

Analytical techniques

$^{40}\text{Ar}/^{39}\text{Ar}$ dating was carried out at Argonlabor Freiberg (ALF), TU Freiberg, Germany. Amphibole and biotite separates were wrapped in Al-foil and placed along with fluence monitors in holes on ultrapure Al discs for irradiation, which was done at the FRG2 research reactor of the Helmholtz-Zentrum Geesthacht, Germany, under Cd-shielding for 72 h at a thermal neutron fluence of $\sim 5 \times 10^{13}$ n/cm²s and a thermal to fast neutron ratio of ~ 40 . Irradiated samples were unwrapped and stepwise heating of amphibole separates (~ 10 to ~ 50 mg) was performed using a Createc high-temperature furnace system (a modified version of this system is described in Pfänder et al. 2014). Heating time was 7 min per step. Stepwise heating of biotite separates (~ 3 mg) was done using a power controlled New Wave floating CO₂ laser system with a beam diameter of 3 mm at a heating time of 5 min per step. Gas purification was performed by two AP10N getter pumps, one at room temperature and one at $\sim 400^\circ\text{C}$. Argon isotope abundances were measured in static mode on a GV Instruments ARGUS noble gas-mass spectrometer equipped with five faraday cups and $10^{12} \Omega$ resistors on mass positions 36 to 39 and a $10^{11} \Omega$ resistor on mass position 40. Each measurement comprises 45 scans at 10 sec integration time each. A more detailed description of the performance of the system as well as typical blank levels are given in Pfänder et al. (2014). Mass bias was corrected by assuming linear mass-dependent isotope fractionation and by using an atmospheric $^{40}\text{Ar}/^{36}\text{Ar}$ ratio of 298.6 ± 0.3 (Lee et al. 2006). Raw-data reduction and time-zero intercept calculations were done using an in-house developed Matlab software package. Isochron, inverse isochron and plateau ages have been calculated with the Excel Add-In ISOPLLOT 3.7 (Ludwig 2008). Fish Canyon Tuff sanidine was used as flux monitor (28.305 ± 0.036 Ma; Renne et al. 2010) and the decay constants of Renne et al. (2010) served as base for all calculations. J-values and interference correction factors along with temperature and power steps, raw data and intensity intercepts are given as supplementary data (Table A1).

Minerals were analysed with a JEOL microprobe at the University of Münster, Germany. Operating conditions were 15 kV and 5 nA, with a peak counting time of 5 s and a background counting time of 3 s with a beam size of 5–20 μm . Background was quantified by measuring the intensity on low- and high angle side of the peak for all quantitative analyses. A set of well characterized natural and synthetic silicate standards have been used for calibration of the microprobe. The ZAF correction procedure was applied to the data; errors in the major oxides are estimated to be about 1–2 %. Representative mineral data are given in Table A2.

Whole rock powders were prepared from rock chips free from surface alteration using a jaw crusher, a ball mill and an agate mortar. Major and some trace elements (except for rare earth elements (REE) and Nb, Ta, Hf, Th and U) were determined on fused lithium-tetraborate glass beads using standard XRF techniques on a fully automated WDS X-ray fluorescence spectrometer equipped with a Rh-tube (Panalytical Magix Pro, Vogel and Kuipers, 1987). Loss on ignition

(LOI) was determined gravimetrically after heating the samples at 1050 °C for one hour (Lechler and Desilets 1987). FeO was measured titrimetrically with standard techniques using potassium permanganate (Heinrichs and Hermann. 1990). Accuracy and reproducibility were controlled by repeated measurements against several international and in-house standards and data for certified reference materials are given in Table A3.

For some samples, trace element concentrations of Nb, Hf, Ta, Th, and U were determined by isotope dilution inductively coupled mass spectrometry (ID-ICPMS) using a ThermoElectron ELEMENT 2 high resolution ICPMS at Max-Planck Institut für Chemie, Mainz. About 100 mg of rock powder are mixed with a multi-element tracer solution (containing enriched isotopes of ^{91}Zr , ^{179}Hf , and ^{235}U). After dissolution, using a standard ICPMS sample preparation procedure, an aliquot of the solution is diluted by a factor of 20000 and measured by sector field ICP-MS. The element concentrations of Zr, Hf, and U are determined by isotope dilution (ID) whereas all other concentrations are obtained by calibration against a multi-element standard solution using Zr, Hf, and U as internal standards. A detailed description of the method can be found in Willbold and Jochum (2005). The combined standard uncertainty has been estimated at 2% (1 RSD) for ID determined elements and 4% (1 RSD) for all other elements (Willbold et al. 2003). Three geological reference materials (JA-1, JA-2, JA-3) were measured together with unknown samples for quality assurance. Given the uncertainty of concentrations as obtained by ID-ICPMS and the uncertainty of reference materials our results are in good agreement with the published values. Data for certified reference materials are given in Table A3.

