

Supplementary Table S1. Operation parameters for fsLA-MIC-ICPMS

Laser ablation	
Type of laser	femto-second
Pulse width	<300 fs
Wavelength	200 nm
Laser fluence	6 J cm ⁻²
Ablation mode	Rotation raster
Spot size	30 μm
Repetition rate	1-10 Hz
Ablation cell	~20 cm ³ (in-house made)
Aerosol carrier gas (He)	1.2 L min ⁻¹
Mass spectrometer	
MC-ICPMS	Thermo Fisher Scientific, Neptune (modified)
RF power	1200 W (27.12 MHz)
Guard electrode	On
Plasma gas (Ar)	15 L min ⁻¹
Auxiliary gas (Ar)	1 L min ⁻¹
Make-up gas (Ar)	1 – 1.3 L min ⁻¹ (mixed with carrier He in 150 ml chamber)
Sample cone	JET-cone (Ni), Normal (N)-cone (Ni) was also used for comparison
Skimmer cone	X-cone (Ni)
Mass resolution	400 (low resolution)
Collector assignment	IC array Center mass: 221.2 IC1: 204(Pb+Hg) IC2: 206(Pb) IC3: 207(Pb) IC4: 208(Pb)

Supplementary Table S2. Pb isotope ratios of BHVO-2G (Pb 1.7 ppm) measured by 200-nm fsLA-MIC-ICP-MS using 30- μ m crater and 10 Hz laser repetition rate

Sample	Measurement data	Bracketing standard	ICP interface	Isotope ratio		Meas. error (2 SE*)	
				$^{207}\text{Pb}/^{206}\text{Pb}$	$^{208}\text{Pb}/^{206}\text{Pb}$	$^{207}\text{Pb}/^{206}\text{Pb}$	$^{208}\text{Pb}/^{206}\text{Pb}$
BHVO-2G_1	Dec. 14, 2011	BCR-2G	N-X	0.836	2.067	0.008	0.015
BHVO-2G_2		BCR-2G	N-X	0.841	2.057	0.008	0.016
BHVO-2G_3		BCR-2G	N-X	0.831	2.049	0.008	0.016
BHVO-2G_4		BCR-2G	N-X	0.837	2.052	0.008	0.016
BHVO-2G_5		BCR-2G	N-X	0.831	2.044	0.008	0.015
BHVO-2G_3	Apr. 26, 2012	GSD-1G	N-X	0.827	2.057	0.012	0.022
BHVO-2G_4		GSD-1G	N-X	0.829	2.047	0.011	0.022
BHVO-2G_5		GSD-1G	N-X	0.823	2.048	0.011	0.021
BHVO-2G_1	Apr. 27, 2012	BCR-2G	N-X	0.833	2.060	0.006	0.012
BHVO-2G_2		BCR-2G	N-X	0.832	2.047	0.006	0.013
BHVO-2G_3		BCR-2G	N-X	0.834	2.058	0.006	0.013
BHVO-2G_4		BCR-2G	N-X	0.830	2.050	0.007	0.013
BHVO-2G_5		BCR-2G	N-X	0.835	2.057	0.006	0.012
BHVO-2G_1	Apr. 27, 2012	BCR-2G	N-X	0.824	2.047	0.007	0.014
BHVO-2G_2		BCR-2G	N-X	0.826	2.059	0.007	0.014
BHVO-2G_3		BCR-2G	N-X	0.830	2.038	0.007	0.014
BHVO-2G_4		BCR-2G	N-X	0.830	2.056	0.007	0.013
BHVO-2G_5		BCR-2G	N-X	0.827	2.038	0.007	0.013
BHVO-2G_1	May. 2, 2012	BCR-2G	N-X	0.831	2.044	0.006	0.012
BHVO-2G_2		BCR-2G	N-X	0.836	2.058	0.006	0.013
BHVO-2G_3		BCR-2G	N-X	0.832	2.050	0.006	0.012
BHVO-2G_4		BCR-2G	N-X	0.836	2.060	0.006	0.012
BHVO-2G_5		BCR-2G	N-X	0.830	2.051	0.006	0.014
BHVO-2G_1	May. 7, 2012	BCR-2G	Jet-X	0.835	2.056	0.005	0.009
BHVO-2G_2		BCR-2G	Jet-X	0.833	2.053	0.004	0.009
BHVO-2G_3		BCR-2G	Jet-X	0.833	2.057	0.005	0.010
BHVO-2G_4		BCR-2G	Jet-X	0.839	2.059	0.005	0.010
BHVO-2G_5		BCR-2G	Jet-X	0.834	2.056	0.005	0.010
BHVO-2G_1	May. 9, 2012	BCR-2G	Jet-X	0.836	2.052	0.006	0.011
BHVO-2G_2		BCR-2G	Jet-X	0.836	2.051	0.005	0.012
BHVO-2G_3		BCR-2G	Jet-X	0.836	2.048	0.006	0.010
BHVO-2G_4		BCR-2G	Jet-X	0.835	2.048	0.006	0.011
BHVO-2G_5		BCR-2G	Jet-X	0.835	2.050	0.005	0.011
BHVO-2G_1	May. 16, 2012	BCR-2G	Jet-X	0.839	2.043	0.007	0.014
BHVO-2G_2		BCR-2G	Jet-X	0.837	2.054	0.007	0.013
BHVO-2G_3		BCR-2G	Jet-X	0.840	2.060	0.007	0.014
BHVO-2G_4		BCR-2G	Jet-X	0.840	2.066	0.007	0.015
BHVO-2G_3	Jun. 26, 2012	BCR-2G	Jet-X	0.833	2.061	0.005	0.012
BHVO-2G_5		BCR-2G	Jet-X	0.834	2.058	0.005	0.011
BHVO-2G_7		BCR-2G	Jet-X	0.832	2.022	0.005	0.011
BHVO-2G_8		BCR-2G	Jet-X	0.831	2.045	0.005	0.012
BHVO-2G_10		BCR-2G	Jet-X	0.829	2.039	0.005	0.011
BHVO-2G_1	Jun. 26, 2012	NIST 612	Jet-X	0.835	2.026	0.005	0.010
BHVO-2G_4		NIST 612	Jet-X	0.831	2.052	0.005	0.010
BHVO-2G_1	Jul. 2, 2012	NIST 612	Jet-X	0.838	2.045	0.007	0.016
BHVO-2G_4		NIST 612	Jet-X	0.831	2.042	0.007	0.015
BHVO-2G_6		NIST 612	Jet-X	0.841	2.045	0.007	0.015
BHVO-2G_1	Jan. 15, 2013	BCR-2G	Jet-X	0.834	2.054	0.005	0.009
BHVO-2G_2		BCR-2G	Jet-X	0.836	2.059	0.005	0.009
BHVO-2G_3		BCR-2G	Jet-X	0.839	2.053	0.005	0.010

