused samples were then mounted into epoxy

pucks and analyzed for a range of trace and major elements by LA-ICP-MS using a Varian 820 quadrupole ICP-MS coupled with a GeoLas Pro 193 nm ArF Excimer laser system. Ablation was conducted in He atmosphere and the ICP-MS was tuned to ensure robust plasma conditions (sensitivity of Th~U~Pb) (e.g., Pettke, 2008) and low oxide production levels (<0.5 % ThO), using a laser energy density of 6 J/cm² and repetition rates of 10 Hz. The beam diameter was set at 120 micrometers and 3 analyses per sample were conducted. All elements were externally standardized using NIST SRM 612 and NIST SRM 610 glass reference materials with concentration values taken from Spandler et al. (2011), using SiO₂, obtained by XRF, as the internal standard. The Glitter software package (Van Achterbergh et al., 2001) was used for data processing.

The same laser ablation ICP-MS system was used for trace element analysis in minerals. The beam diameter varied between 16 and 90 micrometers and analyses were standardized using bracketing analysis of NIST SRM 612 and NIST SRM 610 glasses. The analyses were internally standardized using ⁴³Ca for titanite and apatite, ³¹P for monazite and xenotime, and ²⁹Si for other silicate minerals. Final concentration values were performed using the SILLS software (Guillong et al., 2008). Glass reference (NIST SRM 612 and 610) values for external standardization were taken from Spandler et al. (2011).

Quantitative Zn analysis by LA-ICP-MS can be difficult due to potential ²⁶Mg and ⁴⁰Ar interference (²⁶Mg⁴⁰Ar+) with the ⁶⁶Zn signal. We assessed the degree of ²⁶Mg⁴⁰Ar+ interference with ⁶⁶Zn by monitoring the apparent Zn signal while ablating a pure MgO waver as well as San Carlos Olivine. Our results show that ~ 0.2 ppm Zn is produced per 1 wt.% Mg. All Zn analyses have been corrected for this interference accordingly.

Element distribution budgets for each sample were calculated by multiplying the average element content of each mineral by the respective mineral modal proportion, and then normalizing to bulk rock element content. The mineral modal proportions were calculated via least-squares regressions calculated in Excel, using mineral and bulk-rock compositions. The

calculated mineral proportions agree with petrographic observations in thin sections. The least-squares calculations show low values ($\sum r^2 < 1$) for all samples that reflect precise mineral phase proportion calculations.

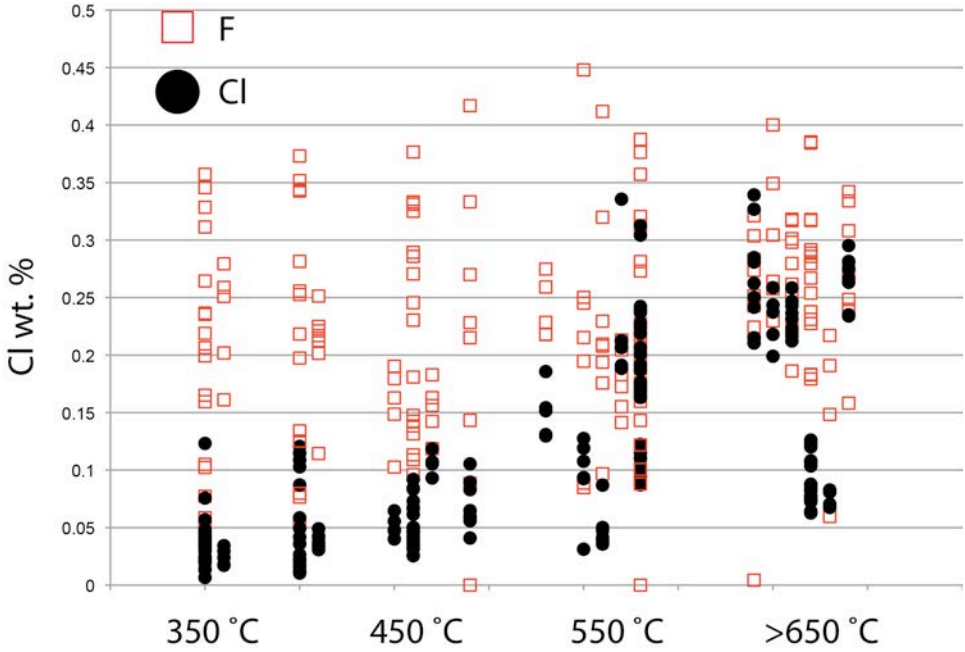


Figure A-1: Cl and F concentration in biotite.

