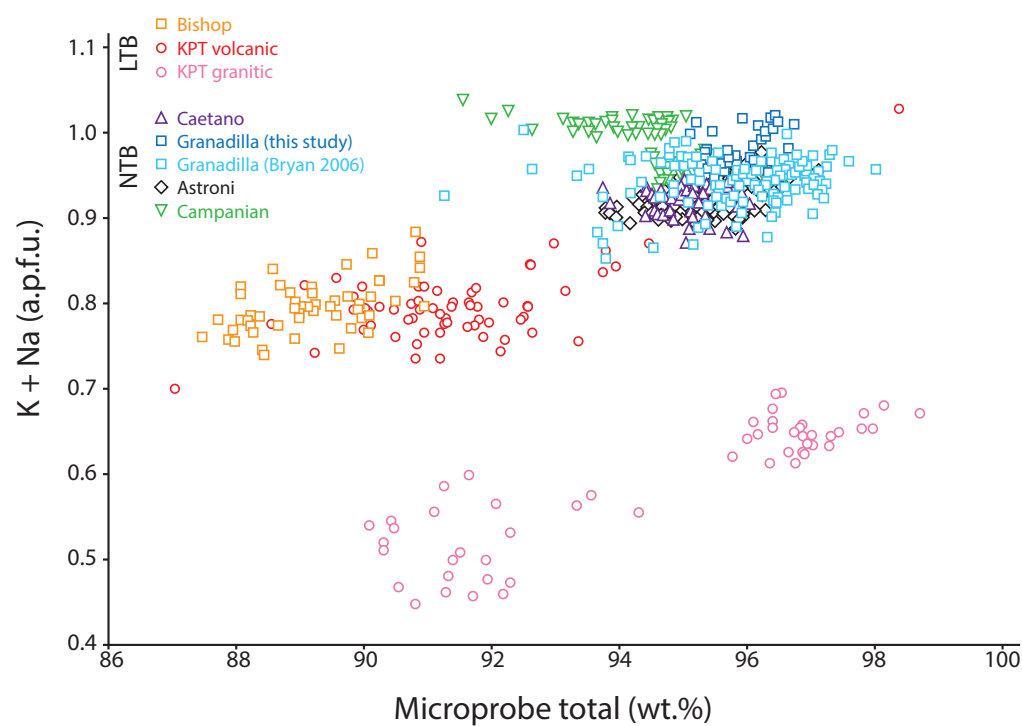
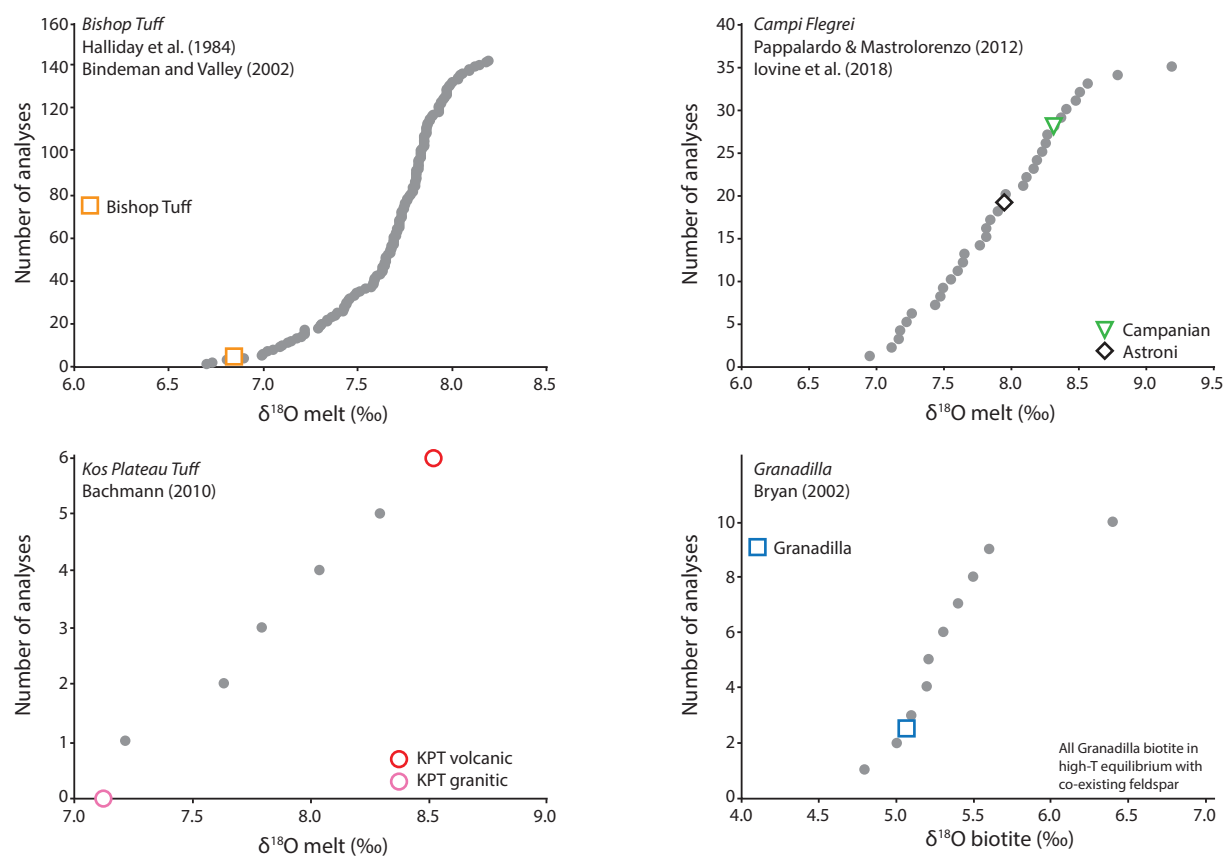


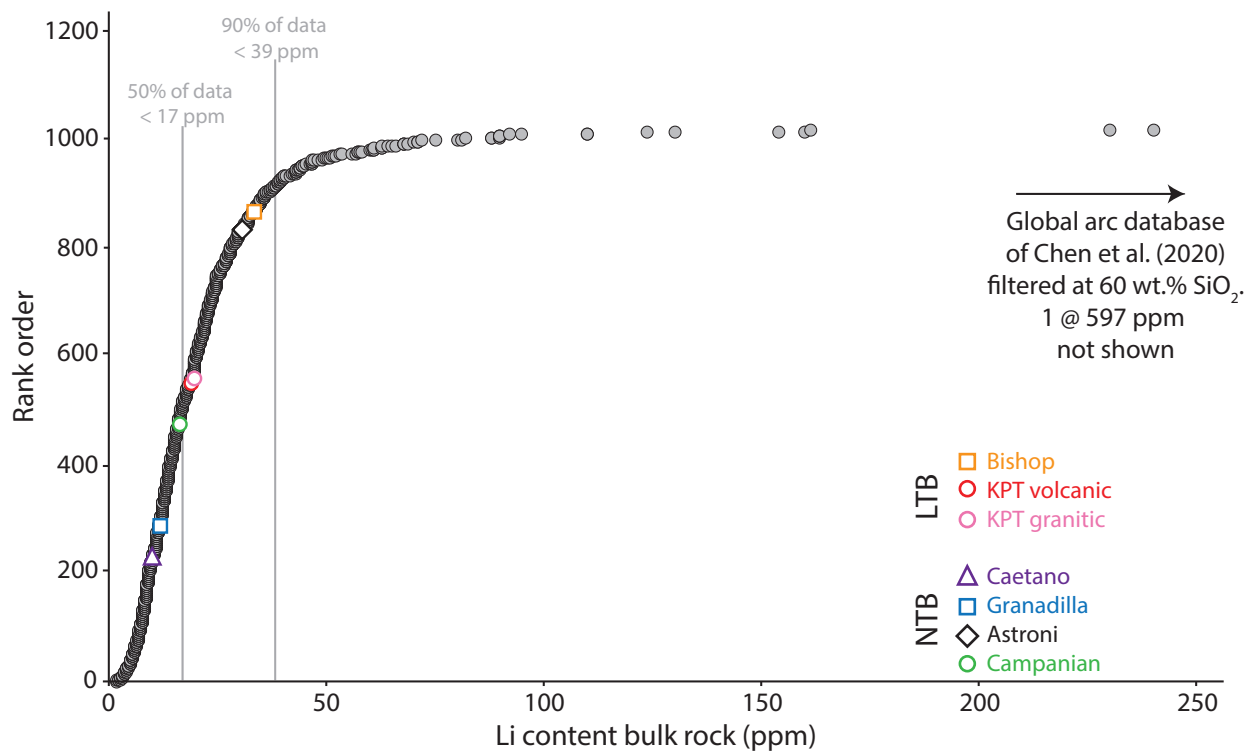
Supplemental Figures



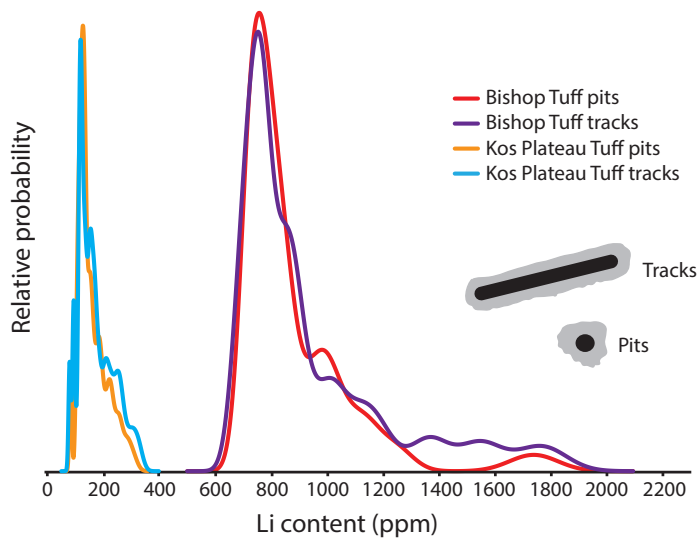
S1: Biotite compositions on a cation (a.p.f.u) basis, with LTB showing lower interlayer (K+Na) occupancy then NTB with additional data from Bryan (2006) and Forni et al. (2018).



S2: O isotopic compositions of the biotites in this study. Data from this study (coloured symbols) are converted to melt