

1. Methods

1.1. Whole rock geochemistry

Representative whole rock powders from 34 samples were prepared using an agate mill and major and trace elements were measured using a Philips PW2400 X-Ray Fluorescence (XRF) spectrometer at the University of Lausanne, Switzerland. The NIMN, NIMG, BHVO and SY2 standards were used for quality control. Glass-fused disks prepared for XRF analyses were fragmented and mounted for additional analyses of trace and rare earth elements (REE) by LA-ICP-MS. Measurements were made using an Agilent 7700x quadrupole ICP-MS, and depending on the expected concentration within samples, either NIST SRM 610 or 612 reference material glasses were used as external standards. The laser settings employed in these analyses employed a 10 Hz repetition rate and a spot size between 80 to 120 μm . Blanks were measured for $\sim 90\text{s}$, after which the laser was switched on and the signal was measured for 45s. The Sr or Al_2O_3 concentrations (as previously determined by XRF) were used as an internal standard. Each sample was ablated three times, and average concentrations were calculated offline using LAMTRACE (Jackson, 2008). The uncertainties of three spots per sample are $\pm 10\%$ for rare earth elements (REE) and $\pm 5\%$ for other trace elements. Whole rock compositions have been normalized to an anhydrous state in all diagrams.

1.2. Sr-Nd-Pb whole rock isotopes

100 mg of whole rock powder was dissolved in 4 ml of concentrated HF and 1 ml of 15 M HNO_3 in closed Teflon vials at 140°C for seven days. The samples were dried down and re-dissolved in 3 ml of 15 M HNO_3 before being dried down again. Sr–Nd–Pb chemical separation followed the methods described in Pin and Santos-Zalduegui (1997) and Chiaradia et al. (2011). Isotopes of Sr, Nd and Pb were analysed at the University of Geneva using a Thermo Neptune PLUS Multi-Collector ICP-MS following the methods described by Beguelin et al. (2015) and Chiaradia et al. (2015). Isotopic ratios were corrected for internal fractionation using $^{88}\text{Sr}/^{86}\text{Sr}=8.375209$ for the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio, $^{146}\text{Nd}/^{144}\text{Nd}=0.7219$ for the

$^{143}\text{Nd}/^{144}\text{Nd}$ ratio and $^{203}\text{Tl}/^{205}\text{Tl}=0.418922$ for the three Pb ratios (a Tl standard was added to the solution). SRM987 ($^{87}\text{Sr}/^{86}\text{Sr}=0.710248$, long-term external reproducibility: 10 ppm), JNdi-1 ($^{143}\text{Nd}/^{144}\text{Nd}=0.512115$; Tanaka et al., 2000; long-term external reproducibility: 10 ppm), and SRM 981 (Pb-isotopes; Baker et al., 2004; long-term external reproducibility of 0.0048% for $^{206}\text{Pb}/^{204}\text{Pb}$, 0.0049% for $^{207}\text{Pb}/^{204}\text{Pb}$ and 0.0062% for $^{208}\text{Pb}/^{204}\text{Pb}$) were used as external standards. Due to a systematic difference between measured and accepted standard ratios, Sr, Nd and Pb isotope ratios were further corrected for external fractionation by a value of -0.039 , $+0.047$ and $+0.5$ amu, respectively. Mass interferences at 84 (^{84}Kr), 86 (^{86}Kr) and 87 (^{87}Rb) were corrected for by monitoring ^{83}Kr and ^{85}Rb . The interference of ^{144}Sm on ^{144}Nd was monitored on ^{147}Sm and corrected with a value of 0.206700 ($^{144}\text{Sm}/^{147}\text{Sm}$). The interference of ^{204}Hg on ^{204}Pb was corrected by monitoring ^{202}Hg .

1.3. Zircon LA-ICP-MS U-Pb geochronology

The U–Pb isotopic composition of zircons was obtained using LA-ICP-MS at the University of Lausanne. Zircons were ablated with an UP-193FX ArF 193 nm excimer ablation system (ESI) using the following parameters: 35 μm beam size, 5 Hz repetition rate, 30 second signal and a beam energy density of 2.2–2.5 J/ cm^2 . Isotopic intensities were measured using an Element XR single-collector sector-field ICP-MS (Thermo Scientific). GEMOC GJ-1 zircon (CA-ID-TIMS ^{206}Pb – ^{238}U age of 600.5 ± 0.4 Ma; Boekhout et al., 2012; Ulianov et al., 2012) was used as the primary standard. Secondary standards used to monitor consistency in the measured U–Pb dates were either Harvard 91500 (1065.4 ± 0.3 Ma; Wiedenbeck et al., 1995) zircon, or Plešovice (337.13 ± 0.37 Ma; Sláma et al., 2008) zircon. Dates were calculated using LAMTRACE (Jackson, 2008) and IsoplotR (Vermeesch, 2018). More details regarding the mass spectrometer setup and data reduction can be found in Ulianov et al. (2012). Statistical analyses of magmatic zircon data were performed using Isoplot 3.71 (Ludwig, 2003). All discordant analyses of magmatic zircons were discarded and only zircons with concordance greater than 90% were accepted and plotted. All uncertainties are reported at $\pm 2\sigma$.