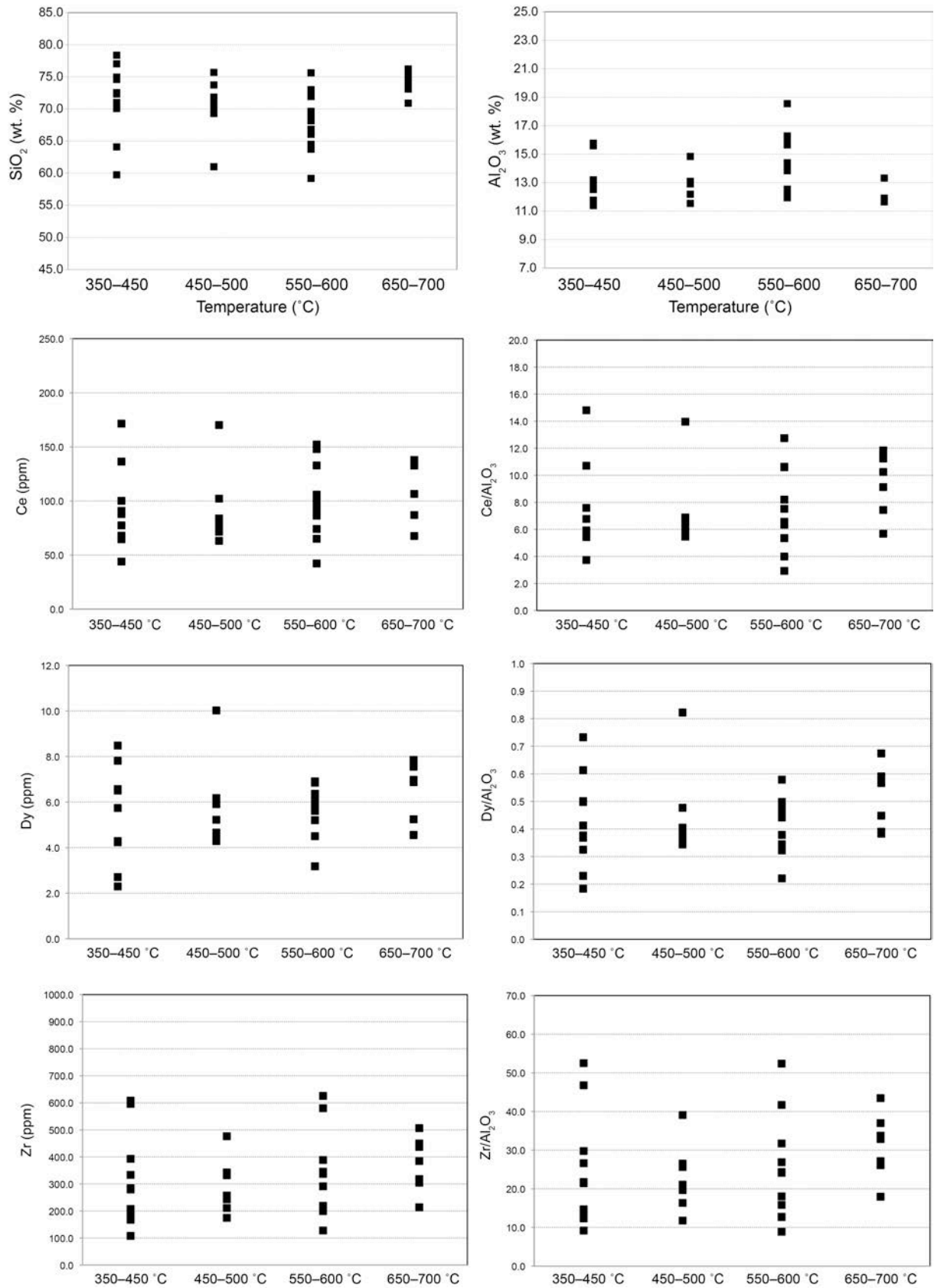


Figure A-2: Major- and trace element bulk rock concentrations.

