## Methods

## Glass major and trace element geochemistry

For correlation purposes glass was analyzed for major and trace element abundances in 57 and 30 samples, respectively. Tephra glasses were embedded in epoxy mounts and exposed via grinding and polishing. Major and minor elemental analysis was carried out by electron microprobe analysis (EMPA) using the JEOL JXA 8200 wavelength dispersive microprobe at the GEOMAR - Helmholtz-Centre for Ocean Research Kiel following the methods from Kutterolf *et al.* (2011). Basic settings for conducting EMPA include: 1) a calibrated measuring program based on international natural and synthetic standards, 2) a constant acceleration voltage of 15 kV, and 3) a beam defocused to 5 μm with a current of 6 nA to prevent Na-loss and counting times of 20 s for peaks and 10 s for backgrounds. Accuracy was monitored by measurements on reference materials Lipari Obsidian (rhyolite; Hunt & Hill, 2001) and Smithsonian basaltic standard “VGA” (Jarosewich *et al.*, 1980). A single spot was analyzed in each glass shard. 45 analyses of unknown glass shards were bracketed by two analyses on the references. Standard deviations of measured oxides are ˂1% for major and ˂5% for minor components.

Trace element concentrations of glass shards were determined by Laser Ablation Inductively Coupled Plasma - Mass Spectrometry (LA-ICP-MS) at the Institute of Earth Sciences at the Academia Sinica in Taipei (Taiwan). A Photon Machines Analyte G2 laser ablation system using a 193 nm ArF Excimer laser was set to a spot size of 16 to 30 μm using 5-10 J/cm2 energy density at 1-5 Hz repetition rate and coupled to a magnetic sector ThermoFinnigan Element XR ICP-MS mass spectrometer. International standard glass (BCR-2g) were measured between sample measurements to monitor accuracy and correct for matrix effects and signal drift in the ICPMS as well as for differences in the ablation efficiency between the sample and the reference material (Günther *et al.*, 1999). Concentrations of NIST reference material SRM 612 were used for external calibration. Calcium concentrations, measured by EMP, were used also as internal standards to calibrate the trace element analyses. For detailed measurement conditions see Stoppa *et al.* (2018) and Schindlbeck *et al.* (2018). The limit of detection (LOD) for most trace elements is generally <100 ppb, with LOD values for rare earth elements (REE) around 10 ppb. The analytical precision is estimated at ~10% (relative) for most trace elements.

## U-Th zircon dating

Based on a representative glass chemistry and comparatively large pumice clast size, sample G17-23 from proximal LCY ignimbrite was selected at a location ~21 km from the source for geochronological analysis of zircon interiors and rims by U-Th dating by secondary ion mass spectrometry (SIMS). Rim analysis of unpolished crystals provides crystallization ages of the outermost crystal face, a prerequisite for subsequent (U-Th)/He dating of zircon to determine the eruption age. Additionally, pumice samples from two other large-volume eruptions that bracket the LCY eruption (W- and I-falls) were used to determine zircon rim crystallization ages by SIMS.

Individual whole pumices were used when possible; however, when sizes of individual pumices were too small to yield sufficient zircon during heavy mineral separation, a composite sample of several small pumice clasts showing similar textural and color features was processed after ultrasonic cleaning of individual clasts. In order to separate zircon crystals from pumice separated from ignimbrite, pumice blocks were individually crushed and sieved to ˂200 µm fractions. Heavy minerals were concentrated by standard physical separation techniques that included washing and hydraulic panning. Zircon crystals were hand-picked with the help of a binocular microscope. In order to dissolve glass attached to zircon, crystals were treated for 5 minutes in 40% HF at room temperature. Glass-free crystals were pressed into indium (In) metal which allowed analysis of the outermost crystal faces, and thus the youngest crystallization event. Additionally, one sample was epoxy-mounted and polished until zircon mid-planes were exposed. Prior to analysis, a conductive Au coat was applied to the mounts.

Zircon U-Th dating was conducted on a CAMECA ims 1280-HR ion microprobe at the Institute of Earth Sciences at Heidelberg University, Germany. 238U-232Th-230Th isotopes in zircon were measured following analysis protocols described in Reid *et al.* (1997) with modifications according to Schmitt (2011). A mass-filtered 16O- primary ion beam with an intensity of 35-45 nA was focused to a spot of 30-35 μm. Secondary ions were accelerated at 10 keV with an energy bandpass of 50 eV and a mass resolving power (*m*/*m*Δ) of 6000. Due to higher secondary ion yields for actinide oxides over atomic species (Schmitt, 2011, Schmitt & Vazquez, 2017), intensities of 230ThO+, 232ThO+ and 238UO+ and reference species 90Zr2O4+ were measured. Simultaneous detection of 230ThO+ and 232ThO+ was achieved using electron-multiplier (EM) and Faraday cup (FC) detectors in order to reduce the analysis time up to ~30% compared to mono-collection analysis. Gains for both detectors were calibrated by measuring 180HfO+ in both detectors at the beginning of the session; no significant drift was found throughout the session. 230ThO+ intensities were corrected by subtracting the averaged intensities measured during each analysis on two mass fixed stations at 244.03 and 246.3 amu. The U-Th relative sensitivity factor (RSF) was calibrated based on secular equilibrium reference zircon AS3 (206Pb/238U age = 1099 Ma; Schmitz *et al.*, 2003). Interspersed analyses of the same reference zircon yielded a weighted mean value for the analytical session of (230Th)/(238U) = 1.008 ± 0.006 (1σ; mean square of weighted deviates MSWD = 1.88; number of spots n = 30). U-Th model ages were calculated from two-point isochrons defined by the U-Th zircon activity ratio and that of LCY whole-rock composition of (230Th)/(232Th) = 0.99 ±0.02 and (238U)/(232Th) = 0.86 ± 0.02 (Rose and Bornhorst, 1981).