equivalent and then compared with melt equivalent $\delta^{18}\text{O}$ values from a range of mineral phases from the literature (shown in grey) for the Bishop, Campi Flegrei, and Kos cases. In the case of the Granadilla, Bryan (2002) measured $\delta^{18}\text{O}$ in biotite and so the comparison can be made directly without the need to calculate melt equivalent values. The biotite values presented by Bryan (2002) are in high-temperature equilibrium with co-existing feldspar.

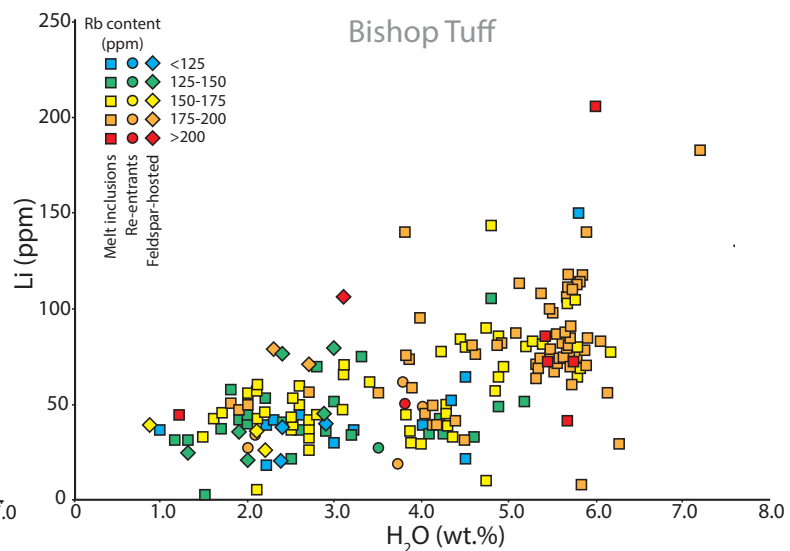
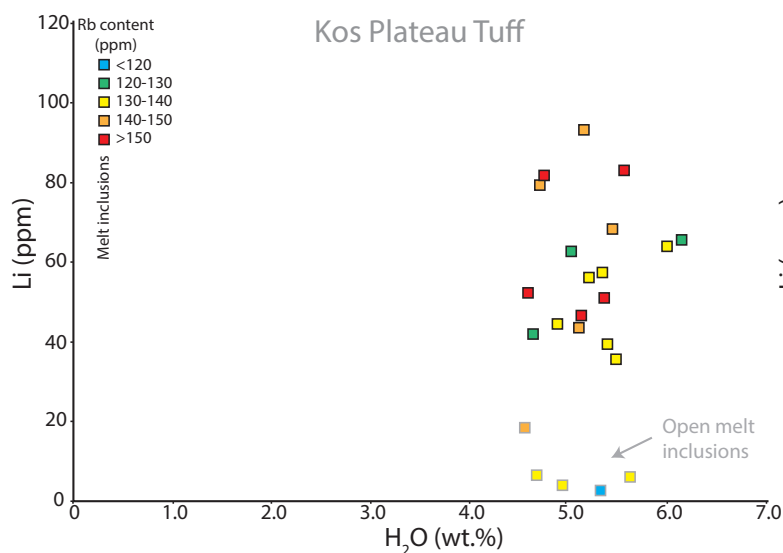


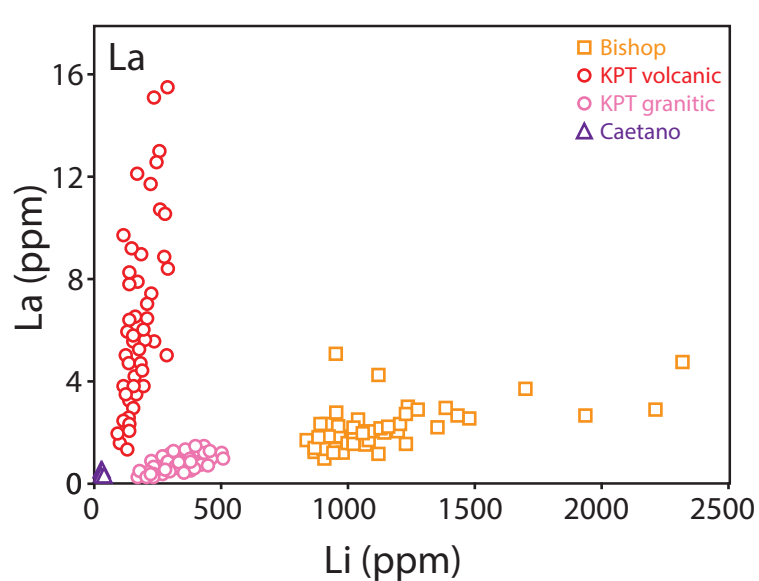
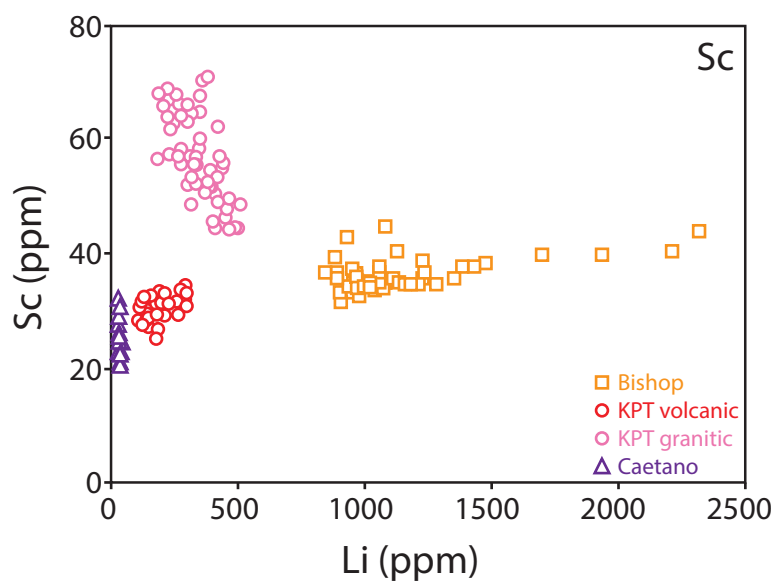
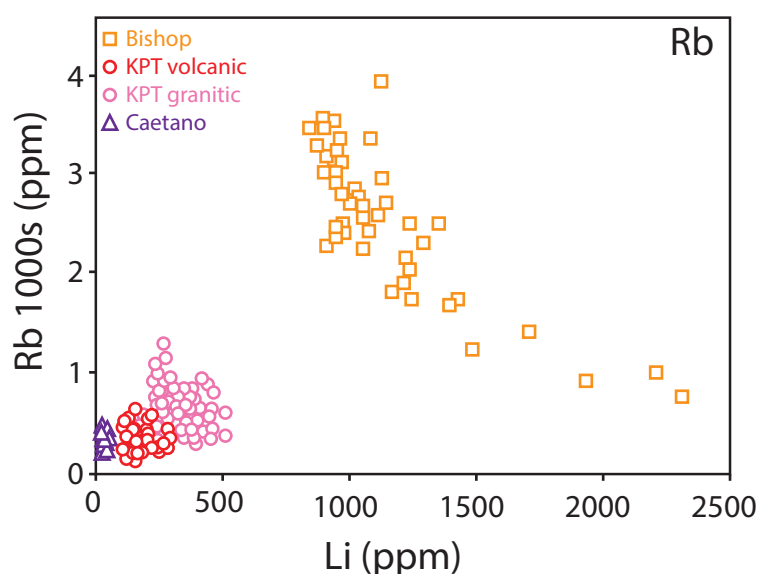
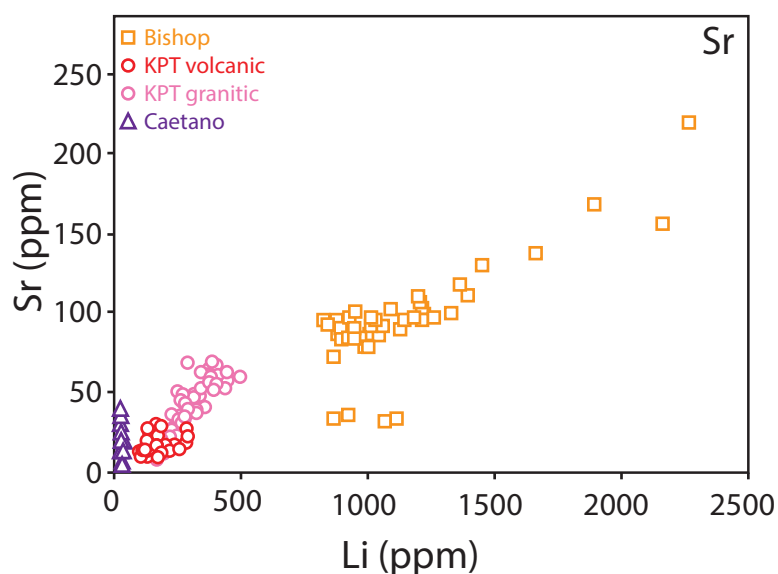
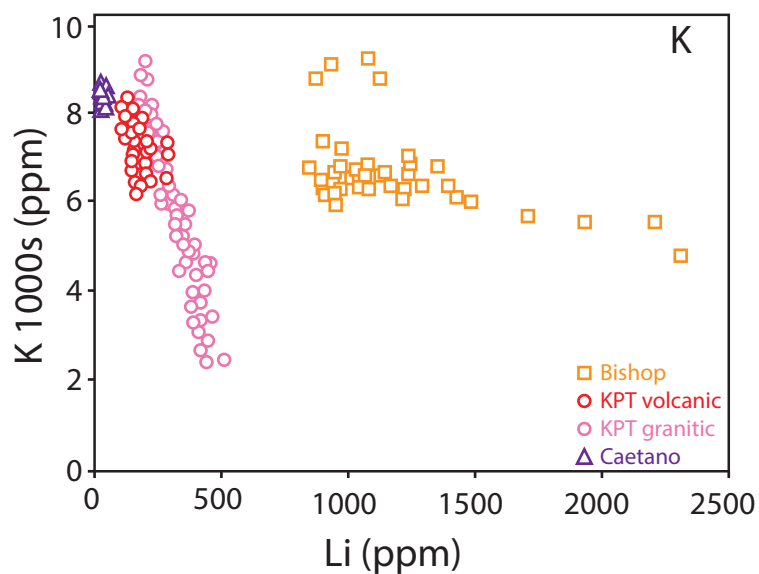
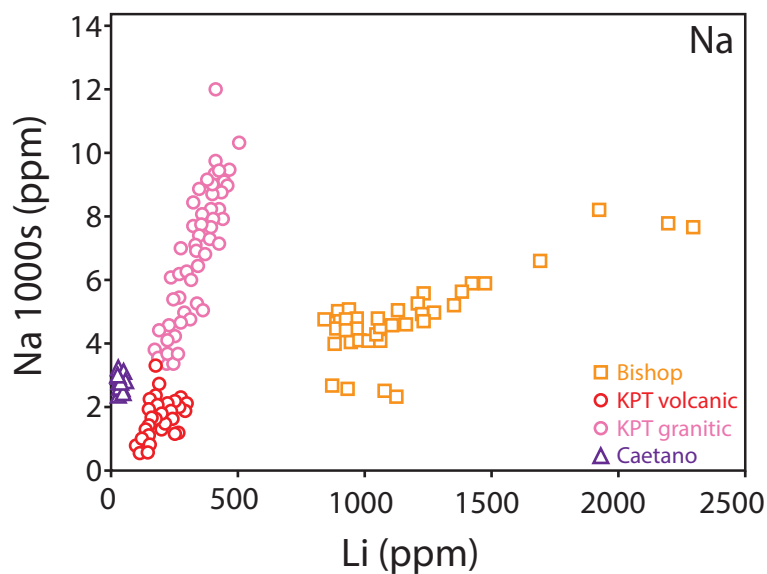
S3: Compilation of the Li contents of bulk rocks from arc suites, filtered at 60 wt.% SiO₂ (modified from Chen et al. 2020). The samples with Li-rich biotites from this study are not high in bulk rock Li.