1.5. Zircon in-situ Hf isotopes

The same zircons that were used for dating were also selected for *in-situ* Hf isotope measurements. Analyses were carried out on a Thermo Neptune Plus MC-ICP-MS coupled to a Teledyne - Photon Machines Analyte G2 ArF excimer laser system equipped with a two volume HelEx-2 ablation cell (d'Abzac et al., 2014) at the University of Geneva. Ablation was performed at a fluence of $\sim 4 \text{ J/cm}^2$, a repetition rate of 5Hz and a spot size of 40 μm (50 μm in some rare cases where enough space was available on the zircon). Helium was used as a carrier gas for the ablated particles and mixed with a small amount of N_2 before entering the Ar-plasma torch to increase sensitivity. Measurements were performed at a low mass resolution over 120 cycles of $\sim 1\text{s}$ for standards and between 80 and 120 cycles for samples (depending on the depth of the zircons). At the beginning of the session, the end and every 15 sample measurements, Mud Tank, Plešovice, MUN4 and GJ-1 zircon standards were measured to evaluate the offset of the measured values to reference values. Blanks were also acquired (120 cycles) at the same intervals as the zircon standard measurements, but without ablation.

Data were reduced off-line using an Excel spreadsheet and consisted of blank subtractions, removing the isobaric interference of ^{176}Lu and ^{176}Yb on mass 176 (e.g. Fisher et al., 2011) and correcting the resulting $^{176}\text{Hf}/^{177}\text{Hf}$ ratio for mass bias using an exponential law (Albarede et al., 2004). β_{Hf} and β_{Yb} mass bias coefficients were calculated from the measured $^{179}\text{Hf}/^{177}\text{Hf}$ and $^{173}\text{Yb}/^{171}\text{Yb}$ with the reference values of Patchett and Tatsumoto (1981; $^{179}\text{Hf}/^{177}\text{Hf}=0.7325$) and Thirlwall and Anczkiewicz (2004; $^{173}\text{Yb}/^{171}\text{Yb}=1.1234$) respectively. Isobaric interferences of ^{176}Yb and ^{176}Lu with ^{176}Hf were corrected using $^{176}\text{Yb}/^{173}\text{Yb}=0.786954$ and $^{176}\text{Lu}/^{175}\text{Lu}=0.02645$ respectively (Thirlwall and Anczkiewicz, 2004). Only non-perturbed spectra were retained. Initial $^{176}\text{Hf}/^{177}\text{Hf}$ ratios and initial ϵ_{Hf} were calculated using the $^{206}\text{Pb}/^{238}\text{U}$ date of the respective crystal, the CHUR parameters of Bouvier et al. (2008); $^{176}\text{Hf}/^{177}\text{Hf} = 0.282785$ and $^{176}\text{Lu}/^{177}\text{Hf} = 0.0336$ and $\lambda^{176}\text{Lu}=1.87 \times 10^{-11} \text{ yr}^{-1}$ (Söderlund et al., 2004).