BHVO-2G_4		BCR-2G	Jet-X	0.837	2.053	0.004	0.010
BHVO-2G_5		BCR-2G	Jet-X	0.834	2.046	0.005	0.010
BHVO-2G_1	Jan. 15, 2013	NIST 612	Jet-X	0.838	2.064	0.005	0.009
BHVO-2G_2		NIST 612	Jet-X	0.831	2.061	0.004	0.009
BHVO-2G_3		NIST 612	Jet-X	0.834	2.065	0.005	0.009
BHVO-2G_4		NIST 612	Jet-X	0.838	2.062	0.004	0.009
BHVO-2G_5		NIST 612	Jet-X	0.830	2.051	0.004	0.009
BHVO-2G_1	Feb. 22, 2013	NIST 612	Jet-X	0.834	2.065	0.004	0.009
BHVO-2G_2		NIST 612	Jet-X	0.839	2.066	0.004	0.008
BHVO-2G_3		NIST 612	Jet-X	0.834	2.064	0.004	0.008
BHVO-2G_4		NIST 612	Jet-X	0.834	2.066	0.004	0.008
Mean (n = 61)				0.834	2.053		
2 SD				0.008	0.018		
2 SD, %				0.96	0.89		
Accuracy**, %				0.19 (0.07)	0.24 (0.13)		
Ref. value ^a				0.832 ± 0.001	2.048 ± 0.003		
LA-ICP-MS value ^b				0.833 ± 0.002	2.050 ± 0.002		

*Bracketing standards error propagated.

**Compared with solution MC-ICP-MS data (Elburg et al., 2005), and LA-SF-ICP-MS data in brackets (Jochum et al., 2005c).

^a Reference values taken from Elburg et al. (2005), errors are +/- 2SD.

^b Reference values taken from Jochum et al. (2005c), errors are +/- 1SE.