For the Rb-Sr, Sm-Nd and Pb whole rock isotope analyses, mm-sized rock chips were leached for 1 hour in 6 N HCl on a hot plate. After leaching, the leachate was decanted and the samples were carefully rinsed with Milli-Q water. Subsequently, they were spiked with ^{149}Sm - ^{150}Nd and ^{85}Rb - ^{84}Sr enriched isotope tracers and digested in concentrated HF-HNO₃ in 3 ml screw-top Teflon vials inside Parr metal bombs at 200°C for three days. After complete dissolution the samples were dried and redissolved in 2.5 N HCl. Rubidium, Sr and REE were separated using standard cation exchange columns with a DOWEX AG 50 W-X 12 resin, using 2.5 N HCl for Rb and Sr and 6 N HCl for the REE. Neodymium and Sm were separated from the other REE on HDEHP coated Teflon columns, using 0.12 N HCl for Nd and 0.3 N HCl for Sm. Isotope analyses were carried out at the Max-Planck-Institut für Chemie at Mainz with a Finnigan MAT 261 multicollector thermal ionization mass spectrometer operating in static mode. Rubidium, Sm and Nd were run on Re double filaments and Sr was run on W single filaments loaded with TaF₅. Neodymium isotopes were normalized to $^{146}\text{Nd}/^{144}\text{Nd}=0.7219$. The total procedural blank for Nd was < 30 pg and is negligible. Repeated measurements of the La Jolla Nd standard gave $^{143}\text{Nd}/^{144}\text{Nd}=0.511857 \pm 0.000026$ (2 s; n=24). The repeatability of the Sr standard (NBS 987) was $^{87}\text{Sr}/^{86}\text{Sr}=0.710230 \pm 0.000021$ (2 s; n=24) and the fractionation was corrected to $^{86}\text{Sr}/^{88}\text{Sr}=0.1194$. Within-run precisions in the $^{87}\text{Sr}/^{86}\text{Sr}$ and

$^{143}\text{Nd}/^{144}\text{Nd}$ ratios are reported in the last two digits. Typical analytical errors in the $^{87}\text{Rb}/^{86}\text{Sr}$ and $^{147}\text{Sm}/^{144}\text{Nd}$ ratios are equal or better than 0.5 % and 0.2%, respectively. The total procedural blank for Sr was < 150 pg and is negligible. For common Pb analyses, the samples were dissolved in 0.6 N HBr and loaded on Teflon columns filled with DOWEX AG 1 X 8 anion exchange resin (100-200 mesh) in chloride form (Mattinson 1986). The Pb was extracted using conventional HBr/HCl techniques and was loaded on Re single filaments using the H_3PO_4 -silica gel method (Cameron et al. 1969). Lead analyses were corrected for mass fractionation by a factor of 0.11 % per amu. The repeatability of the standard NBS 982 was 0.068%, 0.064% and 0.071% for the $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ ratio, respectively. The total procedure blank was < 80 pg Pb and is therefore negligible. Data for certified reference materials are given in Table A4.

