Supplemental Info: Zircon U-Pb geochronology, Hf isotope and trace element concentration analytical methods using UCSB Laser-ablation Split Stream (LASS) Analysis

Sample preparation

For U-Pb geochronology, dateable mineral phases, including zircon, were separated from bulk bedrock samples by standard methods including crushing and pulverizing, water shaking table, heavy liquids, and magnetic separation. Zircons from plutonic rocks were hand-picked and placed on double-sided tape on glass plates, and then mounted in epoxy 2.5 cm rounds. Epoxy mounts were ground to expose grain interiors, polished, and digitally imaged using a camera mounted to a binocular picking microscope.

Analytical Procedures, Standard Reference Materials Results, and Analytical Uncertainty

U-Pb ratios, Hf isotopes, and trace-element compositions for zircons were measured simultaneously at University of California, Santa Barbara (UCSB) by laser ablation split-stream (LASS; Kylander-Clark et al., 2013) in May 2018 using the Photon Machines Analyte 193-nm excimer laser, ablating sample material using a 10 Hz repetition rate and a set fluence for a single session that varied between ~1-2 J/cm^2. Spot sizes ranged from 65um to 50um, depending on grain size of sample zircons. Spot sizes and laser-run conditions for each sample are given in geology_LASS_Geochronology_DarbyMtns_AK_Drewes.csv. The laser was fired twice to remove surface contamination (primarily common Pb), and this material was allowed to wash out for 40 seconds. Hf (and Lu) isotopes were measured on the Nu Instruments Nu Plasma 3D (HR-MC-ICP-MS) by static measurement of masses 180 to 171 on Faraday cups. U-Pb-Th isotopes (^{238}U, ^{232}Th, ^{208}Pb, ^{207}Pb, ^{206}Pb, ^{204}Pb+^{204}Hg) and trace-element compositions were measured on an Agilent 7700x quadrupole ICP-MS. Trace-elemental abundances are determined by counting ^{31}P, ^{49}Ti, ^{88}Sr, ^{89}Y, ^{93}Nb, ^{139}La, ^{140}Ce, ^{141}Pr, ^{146}Nd, ^{147}Sm, ^{153}Eu, ^{157}Gd, ^{159}Tb, ^{163}Dy, ^{165}Ho, ^{177}Er, ^{169}Tm, ^{172}Yb, ^{175}Lu, ^{178}Hf, ^{181}Ta, collected in time-resolved, single point-per- peak mode (dwell times set to from 10 to 5ms), and are calibrated to ^{90}Zr (Zr = 48 wt%) with analysis of matrix-matched reference materials (e.g., 91500 and GJ1) interleaved among samples. Spectrometry methods and configuration is detailed in Kylander-Clark et al. (2013). All raw mass spectrometry data acquired during laser ablation were processed using Iolite (ver. 2.5; Woodhead and Hergt, 2005; Woodhead et al., 2007; Paton et al., 2010; 2011). Peak mass bias corrections and peak stripping were done using a modified version of the Iolite Hf routine, following recommendations of Fisher et al., (2014).

