Detailed analytical methods

In situ zircon U-Pb isotopes, trace element contents and Lu-Hf isotopes were analysed at ETH Zürich. U-Pb isotopes were measured simultaneously with trace elements (from the same ablated volume) using a Thermo Element XR SF-ICP-MS coupled with a Resonetics Resolution 155 type ablation system. We used a 30 μm spot size, 5 Hz repetition rate, 2.0 J/cm^2 energy density (fluence) and 40 s ablation time after five cleaning pulses and 30 s of gas blank acquisition. We performed the analyses in six sessions during which the analytical conditions were similar. For U-Pb dating, GJ-1 reference zircon (Jackson et al., 2004, Horstwood et al., 2016) was used as a primary reference material, while zircons 91500, Plešovice, AUSZ7-1, AUSZ7-5, Rak-17, Temora2 and LG-0302 were measured as validation reference materials (Wiedenbeck et al., 1995; Sláma et al., 2008; Kennedy et al., 2014; von Quadt et al., 2016, Webb et al., 2020; Black et al. 2004; Gutiérrez et al. 2018, respectively). The average precision of these reference materials (RM) ranged from 0.9% to 3.2% (2 SE) in case of secondary RM older than 30 Ma, while the younger have 6.7–12.5% (2 SE) average precisions (further details are in Supplementary Data, Table S2). Validation reference materials for dating, covering an age interval between 1063.5 Ma and 2.409 Ma, were used to correct the matrix dependent age offsets following the procedure described in Sliwinski et al. (2017). Data reduction of in-situ dating by LA-ICP-MS was done using IOLITE (Paton et al., 2011) combined with VizualAge (Petrus and Kamber, 2012). The Th disequilibrium correction was performed after alpha dose-correction (Sliwinski et al., 2017) assuming a constant Th/U partition coefficient ratio of 0.33 ± 0.06 (1σ; Rubatto and Hermann, 2007) and using the equations of Schärer (1984). The in-situ dates were not corrected for common Pb contents; however, during data reduction, integration intervals were set to exclude the common Pb contaminated signal intervals and data were filtered according to their discordance ([(^{207}Pb/^{235}U Age)–(^{206}Pb/^{238}U Age)]/(^{207}Pb/^{235}U Age) < 10%). Average uncertainty of the individual zircon dates is given as 2 SE and it is between 1.4 and 2.3%. For the in-situ zircon trace element analysis we used NIST610 as primary reference material and zircon 91500 for quality control, and an in-house synthetic reference material (Synthetic Zircon Blank) to correct the Nb concentrations. Target elements were Si, Zr, REE, Y, Hf, P, Nb, Ta, U, Th, Ti (see Supplementary Data, Table S2), and either Al or Ba were measured for monitoring glass, apatite inclusions. Si (15.2 wt% in zircon) was used as internal standard for data reduction done by IOLITE. Zircon compositions with Al above detection limit were discarded from the database.