For Lu-Hf isotope analyses, approximately 100 mg of whole rock powder were doped with a ^{176}Lu - ^{178}Hf isotope tracer. Whole rocks digested in concentrated HF-HNO₃ in 3 ml screw-top Teflon vials inside Parr metal bombs at 200°C for three days. After evaporation to dryness, each sample was treated three times with concentrated nitric acid and traces of HF. In a final step, samples were taken up in 6 M HCl to convert them into a chlorite form, and for each sample a clear solution was achieved. The aliquot fractions were dried down and converted to nitrate once to ensure complete spike sample equilibrium. Separation of Lu and Hf from the rock followed the protocol of Münker et al. (2001). Procedural blanks for Lu and Hf were 5 and 10 pg, respectively. Isotope analyses of Lu and Hf were carried out on a Thermo Fisher® Neptune MC-ICP-MS in Amsterdam using a Cetac® Aridus desolvating nebulizer system in static mode. Mass bias corrections for Hf was performed with $^{179}\text{Hf}/^{177}\text{Hf} = 0.7325$ using the exponential law (Russel et al., 1978). Interferences of ^{176}Yb and ^{176}Lu on ^{176}Hf were corrected with $^{176}\text{Lu}/^{175}\text{Lu} = 0.02655$ and $^{176}\text{Yb}/^{173}\text{Yb} = 0.79631$ (Vervoort et al. 2004). The $^{176}\text{Hf}/^{177}\text{Hf}$ for the JMC-475 standard solution during the course of the study was 0.282163 ± 0.000008 (2 s.d. of the mean). Lutetium isotopes were analyzed using natural sample Yb (Blichert-Toft et al. 1997) and additionally admixed Re (e.g., Scherer et al., 2001). Isotope ratios obtained by the double normalization methods always agreed within 0.02% for intensities on ^{173}Yb of at least 10 mV. Repeated analyses of standard solution with different Lu/Yb ratios of up to 1 showed no offset of the natural Lu isotope compositions outside the analytical uncertainty of 0.05%. For mass bias correction a $^{171}\text{Yb}/^{173}\text{Yb} = 0.883129$ (Vervoort et al., 2004) and $^{187}\text{Re}/^{185}\text{Re} = 1.6738$ (Gramlich et al. 1973) was used. Repeated analyses of spiked Lu-Hf mixed gravimetric standard solution yielded an error on Lu/Hf of 0.5%, which was applied to all samples in the study. Data for certified reference materials are given in Table A4.

Oxygen isotope analyses for samples CS 42, CS 56, CS 66, CS 88, CS 22, CS 39, CS 53, CS 59 and CS 89 were performed at the University of Bonn on ~ 10 mg aliquots of powdered whole-rock samples, using purified fluorine for oxygen extraction, followed by conversion to CO₂ (Clayton and Mayeda 1963). The $^{18}\text{O}/^{16}\text{O}$ measurements were made on a

SIRA-9 triple-collector mass spectrometer manufactured by VG-Isogas. Analytical uncertainties are $< 0.2\%$. All samples were measured twice and the average of these measurements is given in Table 2. All values are reported as per mil deviations relative to SMOW (Standard Mean Ocean Water) and refer to a certified value of $9.6 \pm 0.2\%$ for NBS 28. Oxygen isotope analyses for samples CS 69, CS 32, CS 54, CS 57, CS 91 and CS 97 were analyzed at Universität Göttingen using infrared (IR) laser fluorination in combination with gas chromatography isotope ratio monitoring gas mass spectrometry (GC-irmMS) (Jones et al. 1999; Sharp 1990). The gas extraction technique is described in Gehler et al. (2012). In brief, 1.0 – 1.3 mg of sample material was loaded along with MORB glass and NBS-28 quartz into a 18-pit nickel sample holder. After evacuation overnight and pre-fluorination, samples were reacted in a ~ 20 mbar atmosphere of purified F_2 gas (Asprey 1976) by means of heating with a SYNRAD 50 W CO_2 -laser. Liberated O_2 was cleaned from excess F_2 by reacting with NaCl ($110^\circ C$). The purified sample O_2 was expanded into a stainless-steel capillary and transported with He carrier gas through a second trap, where O_2 was cryofocused at $-196^\circ C$ on a molecular sieve. Sample O_2 was then released at $92^\circ C$ back into the He carrier gas stream and transported through a 5 Å molecular sieve GC column of a Thermo Gasbench-II. Sample O_2 was injected via an open split valve of the GasBench-II into the source of a THERMO MAT253 gas mass spectrometer. The signals of $^{16}O^{16}O$ and $^{18}O^{16}O$ were simultaneously monitored on Faraday cups. Sample peaks ($m/z = 32$) had an amplitude of 20 – 30 V and a full width at half maximum of ~ 20 s. Reference O_2 was injected before the sample through a second open split valve of the GasBench-II. The external error of a single analysis was $\pm 0.25\%$.