Sample spot analyses were bracketed before and after every eighth spot by analyses of zircon reference materials (RM) 91500 (Weidenbeck et al., 1995) plus a rotation of GJ-1 (Jackson et al., 2004), Mud Tank (Black and Gulson, 1978), Plešovice (Slama et al., 2008), and Temora2 (Black et al., 2003, 2004). The 91500 zircon was used as the primary RM for U-Pb geochronology and trace-element analyses. Mud Tank (U-Pb age = 733.9 ± 3.0, MSWD = 1.22; ^{176}Hf/^{177}Hf = 0.282490 ± 0.000012; n = 58) was used as the primary RM for Hf isotopes. However, no Hf isotope correction was applied because measured Hf isotope ratios of Mud Tank agree within analytical uncertainty of the accepted ratio ^{176}Hf/^{177}Hf = 0.282507 ± 0.000006, which corresponds to ^{176}Hf/^{177}Hf = 0.282160 for JMC475 (Woodhead and Hergt, 2005). Uncorrected Hf isotopes for other standards run during this session, including 91500 (U-Pb age = 1062.2 ± 1.4, MSWD = 0.78; ^{176}Hf/^{177}Hf = 0.282309 ± 7, MSWD = 0.54; n = 405), GJ-1 (U- Pb age = 603.7 ± 1.2, MSWD = 0.85; ^{176}Hf/^{177}Hf = 0.282013 ± 9, MSWD = 0.57; n = 177), Plešovice (U-Pb age = 338.6 ± 0.9, MSWD = 0.90; ^{176}Hf/^{177}Hf = 0.282490 ± 7, MSWD = 0.90; n=114), and Temora2 (U-Pb age = 421.3 ± 1.2, MSWD = 1.0; ^{176}Hf/^{177}Hf = 0.282668 ± 9, MSWD = 1.0; n=113), were in even better agreement relative to assumed values (Woodhead and Hergt, 2005; Slama et al., 2008; Morel et al. 2008). RM zircon GJ-1 was a backup standard for trace elements, with Plešovice and Temora2 as secondary geochronology standards, and the synthetic zircon standards MUNZirc1 and MUNZirc4, typically run as part of a RM block at the beginning of each analytical session (Fisher et al., 2011; 2014), served as additional secondary Hf isotope standards. Two-sigma analytical uncertainties associated with U-Pb ages reported here are better than the approximately 2 percent long-term empirical scatter of ages measured at the UCSB laboratory.

Internal uncertainty (2 times the percent standard error, hereafter %2SE) for trace element concentrations, based on repeated analyses of SRM GJ1 interleaved through all runs during this session, is about ±6% for Hf, 25% for U, Th, and the heavy REE (HREE) +Y, ± 30% for the middle and most light REE (MREE and LREE), but higher for Pr and La, where abundances are at or below detection limits, similar to uncertainties reported in Kylander-Clark et al. (2013). Across all sessions that E. Todd acquired data from this lab, GJ1 analyses at all spot sizes have calculated external reproducibility (2 times the percent standard deviation, hereafter %2SD) for the REE's (except La, Pr), Ti, Y, and Nb are roughly linearly correlated with mean of internal uncertainties (%2SE) for these elements, with a slope of ~1 and an intercept at about 2%. This indicates that empirical uncertainty may be estimated for all samples to be at least about 2% higher than the internal uncertainty for most elements. Using this same approach, but limiting to 65um spot sizes only, the slope remains ~1, but the intercept is near zero (i.e., %2SD is roughly equal to %2SE), whereas 50um spot sizes (slope also ~1) have an intercept near 4% (i.e., %2SD is approximately equal to %2SE +4%).

Data Presentation and Screening

All spot analyses are shown in geology_LASS_Geochronology_DarbyMtns_AK_Drewes.csv. Data in the table are generally arranged by sample ID, with individual spot analyses listed in the order they were analyzed. In a few cases where there are discreet age populations among analyzed grains from a single sample, spot analyses are reorganized in order of increasing U-Pb age. Ages shown on geology_LASS_Geochronology_DarbyMtns_AK_Drewes.csv include the $^{206}Pb/^{238}U$-calculated age, the $^{207}Pb/^{206}Pb$-calculated age, and a "Preferred Age," which for all samples included here is the ^{207}Pb-corrected $^{206}Pb/^{238}U$ age. All ages were calculated using Isoplot 4.15 (Ludwig, 2002).

A summary of all U-Pb ages, Lu-Hf isotopic data, and trace-element analyses is also included in the accompanying data distribution in geology_LASS_SampleSummary_DarbyMtns_AK_Drewes.csv. Results in this table provide summary data for a single sample, with the Date and Timestamp fields for the most recent analysis (within one session, or among sessions in instances where a sample was analyzed across more than one session). Sample entries in Geology_LASS_SampleSummary_DarbyMtns_AK_Drewes.csv show representative weighted-mean and uncertainty (2 sigma) for U/Pb ages and Hf isotopes, determined using Isoplot 4.15 (Ludwig, 2002; revised 2012), and median trace-element compositions, each calculated for the youngest population for a single sample. These younger "main" populations are considered most likely to be contemporaneous with crystallization of the host rock, and are distinguished from any "inherited" grains, or grains from metamorphic zircon-forming events. Grains with discordant ages (that is, a ratio of $^{206}Pb/^{238}U$- and $^{207}Pb/^{235}U$- calculated ages of <0.80 or >1.05) were typically excluded from calculated weighted mean ages, hafnium isotope ratios, and representative trace-element concentration medians, except in cases where noted for nominally discordant grains with ages consistent with concordant grain populations (geology_LASS_Geochronology_DarbyMtns_AK_Drewes). Grains with elevated U/Th (>10), generally thought to be consistent with a metamorphic origin, were excluded from calculated values in geology_LASS_SampleSummary_DarbyMtns_AK_Drewes.csv only in cases where they varied significantly from otherwise low U/Th populations.