Zircons from four key samples were also dated by chemical abrasion isotope dilution thermal ionisation mass spectrometry (CA-ID-TIMS) at ETH Zürich. Select grains, most of which were previously analysed by LA-ICP-MS, were extracted from zircon mounts and individually annealed in a muffle furnace at 900 °C for 48 h. The annealed crystals were then subjected to chemical abrasion (Mattinson, 2005) at 190 °C for 12 hours in 29 M HF in individual 200 µl PFA microcapsules placed in a Parr digestion vessel. The zircons were then kept on a hotplate at 80 °C in 6 M HCl for several hours, followed by repeated rinsing with HNO_3. The crystals were then loaded into individual 200 µl PFA microcapsules, spiked with the EARTHTIME ^{205}Pb-^{233}U-^{235}U tracer solution (Condon et al., 2015; McLean et al., 2015) and dissolved in ca. 70 µl 29 M HF and trace HNO_3 in a Parr digestion vessel at 210 °C for 60 h. Following dissolution, the samples were dried down and converted to chloride by placing them back in the oven overnight in 6 M HCl. The solutions were then dried down and re-dissolved in 3 M HCl, and purified to U and Pb through anion exchange column chromatography (modified from Krogh, 1973). Once purified, the U and Pb fractions were combined in cleaned 7 ml PFA beakers and dried down with trace 0.02 M H_3PO_4 prior to loading on single outgassed zone-refined Re filaments with a Si-gel emitter (Gerstenberger and Haase, 1997). U and Pb isotope analyses were conducted with a Triton Plus TIMS. Both Pb and UO_2 analyses were performed using static multicollection in Faraday cups connected to 10^{13} Ω resistors, except for ^{204}Pb collected in the axial secondary electron multiplier (von Quadt et al., 2016). $^{18}O/^{16}O$ in uranium runs was determined online by monitoring mass 272 (Wotzlaw et al., 2017). Mass fractionation of Pb was corrected using an instrumental mass fractionation factor derived from compiled analyses of ^{202}Pb-^{205}Pb-spiked samples. For U, instrumental mass fractionation was corrected with the known $^{233}U/^{235}U$ of the spike and assuming a sample $^{238}U/^{235}U$ ratio of 137.818 ± 0.045 (2σ; Hiess et al., 2012). All common Pb was considered laboratory blank and was corrected using the long-term isotopic composition of the Pb blank of the lab (Supplementary Data, Table S2). Data were processed with the Tripoli and Redux U–Pb software packages (Bowring et al., 2011; McLean et al., 2011). All $^{206}Pb/^{238}U$ dates were corrected for initial $^{230}Th/^{238}U$ disequilibrium using a fixed partition coefficient ratio derived from a compilation of natural and experimental data ($D^{Th/U}_{[zircon]}/D^{Th/U}_{[melt]}$ = 0.183 ± 0.061, 1σ) and assuming that variations in Th/U between zircons reflect changes in magma composition rather than in U/Th partitioning.

In-situ Lu-Hf isotopic analysis was performed using Resolution 193 nm ArF laser ablation system coupled to a Nu2 multicollector inductively-couples-plasma mass-spectrometer (MC-

ICP-MS) at the ETH Zürich. Ablation was carried out using He as a sweep gas with a flow rate of ~0.37 l/min and combined with Ar (~0.85 l/min) using a 50 µm spot size and a 5 Hz laser pulse repetition rate and an energy density of 4 J/cm^2. Data reduction was done by using IOLITE 4.0. Lutetium and Yb were analysed in order to correct for isobaric interferences on ^{176}Hf using ^{176}Yb/^{173}Yb = 0.796218 (Thirlwall and Anckiewicz, 2004). Mass bias correction coefficients for Hf, Yb, and Lu isotopes were determined based on the natural ratio of ^{179}Hf/^{177}Hf and ^{173}Yb/^{171}Yb (^{179}Hf/^{177}Hf = 0.7325; ^{173}Yb/^{171}Yb = 1.132685; Chu et al., 2002), respectively. The Lu mass bias fractionation was assumed to be the same as Yb. The accuracy and precision of the data obtained was monitored through the systematic measurements of the well characterized RM Mud Tank (0.282507; Woodhead and Hergt, 2005), 91500 (0.282307; Wu et al., 2006), Temora (0.282680; Wu et al., 2009), and GHR1 (0.283050; Eddy et al. 2019). The standard reference materials were chosen in order to have a range in Yb/Hf ratios to test the accuracy of the ^{176}Yb correction following the protocols of Fisher et al. (2014). Repeated standard analyses yielded results for Mud Tank: ^{176}Hf/^{177}Hf = 0.282492 ± 0.000021 (n = 83); 91500: 0.282309 ± 0.000039 (n = 31); Temora: 0.282675 ± 0.000032 (n = 28); GHR1: 0.283061 ± 0.000030 (n = 27) with uncertainties reported as 2 standard deviations. Initial ^{176}Hf/^{177}Hf ratios and εHf were calculated using the crystallization age of each sample, the decay constant of Söderlund et al. (2004) for ^{176}Lu (1.867x10^{-11} a^{-1}) and the CHUR parameters of Bouvier et al. (2008). All uncertainties are reported at the 2 sigma level.