Tab.A2. Major element composition for selected amphiboles and plagioclases from three andesites covering nearly the full spectrum of MgO content. X Mg= Mg/(Mg+Fe)

Sample	CS 22		CS 66		CS 53				
Amphibole									
	rim	core	rim	core	rim	core			
SiO ₂	46.12	46.43	45.97	45.96	46.44	45.51			
TiO ₂	1.50	1.63	2.08	1.95	1.31	1.48			
Al ₂ O ₃	8.06	8.32	8.44	8.55	7.89	8.29			
FeO	14.22	14.08	13.52	13.20	15.71	15.75			
MnO	0.53	0.53	0.13	0.46	0.43	0.37			
MgO	13.86	13.30	13.92	13.60	12.30	12.38			
CaO	11.62	11.81	11.58	11.50	11.85	11.88			
Na ₂ O	1.29	1.35	1.57	1.41	1.22	1.30			
K ₂ O	0.74	0.81	0.62	0.66	0.91	0.90			
H ₂ O	1.99	1.99	1.99	1.99	1.99	1.99			
total	99.93	100.2	99.82	99.28	100.0	99.84			
X Mg	0.64	0.63	0.65	0.65	0.58	0.58			
Sample	CS 22		CS 66		CS 53				
Plagioclase									
SiO ₂	57.06	59.88	63.53	63.78	59.71	57.71	66.15	65.19	66.42
Al ₂ O ₃	27.35	24.99	22.94	23.13	25.66	26.57	21.96	23.33	22.24
CaO	10.14	7.64	5.87	5.33	7.24	9.53	3.49	3.09	3.08
Na ₂ O	5.88	7.24	8.00	8.53	7.00	5.93	9.54	7.31	8.42
K ₂ O	0.12	0.20	0.24	0.13	0.61	0.15	0.05	1.07	0.53
total	100.5	99.9	100.6	100.9	100.2	99.9	101.2	100.0	100.7
Ab	51	62	70	74	61	53	83	75	80
An	48	36	28	25	35	47	17	18	16
Or	1	1	1	1	4	1	0	7	3

Table A3: major and trace element data for certified reference materials used in this study and recommended values.
Recommended major and trace element concentrations for JA-1, JA-2, JA-3, BIR-1 and W-2 are from Govindaraju (1994).
Recommended trace element concentrations for SY-4 are from Bowman (1995).

	JA-1	JA-1 rec.	JA-2	JA-2 rec.	JA-3	JA-3 rec.	BIR-1a	BIR-1a rec.	W-2	W-2 rec.	SY-4	SY-4 rec.
SiO ₂					61.84	62.26						
TiO ₂					0.65	0.68						
Al ₂ O ₃					15.69	15.57						
Fe ₂ O ₃					6.56	6.59						
MnO					0.10	0.106						
MgO					3.59	3.65						
CaO					6.42	6.28						
Na ₂ O					3.17	3.17						
K ₂ O					1.34	1.41						
P ₂ O ₅					0.11	0.11						
Nb	1.51	1.7	10.0	9.8	3.73	3.0						
Hf	2.34	2.41	2.53	2.89	2.98	3.43						
Ta	0.105	0.10	0.518	0.61	0.201	0.14						
Th	0.746	0.82	4.538	4.7	3.289	3.4						
U	0.35	0.34	2.08	2.4	0.95	1.4						
V							314	310	271	262		
Cr							370	370				
Co							48	52	41	43		
Ni							180	170				
Cu									120	110		
Zn							70	70	80	80	100	93
Ga							15	16	17	17	35	35
Rb									19	21	54	55
Sr							101	110	188	190	1120	1191
Y							14.6	16			118	119
Zr									95	94		
Nb							0.5	0.6	7.7	7.9	14.2	13
Cs									0.8	0.99	1.5	1.5
Ba									167	182	337	340
La							0.6	0.63	11.2	10	63.3	58
Ce							1.9	1.9	23.8	23	129	122
Pr											14.9	15
Nd							2.3	2.5	12.7	13	58.6	57
Sm							1	1.1	3.3	3.3	13.1	12.7
Eu							0.53	0.55	1.1	1	1.93	2
Gd											13.6	14
Tb									0.59	0.63	2.57	2.6
Dy									3.8	3.6	18.9	18.2
Ho									0.75	0.76	4.31	4.3
Er											14.3	14.2
Tm											2.2	2.3
Yb							1.6	1.7	2	2.1	14.8	14.8
Lu									0.32	0.33	2.17	2.1
Hf									2.3	2.6	10.3	10.6
Ta									0.49	0.5	0.82	0.9
Pb							< 5	3			10	10
Th									2.3	2.4	1.2	1.4
U									0.51	0.53	0.8	0.8

Bowman, W.S. (1995). Canadian diorite gneiss SY-4: Preparation and Certification by eight-nine international laboratories. Geostandard Newsletter 19, 101-124.
Govindaraju, K. (Ed.) (1994). 1994 compilation of working values and sample description for 383 geostandards. Special Volume Geostandards Newsletter 18, 1-158.