Trace-element median values in geology_LASS_SampleSummary_DarbyMtns_AK_Drewes.csv additionally exclude zircon grains with anomalous trace-element concentrations that were presumed to have been imparted on the signal by ablating non-zircon nano-inclusions within zircon grains (for example, apatite or ilmenite). This impurity monitoring typically used Si, P, Ti, V, Sr, and REE concentrations, but the monitor trace elements varied among different sessions, so in some cases more elements were available for inclusion monitoring than in others. Normalized REE patterns and MREE and HREE concentrations are also used to determine if uncontaminated zircon grains represent a single population. The consideration of median trace-element concentrations for a zircon population additionally excludes La and Pr concentrations from a single grain in cases where $(La/Pr)_N$ and/or $(Pr/Nd)_N$ >1.0. Anomalous La and/or Pr alone was not used as criterion to screen for inclusions because these elements are disproportionately sensitive to contamination due to 1) their presence in such low concentrations (typically 10's of ppb) in normal zircon, so even trivial proportions of inclusions (many of which have high LREE, such as apatite or monazite) may disproportionately affect La and Pr several orders of magnitude above normal "zircon" abundances, and 2) the particularly large relative analytical uncertainty for these low-concentration elements. Grains with anomalous trace elements, where nano-inclusions were suspected, did not obviously impart aberrant weighted mean U-Pb ages or Hf isotope ratios (likely due to extremely high Hf $D_{zircon/melt}$), so nano-inclusion filtering did not extend to the omission of ages or Hf isotopes measured on a single spot from their respective weighted means.

Spot analyses excluded from calculations of weighted mean ages and Hf isotope ratios, and median trace-element concentrations shown are indicated in geology_LASS_Geochronology_DarbyMtns_AK_Drewes.csv by "X" in either of three columns: "**U/Pb_x_from_wtmn**", "**Hf_x_from_wtmn**", and "**TE_x_from_median**", respectively. The column labeled "**Comment_exclude**" in geology_LASS_Geochronology_DarbyMtns_AK_Drewes.csv provides a short annotation justifying their exclusion from the calculated representative values (e.g., "disc." if discordant, "Inherited" if ages are older than the main/youngest grain population ages, and/or "P_LREE (Apt)" if phosphate nanomineral inclusions are indicated by trace element concentrations, etc.).

Hafnium isotope epsilon (εHf) values and age-corrected Hf isotope ratio ($^{176}Hf/^{177}Hf_{(T)}$) and epsilon values ($\varepsilon Hf_{(T)}$) were calculated assuming chondritic $^{176}Hf/^{177}Hf$ and $^{176}Lu/^{177}Hf$ ratios of 0.282785 and 0.0336 (Bouvier et al., 2009) and a lutetium-176 decay constant of $1.86x10^{-11}$ (Scherer et al., 2001) (age-corrected values were calculated for spots with concordant ages only). Model ages (TDM_{Hf}), in billions of years, were calculated assuming present-day depleted mantle $^{176}Lu/^{177}Hf$ and $^{176}Hf/^{177}Hf$ values of 0.0384 and 0.28325 (Griffin and others, 2000).

Equilibrium crystallization temperatures (˚C) for zircons (see above) were calculated using the Ti-in-zircon thermometer, after Ferry and Watson (2007), assuming SiO_2 activity ($aSiO_2$) of 1.0 for silica saturated rocks and 0.7 for silica under-saturated rocks; TiO_2 activity ($aTiO_2$) is assumed to be 0.7. Temperatures in geology_LASS_SampleSummary_DarbyMtns_AK_Drewes.csv are calculated from representative median Ti concentrations of a sample population (i.e., they are not the mean of single-grain temperatures in geology_LASS_Geochronology_DarbyMtns_AK_Drewes.csv).

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