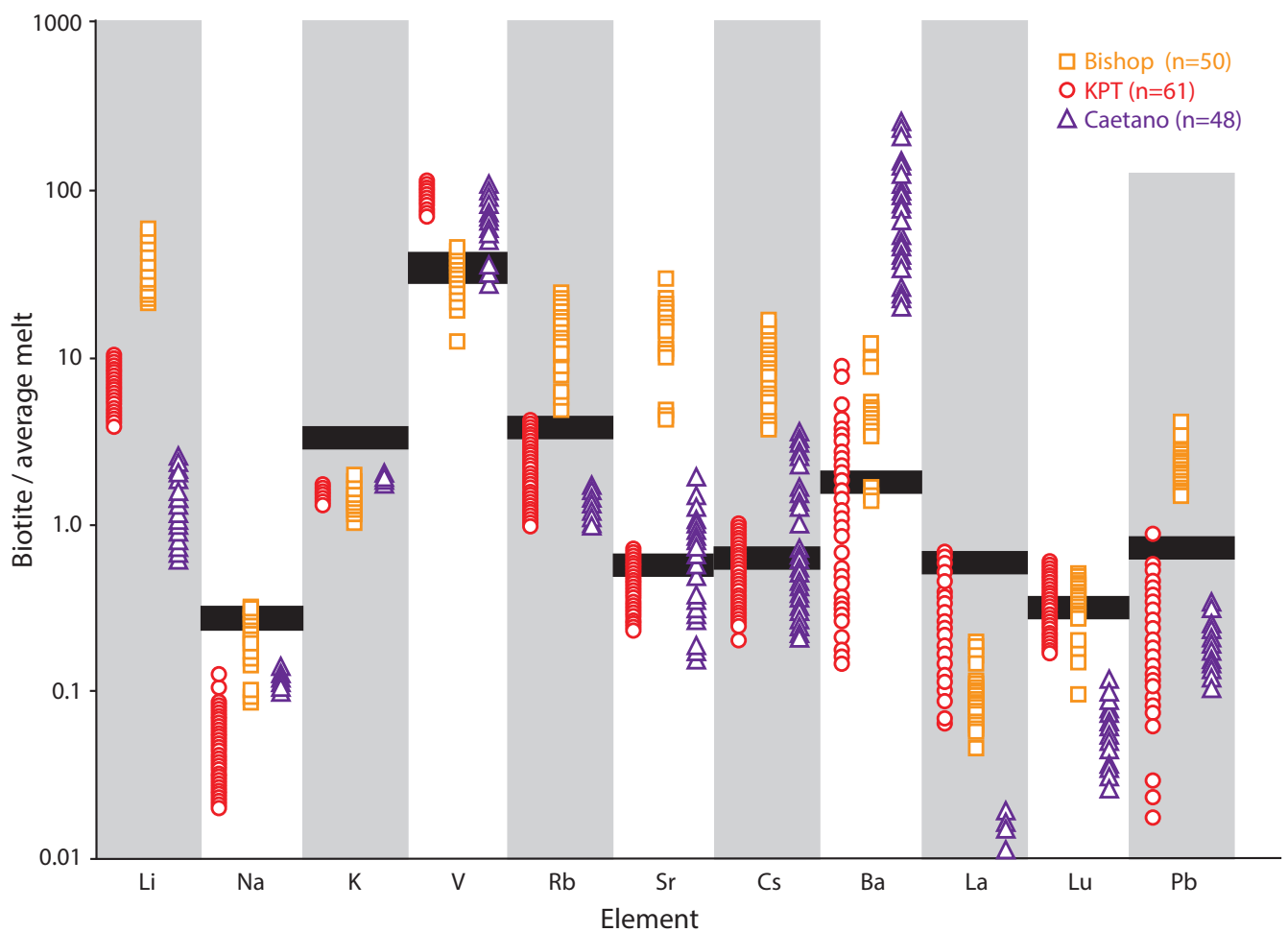
S4: Comparison of LA-ICPMS data from analyses via pits and tracks showing that the same range of Li compositions are found in both methods.

S5 (below): Published melt inclusion lithium contents of the Bishop Tuff (Dunbar and Hervig, 1992; Wallace et al. 1999; Myers et al. 2018) and Kos Plateau Tuff (Bachmann et al., 2009) illustrating that they are not high-lithium magmas.

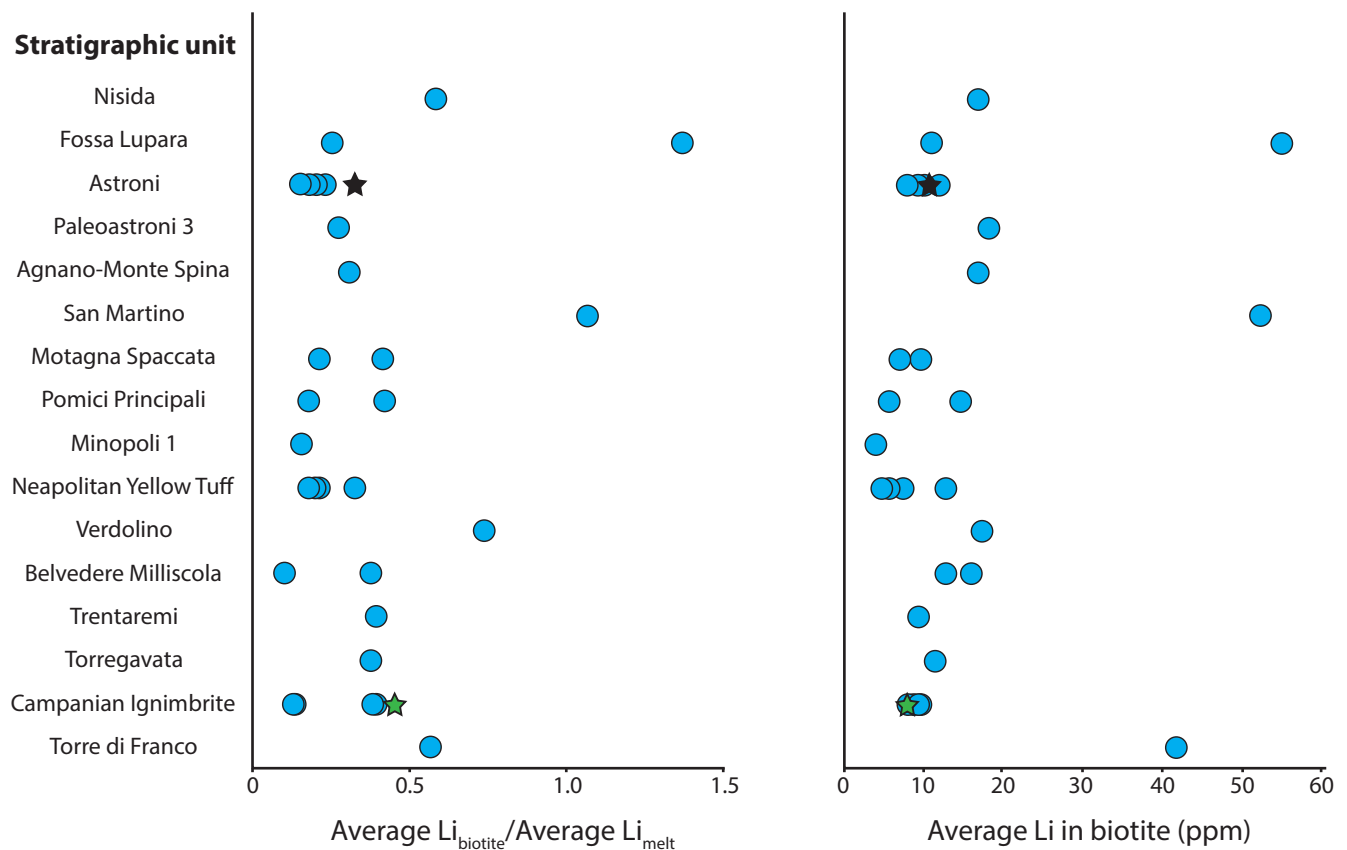