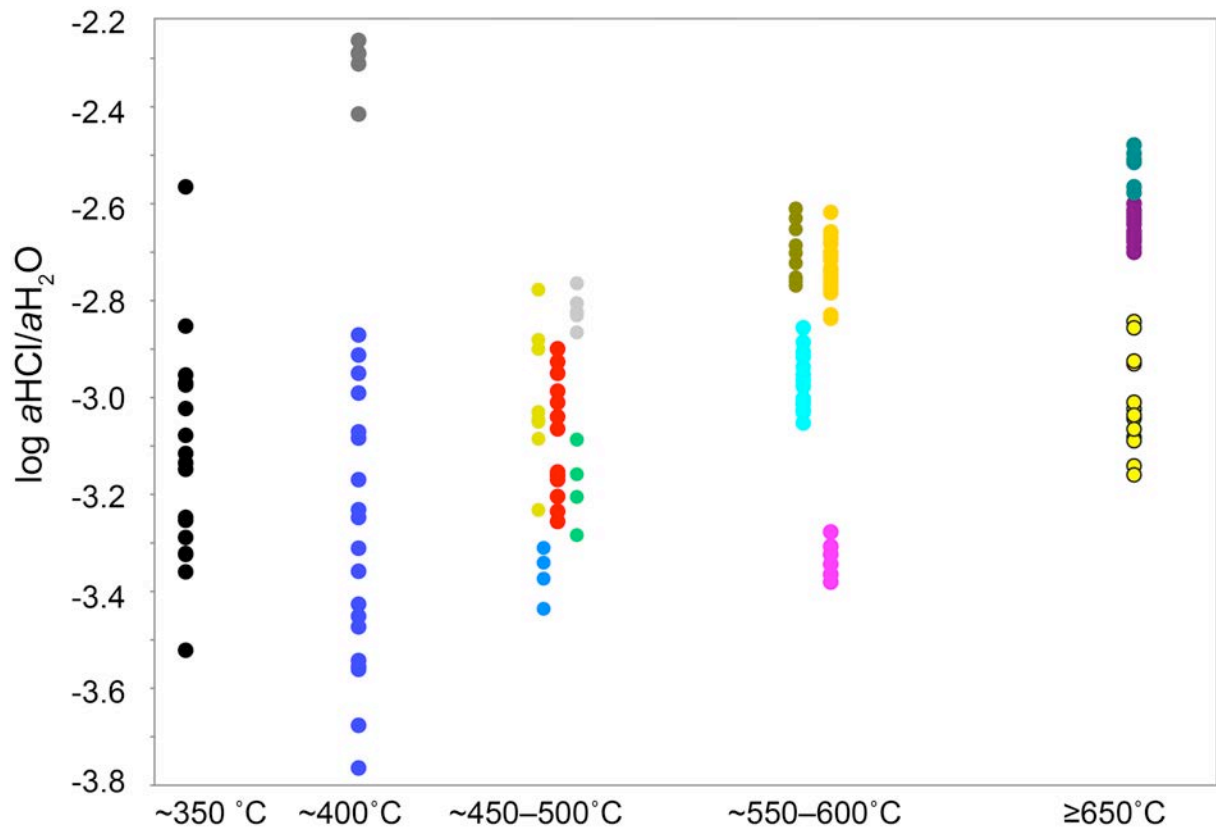


Figure A-3 $\log(a_{\text{HCl}}/a_{\text{H}_2\text{O}})$ of fluid calculated based biotite composition following the procedure by Coulson et al. (2001).