## An internal isochron method for U-Th-Raman thermochronology

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Phase mapping of compositionally zoned zircon by Raman spectroscopy produces detailed maps of alpha damage (Fig. 1a). These maps look nearly identical to the spatial distribution of actinide concentration determined by LA-ICP-MS (Fig. 1b). Plotting Raman shift against effective uranium concentration (eU = U + 0.235 Th) produces simple exponential trends between crystalline and metamict endmembers, which are characterised by ER peak locations of ~356 cm<sup>-1</sup> and ~351.3 cm<sup>-1</sup>, respectively (Fig. 1c). These exponential trends can be linearised using a simple logistic transformation of the Raman shifts, and parameterised by linear regression. The exponential curve forms an isochron. It defines a relative timescale: doubling the age (from t' to 2t' in Fig. 1c) requires approximately half the eU concentration (from x to x/2) to produce the same Raman shift (y). More accurate results are obtained by considering the data in three dimensional U-Th-Raman space. The relative timescale can be converted to an absolute timescale by calibration against a reference material of known cooling age. The isochron method offers several advantages over 'conventional' U-Th-Raman methods:

1) improved reproducibility: the mapping approach avoids misregistration issues between the U,Th and Raman measurements, which would bias the result;

2) better precision: the analytical uncertainty of individual Raman shift measurements is difficult to quantify, but our repeat measurements indicate that it is high. In our linear regression approach, precision roughly scales with the square root of the number of measurements. This typically results in an order of magnitude improvement;

3) quality control: previous research has shown that partial annealing of radiation damage is a complex process with several phases. Should a zircon crystal be affected by this complexity, then this will result in detectable deviations from the simple exponential trend;

We applied the method to 44 zircons from three Indian samples. Our data exhibit excellent consistency within each zircon, and between different zircons from the same sample. Due to the aforementioned complexity of the annealing process, we prefer to refer to U-Th-He Raman dates as 'model ages'. With this caveat in mind, the method holds great potential, especially in detrital thermochronology.