## Zircon (U-Th)/He analytical procedures

The (U–Th)/He part of ZDD was conducted at the John de Laeter Centre (Curtin University), Perth, Australia, following protocols described in Danišík *et al.* (2012). Zircon crystals were plucked out from the In mounts and then photographed, their physical dimensions measured, and then packed into Nb microtubes which were loaded into the He extraction line. 4He together with other gases were extracted at ~1250°C under ultra-high vacuum using a diode laser, cleaned on Ti-Zr getters, spiked with pure 3He gas. 4He was measured by isotope dilution on a Pfeiffer Prisma QMS-200 mass spectrometer. A "re-extract" was run after each sample to verify the complete outgassing of the crystals. Helium gas results were corrected for blank, determined by heating empty microtubes using the same procedure. After the 4He measurements, tubes containing the crystals were retrieved from the laser cell, spiked with 235U and 230Th, and dissolved in Parr bombs using HF and HCl (Evans *et al.*, 2005). Sample, blank, and spiked standard solutions were analyzed by isotope dilution for 238U and 232Th, and by external calibration for 147Sm on an Agilent 7500 ICP-MS. The total analytical uncertainty (TAU) was calculated as a square root of the sum of squares of uncertainty on He and weighted uncertainties on U, Th, and Sm measurements. The zircon (U–Th)/He ages were corrected for alpha ejection (Ft-correction) after Farley *et al.* (1996), whereby a homogenous distribution of U, Th, and Sm was assumed for the crystals. The accuracy of zircon (U–Th)/He dating procedure was monitored by replicate analyses of Fish Canyon Tuff zircon (n = 12) measured throughout this study as an internal standard, yielding a mean (U–Th)/He age of 28.4 ± 0.9 Ma (1σ uncertainties stated throughout this study, which is in good agreement with the reference (U-Th)/He age of 28.3 ± 1.3 Ma (Reiners, 2005) and a U-Pb age of 28.48 ± 0.03 Ma (Schmitz & Bowring, 2001).

In addition to the Ft-correction, the Ft-corrected (U–Th)/He ages were corrected for disequilibrium and pre-eruptive crystal residence by using the MCHeCalc software (Schmitt *et al*. 2010). The software requires as input parameters the Ft-corrected (U–Th)/He ages and uncertainties, crystallization ages and uncertainties, and D230 and D231 parameters describing zircon-melt fractionation of Th and Pa relative to U. D230 was calculated by dividing measured Th/U ratios of zircons by measured whole-rock/glass Th/U; for D231 a Pa/U zircon–rhyolite melt partitioning ratio of 3 was adopted.

In order to avoid overestimation of the disequilibrium correction, zircon (U–Th)/He ages were further corrected following the method of Friedrichs *et al.* (2021) that accounts for intra-grain variations in 238U-230Th disequilibria resulting from protracted crystallization where interiors can be significantly older than rims. The average time difference between rim and core age crystallization modes for LCY ignimbrite zircon population is *ca*. 9 ka, which translates into a 238U-230Th disequilibrium isochron slope of ~0.08. This suggests that at the time of crystallization of outermost rim domains, zircon cores already lost ~8 % of their initial 230Th deficit. Approximating a typical zircon crystal as a sphere with radius of ~45µm (Friedrichs *et al.*, 2021), the outermost ~9 μm thick shell represents ~50 % of its total volume in which, due to alpha ejection, only ~33 % of its total retained 4He is accumulated. Therefore, disequilibrium corrections as calculated by the MCHeCalc software (Schmitt et al., 2010) were only fully applied to 33 % of accumulated 4He (from the outermost 9 μm shells of zircon crystals), but reduced for 8 ± 8 % for the remaining fraction (from generally older zircon cores, Fig. 8a). Inferring a theoretical interface between rim and core domains at a depth of 9 μm (and thus 50 % of crystal volume) is justified by analyzing both the youngest (rims) and oldest zircon growth (cores). Uncertainties of the MCHeCalc Monte Carlo simulations accounting for disequilibrium corrections as well as these population-based interior age corrections were propagated into final best estimate (U–Th)/He age uncertainties. Both Ft-, disequilibrium-, and population-based interior age corrections assume a homogenous distribution of U, Th, and Sm within the zircon crystals, which is justified by overlapping U and Th abundances between rim and core analyses (Supporting Information Table 1).

The eruption age for single samples was calculated as the error-weighted mean from the disequilibrium corrected single-grain (U-Th)/He ages and standard error using IsoplotR (Vermeesch, 2018). However, the final LCY eruption age was calculated from combining the disequilibrium corrected single-grain (U-Th)/He ages of the three analyzed LCY samples (n = 48 grains). Mean square weighted deviation (MSWD) values were >1 in all samples suggesting non-analytical scatter e.g., due to heterogeneity in U abundances; in order to provide a conservative age uncertainty, all standard errors of the weighted average were multiplied by the square-root of the MSWD.

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