References

- Albarède, F., Telouk, P., Blichert-Toft, J., Boyet, M., Agraniér, A., Nelson, B., 2004. Precise and accurate isotopic measurements using multiple-collector ICPMS. *Geochim. Cosmochim. Acta* 68, 2725–2744.
- Baker, J., Peate, D., Waight, T., & Meyzen, C. 2004. Pb isotopic analysis of standards and samples using a ^{207}Pb – ^{204}Pb double spike and thallium to correct for mass bias with a double-focusing MC-ICP-MS. *Chemical Geology*, 211(3-4), 275-303.
- Béguelin, P., Chiaradia, M., Beate, B., & Spikings, R. (2015). The Yanaurcu volcano (Western Cordillera, Ecuador): A field, petrographic, geochemical, isotopic and geochronological study. *Lithos*, 218, 37-53.
- Boekhout, F., Spikings, R., Sempere, T., Chiaradia, M., Ulianov, A., & Schaltegger, U. (2012). Mesozoic arc magmatism along the southern Peruvian margin during Gondwana breakup and dispersal. *Lithos*, 146, 48-64.
- Bouvier, A., Vervoort, J.D., Patchett, P.J., 2008. The Lu–Hf and Sm–Nd isotopic composition of CHUR: Constraints from unequilibrated chondrites and implications for the bulk composition of terrestrial planets. *Earth and Planetary Science Letters* 273, 48–57.
- Chiaradia, M. (2015), Crustal thickness control on Sr/Y signatures of recent arc magmas: An Earth scale perspective, *Sci. Rep.*, 5, 8115, doi:10.1038/srep08115.
- Chiaradia, M., Müntener, O., & Beate, B. 2011. Enriched basaltic andesites from mid-crustal fractional crystallization, recharge, and assimilation (Pilavo Volcano, Western Cordillera of Ecuador). *Journal of Petrology*, 52(6), 1107-1141.
- D'Abzac, F.-X., Czaja, A.D., Beard, B., Schauer, J.J., Johnson, C.M., 2014. Iron distribution in size-resolved aerosols generated by UV-Femtosecond laser ablation: influence of cell geometry and implications for in situ isotopic determination by LA-MC-ICP-MS. *Geostandards and Geoanalytical Research* 38, 293-309.
- Fisher, C.M., Hanchar, J.M., Samson, S.D., Dhuime, B., Blichert-Toft, J., Vervoort, J.D., Lam, R., 2011. Synthetic zircon doped with hafnium and rare earth elements: a reference material for in situ hafnium isotope analysis. *Chem. Geol.* 286, 32–47.
- Jackson, S. E. 2008. LAMTRACE data reduction software for LA-ICP-MS. In *Laser ablation ICP-MS in the earth sciences: current practices and outstanding issues* (Vol. 40, pp. 305-307). Vancouver, BC, Canada: Mineralogical Association of Canada.
- Jackson, S. E., 2008. Lamtrace data reduction software for LA-ICP-MS. In: Sylvester, P. (ed.) *Laser Ablation ICP-MS in the Earth Sciences: Current Practices and Outstanding Issues*. Short Course Series, Vol. 40. Mineralogical Association of Canada, pp. 305–307.

- Ludwig, K.R., 2003, Isoplot/Ex version 3.00. A Geochronological Toolkit for Microsoft Excel, User's Manual: Berkeley Geochronology Center Special Publications, v. 4, p. 1–70.
- Patchett, P. J., Tatsumoto, M. 1980. A Routine high-precision method for Lu-Hf isotope geochemistry and chronology. *Contrib. Miner. Petrol.*, 75, pp. 263-267.
- Pin C. and Zalduegui J. F. S. (1997) Sequential separation of light rare-earth elements, thorium and uranium by miniaturized extraction chromatography: Application to isotopic analyses of silicate rocks. *Anal. Chim. Acta* 339, 79–89.
- Slama, J., Košler, J., Condon, D.J., Crowley, J.L., Gerdes, A., Hanchar, J.M., Horstwood, M.S.A., Morris, G.A., Nasdala, L., Norberg, N., Schaltegger, U., Schoene, B., Tubrett, M.N., Whitehouse, M.J., 2008. Plešovice zircon — a new natural reference material for U–Pb and Hf isotopic microanalysis. *Chemical Geology* 249 (1–2), 1–35
- Söderlund, U., Patchett, J., Vervoort, J., Isachsen, C., 2004. The ^{176}Lu decay constant determined by Lu–Hf and U–Pb isotope systematics of Precambrian mafic intrusions. *Earth Planet. Sci. Lett.* 219, 311–324.
- Tanaka, T., and 19 co-authors., 2000, JNdi-1: A neodymium isotopic reference in consistency with La Jolla neodymium: *Chemical Geology*, v. 168, p. 279–281.
- Thirlwall, M.F., Anczkiewicz, R., 2004. Multidynamic isotope ratio analysis using MC-ICP-MS and the causes of secular drift in Hf, Nd and Pb isotope ratios. *Int. J. Mass Spectrom.* 235, 59–81.
- Ulianov, A., Muntener, O., Schaltegger, U., Bussy, F., 2012. The data treatment dependant variability of U–Pb zircon ages obtained using mono-collector, sector field, laser ablation ICP-MS. *Journal of Analytical Atomic Spectrometry* 27, 663–676.
- Vermeesch, P., 2018, IsoplotR: A free and open toolbox for geochronology: *Geoscience Frontiers*, <https://doi.org/10.1016/j.gsf.2018.04.001>
- Wiedenbeck, M., Alle, P., Corfu, F., Griffin, W.L., Meier, M., Oberli, F., von Quadt, A., Roddick, J.C., Spiegel, W., 1995. 3 natural zircon standards for U-Th-Pb, Lu-Hf, trace-element and REE analyses. *Geostandards Newsletter* 19, 1–23.