S6: Trace elemental data from the rhyolitic biotites in this study showing different behaviour even between the Kos and Bishop Tuff LTB.



S7: Apparent partitioning values for a range of elements from biotites using the average co-existing melt compositions. The black bands represent the experimentally derived values from Were and Keppler (2021). Unfortunately no data exist for lithium. The notably high values for Ba in the Caetano system are consistent with the cumulate remelting scenario noted by Watts et al. (2016). Such a cumulate melting behaviour is not captured in MELTS simulations and helps explain why Caetano forms NTB.



S8: Compiled Campi Flegrei data from Forni et al. (2018) illustrating that the Li partitioning into biotite from the whole volcanic field is similar to the examples we focus on (stars).

References

- Bindeman I.N., Valley J.W., 2002, Oxygen isotope study of the Long Valley magma system, California: isotope thermometry and convection in large silicic magma bodies. *Contributions to Mineralogy and Petrology* 144, 185-205.
- Iovine R.S., Mazzeo F.C., Worner G., Pelullo C., Cirillo G., Arienzo I., Pack A., D' Antonio M., 2018, Coupled $\delta^{18}\text{O}$ - $\delta^{17}\text{O}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ isotope compositions suggest a radiogenic and ^{18}O -enriched magma source for Neapolitan volcanoes (Southern Italy). *Lithos* 316-317, 199-211.
- Halliday A.N., Fallick A.E., Hutchinson J., Hildreth W., 1984, A Nd, Sr and O isotopic investigation into the causes of chemical and isotopic zonation in the Bishop Tuff, California. *Earth and Planetary Science Letters* 68, 379-391.
- Pappalardo L., Mastrolorenzo G., 2012, Rapid differentiation in a sill-like magma reservoir: a case study from the campi flegrei caldera. *Scientific Reports* 2, 712.
- Were P., Keppler H., 2021, Trace element fractionation between biotite, allanite, and granitic melt. *Contributions to Mineralogy and Petrology* 176, 74.