Table A4: Isotope data for certified reference materials used in this study and recommended values.

	BCR-2	±2.s.e.	BCR-2(1)	±2.s.e.	BCR-2(2)	±2.s.e.	BCR-2(4)	±2.s.e.
	(this study)							
$^{87}\text{Sr}/^{86}\text{Sr}$	0.704965	0.000012	0.704958	0.000008	0.705013	0.000010	0.705000	0.000011
$^{143}\text{Nd}/^{144}\text{Nd}$	0.512637	0.000012	0.512633	0.000007	0.512637	0.000012	0.512637	0.000013
	BCR-2(4)	±2.s.e.	BCR-2	±2.s.e.	BCR-2(2)	±2.s.e.		
			(this study)					
$^{206}\text{Pb}/^{204}\text{Pb}$	18.8029	0.000010	18.752	0.010	18.7259	0.0197		
$^{207}\text{Pb}/^{204}\text{Pb}$	15.6239	0.000008	15.620	0.022	15.6429	0.0040		
$^{208}\text{Pb}/^{204}\text{Pb}$	38.8287	0.000025	38.695	0.031	38.7237	0.0405		
	BCR-2	±2.s.e.	BCR-2(3)	±2.s.e.	BCR-2(4)	±2.s.e.		
	(this study)							
$^{176}\text{Hf}/^{177}\text{Hf}$	0.282890	0.000003	0.282870	0.000008	0.282866	0.000011		

(1) Razcek et al. 2007

(2) Weis et al. 2006

(3) Weis et al. 2007

(4) Jweda et al. (2015)

Razcek, I., Jochum, K.P., Hofmann, A.W. 2007. Neodymium and Strontium isotope data for USGS Reference materials BCR-1. BCR-2. BHVO-1. BHVO-2. AGV-1. AGV-2. GSP-1. GSP-2 and eight MPI-DING reference glasses. *Geostandards Newsletters* 27: 173-179.

Weis, D., Kiefer, B., Maerschalk, C., Barling, J., de Jong, J., Williams, G.A., Hanano, D., Pretorius, W., Matielli, N., Scoates, J.S., Goolaerts, A., Friedman, R. M., Mahoney, J.B, 2006. High-precision isotopic characterization of USGS reference materials by TIMS and MC-ICP-MS. *Geochemistry Geophysics Geosystems* 7:Q08006.

Weis, D., Kiefer, B., Hanano, D., Silva, I.N., Barling, J., Pretorius, W., Maerschalk, C., Matielli, N. 2007. Hf isotopic characterization of U.S. Geological Survey reference materials. *Geochemistry Geophysics Geosystems* 7:Q06006.

Jweda, J., Bolge, L., Class, C., Goldstein, S.L., 2015. High-precision Sr-Nd-Hf-Pb isotopic composition of USGS reference material BCR-2. *Geostandards and Geoanalytical Research* 40, 101-115.

Table A5: Composition and parameters used for the EC-AFC model calculations (Spera and Bohrsen, 2001).

Sample VS18 is from Voshage et al. (1990) and sample GL11 is from Pinarelli et al. (2002).

Magma liquidus temperature t_{lm}	1250°C			
Magma temperature t_{m0}	1250°C			
Assimilant liquidus temperature	850°C			
Country rock temperature t_{a0}	850°C			
Solidus temperature	800°C			
magma specific heat capacity C_{pm}	1484 Jkg ⁻¹ K ⁻¹			
Assimilant specific heat capacity C_{pa}	1388 Jkg ⁻¹ K ⁻¹			
Crystallization enthalpy	396000 Jkg ⁻¹			
Fusion enthalpy	354000 Jkg ⁻¹			
Equilibration temperature	800°C			
Element	Sr	Nd	Sr	Nd
Melt (sample CS 56)	250	15	250	15
Bulk Do	1	0.8	1	0.8
Assimilant (VS18)	207	33		
Assimilant (GL11)			293	52
Bulk Do	1	0.8	1	0.8
	⁸⁷ Sr/ ⁸⁶ Sr	¹⁴³ Nd/ ¹⁴⁴ Nd	⁸⁷ Sr/ ⁸⁶ Sr	¹⁴³ Nd/ ¹⁴⁴ Nd
Melt	0.708500	0.512500	0.70800	0.512500
Assimilant (VS 18)	0.721000	0.512070		
Assimilant (GL11)			0.71700	0.512050