Bulk rock composition of pumices and pyroclastic rocks was determined at the Bureau Veritas Mineral Laboratories (ACME Labs; http://www.acmelab.com/) using ICP-OES and ICP-MS technique for major-minor and trace elements, respectively. Internal standards and duplicate sample analysis by sessions were used to check the reliability of the results.

Major and trace element composition of volcanic glass was analysed for each pyroclastic unit, except for SAU (where no fresh glasses were found) and from the Lénárddaróc outcrop. Major elements in 20-22 glass shards per sample were analyzed at GEOMAR (Kiel, Germany) using JEOL JXA 8200 electron microprobe. The analytical conditions were 15 kV accelerating voltage, 6 nA current and 5 µm electron beam size for all analyses. The conditions correspond to the electron beam current density of 0.076 nA µm^{-2} on sample, which is within the recommended range (<0.1 nA µm^{-2}) to minimize the Na loss during analysis, especially in a combination with short (5s in our work) counting time for Na (e.g., Kuehn et al., 2011). Full analytical details and data on long-term analytical precision can be

found in Portnyagin et al. (2020). Some elements (e.g. Ti, Fe, Mn, Mg), which are typically analysed by electron microprobe with high precision, occur in the glasses studied at very low concentrations approaching and below the quantification limit of microprobe (typically 200-300 ppm at the given analytical conditions). High precision quantitative data for these elements are provided by LA-ICP-MS analyses. Data on reference glasses analyzed along with the samples in this study are provided in Supplementary Data, T table S3.

Trace element analyses were obtained at the Institute of Geosciences, Christian-Albrecht University of Kiel (Kiel, Germany) using ICP-MS Agilent 8900 and a Coherent GeoLas ArF 193 nm Excimer LA system operated with a fluence of 5 J cm^{-2}, at a repetition rate of 10 Hz and 24 μm ablation craters. Analyses were performed using a Rotatable Channel Cell (RCC) sample chamber (Wuhan Sample Solution Analytical Technology Co. Ltd.). Helium (0.7 L min^{-1}) with addition of 14 mL min^{-1} H$_2$ were used as carrier gas. The carrier gas was mixed with Ar (~1 L min^{-1}) prior to introduction to the ICP-MS. Ten major elements (Si, Ti, Al, Fe, Mn, Mg, Ca, Na, K, P) and 31 trace elements were analyzed. Analyses included 20 s background (laser-off) and 30 s signal (laser-on) measurements. Dwell time for different elements varied from 5 to 20 ms depending on their abundance. One complete measurement cycle lasted 0.607 ms and initial data reduction was performed in Glitter software (Griffin et al., 2008), that included manual selection of intervals for signal integration and preliminary calibration. The intensities corrected for background and averaged over the selected intervals (typically 10-20 s) were normalized to the intensity of ^{43}Ca isotope and converted to concentrations by matching the sum of major element oxides to 100 wt% (e.g. Pettke et al., 2004). Some data for phosphorus have been excluded due to unexpected contamination by the epoxy EpoThin-2 from Buehler used in this study. The previously distributed epoxy EpoThin-1 from the same provider had negligibly low P content and has been used in a numerous works (e.g. Portnyagin et al., 2020 and references therein). The calibration and correction of instrumental drift used data on ATHO-G reference glass (Jochum et al., 2006), which was measured in duplicate after every 18 points on unknown samples. Typically, 10-12 glass shards were analyzed for every sample. Data on reference glasses analyzed along with the samples in this study are provided in Supplementary Data, Table S3. Further analytical details can be found in (Portnyagin et al., 2020).

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