Fig. A1: Elemental composition of amphibole from samples CS66, CS22 and CS53

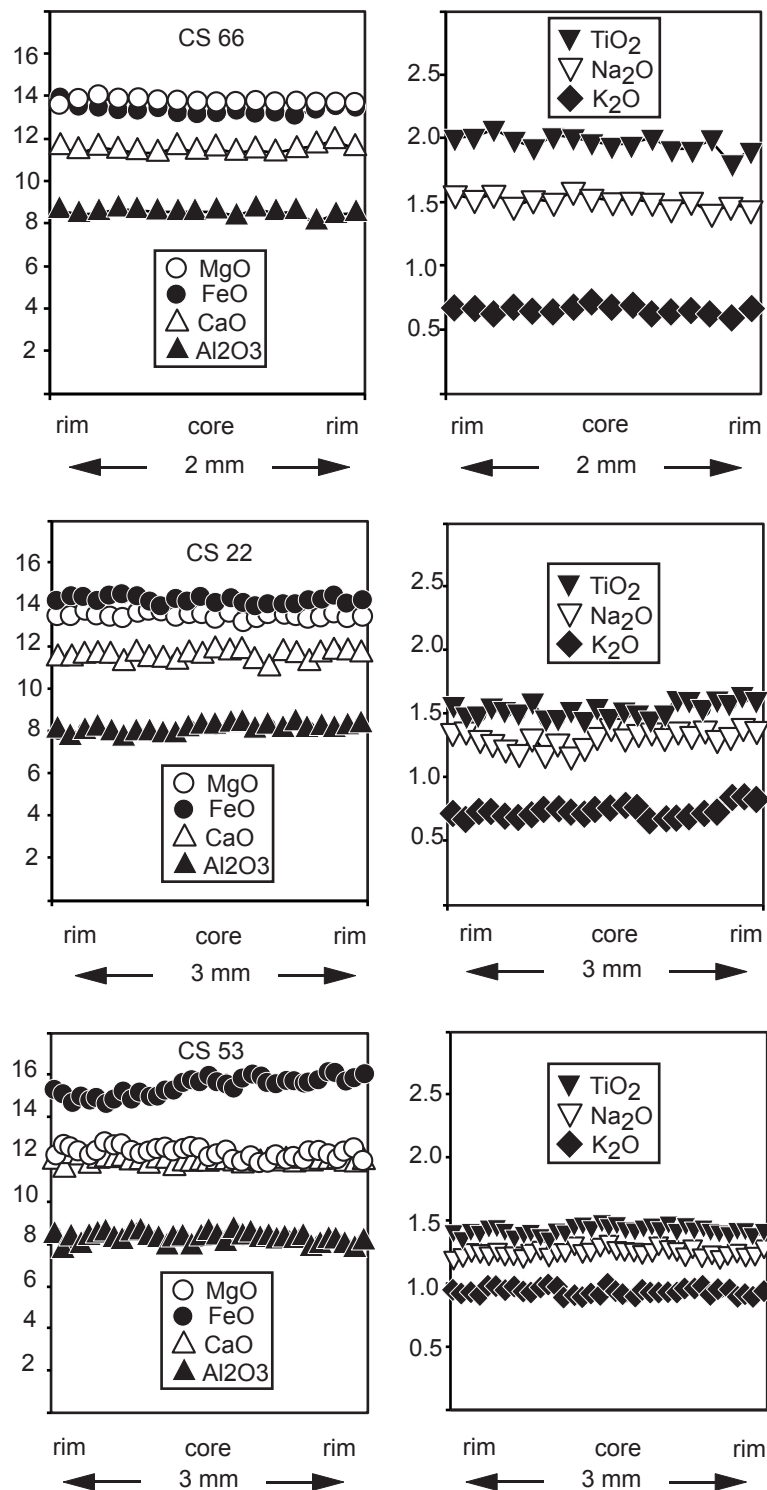
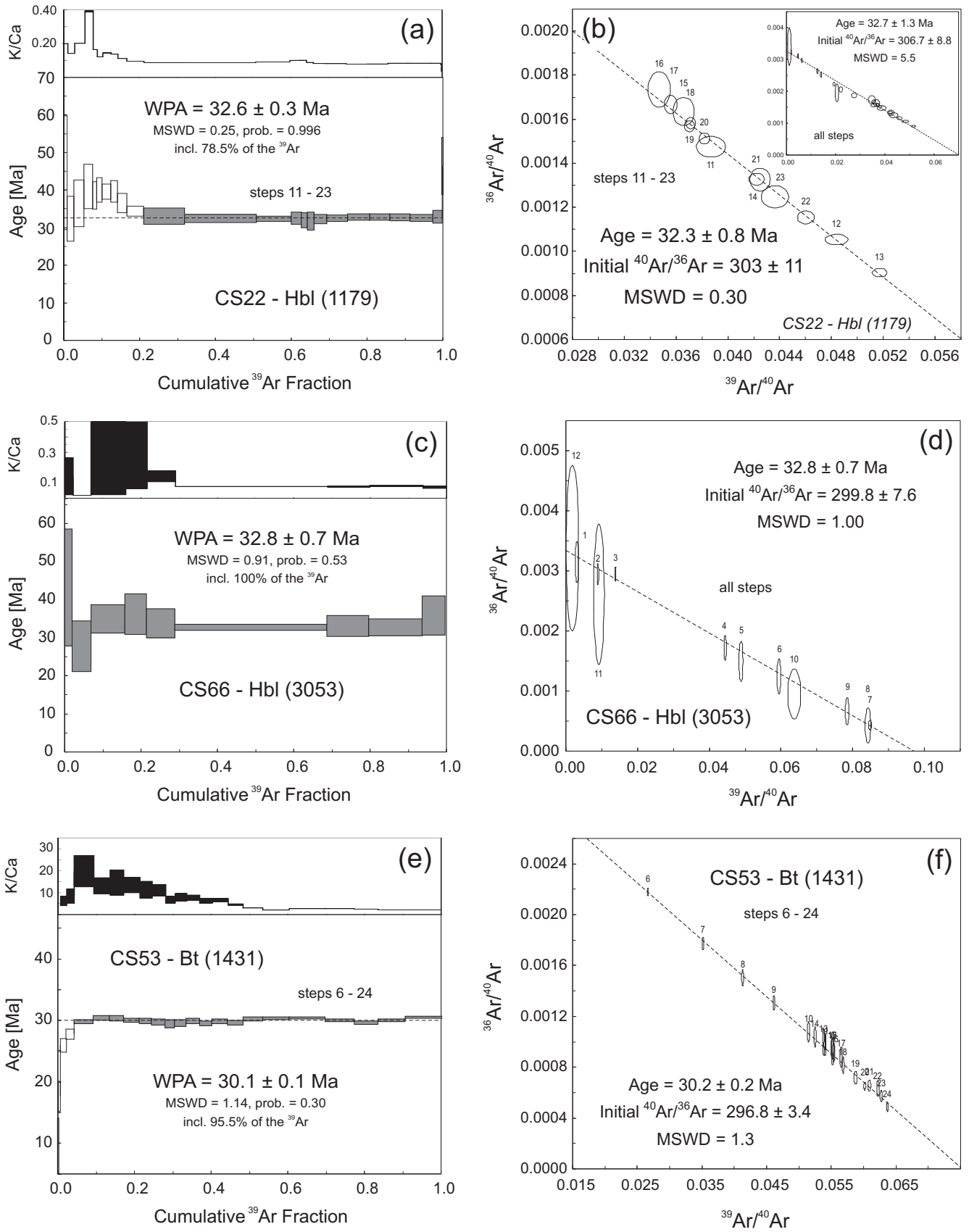


Fig. A2: Ar-Ar age spectra and inverse isochron plots



Age spectra and inverse isochron plots of two amphibole (a-d) and one biotite (e - f) separate. Temperature steps used to calculate the plateau ages are dark grey, those omitted are white. In the inverse isochron diagrams, only plateau steps have been used to calculate the inverse isochron age and the initial $^{40}\text{Ar}/^{36}\text{Ar}$ composition (black symbols and labels). All errors are reported on a 2σ confidence level.