Supplementary Table S3. Pb isotope ratio analyses of geological reference glasses from MPI-DING and USGS measured by 200-nm fsLA-MIC-ICP-MS

Sample	Bracketing ICP		Laser repetition	Isotope ratio		2SE	
	standard	interface		$^{207}\text{Pb}/^{206}\text{Pb}$	$^{208}\text{Pb}/^{206}\text{Pb}$	$^{207}\text{Pb}/^{206}\text{Pb}$	$^{208}\text{Pb}/^{206}\text{Pb}$
KL2-G (MPI-DING basalt, Pb 2.07 ppm)							
KL2-G_1	BCR-2G	JET-X	10 Hz	0.833	2.045	0.004	0.009
KL2-G_2	BCR-2G	JET-X	10 Hz	0.832	2.048	0.004	0.008
KL2-G_3	BCR-2G	JET-X	10 Hz	0.828	2.038	0.004	0.008
KL2-G_4	BCR-2G	JET-X	10 Hz	0.831	2.046	0.005	0.008
KL2-G_5	BCR-2G	JET-X	10 Hz	0.829	2.042	0.005	0.008
KL2-G_1	BCR-2G	JET-X	10 Hz	0.831	2.049	0.005	0.010
KL2-G_2	BCR-2G	JET-X	10 Hz	0.839	2.057	0.005	0.010
KL2-G_3	BCR-2G	JET-X	10 Hz	0.835	2.036	0.005	0.010
KL2-G_4	BCR-2G	JET-X	10 Hz	0.830	2.037	0.005	0.010
KL2-G_5	BCR-2G	JET-X	10 Hz	0.829	2.039	0.005	0.011
KL2-G_1	NIST612	JET-X	10 Hz	0.825	2.036	0.005	0.011
KL2-G_2	NIST612	JET-X	10 Hz	0.818	2.025	0.005	0.011
KL2-G_3	NIST612	JET-X	10 Hz	0.826	2.028	0.005	0.011
KL2-G_4	NIST612	JET-X	10 Hz	0.819	2.021	0.005	0.010
KL2-G_5	NIST612	JET-X	10 Hz	0.826	2.023	0.005	0.011
Mean				0.829	2.038		
2 SD				0.011	0.021		
2 SD, %				1.34	1.02		
Accuracy, %				0.95	0.73		
Reference value (a)				0.821	2.023		
(b)				0.821	2.024		
StHs6/80-G (MPI-DING andesite, Pb 10.3 ppm)							
StHs6/80-G_1	NIST612	JET-X	5 Hz	0.834	2.050	0.004	0.008
StHs6/80-G_2	NIST612	JET-X	5 Hz	0.826	2.043	0.004	0.007
StHs6/80-G_3	NIST612	JET-X	5 Hz	0.823	2.037	0.003	0.008
StHs6/80-G_4	NIST612	JET-X	5 Hz	0.825	2.048	0.004	0.008
StHs6/80-G_5	NIST612	JET-X	5 Hz	0.829	2.047	0.004	0.008
Mean				0.828	2.045		
2 SD				0.009	0.010		
2 SD, %				1.05	0.51		
Accuracy, %				0.15	0.34		
Reference value (a)				0.826	2.038		
(b)				0.826	2.038		
ATHO-G (MPI-DING rhyolite, Pb 5.67 ppm)							
ATHO-G_1	BCR-2G	JET-X	10 Hz	0.845	2.080	0.003	0.006
ATHO-G_2	BCR-2G	JET-X	10 Hz	0.847	2.080	0.003	0.006
ATHO-G_3	BCR-2G	JET-X	10 Hz	0.845	2.074	0.003	0.006
ATHO-G_4	BCR-2G	JET-X	10 Hz	0.845	2.076	0.003	0.006
ATHO-G_5	BCR-2G	JET-X	10 Hz	0.844	2.076	0.003	0.006
ATHO-G_1	BCR-2G	JET-X	10 Hz	0.847	2.085	0.003	0.006
ATHO-G_2	BCR-2G	JET-X	10 Hz	0.847	2.075	0.003	0.006
ATHO-G_3	BCR-2G	JET-X	10 Hz	0.847	2.076	0.003	0.007
ATHO-G_4	BCR-2G	JET-X	10 Hz	0.843	2.078	0.003	0.006
ATHO-G_5	BCR-2G	JET-X	10 Hz	0.843	2.074	0.003	0.006
ATHO-G_1	NIST612	JET-X	10 Hz	0.838	2.071	0.003	0.008
ATHO-G_2	NIST612	JET-X	5 Hz	0.843	2.076	0.004	0.009

ATHO-G_3	NIST612	JET-X	5 Hz	0.851	2.084	0.004	0.009
ATHO-G_4	NIST612	JET-X	5 Hz	0.848	2.082	0.004	0.009
ATHO-G_5	NIST612	JET-X	5 Hz	0.842	2.076	0.004	0.010
Mean				0.845	2.078		
2 SD				0.006	0.008		
2 SD, %				0.71	0.39		
Accuracy, %				0.34	0.17		
Reference value (a)				0.842	2.074		
(b)				0.842	2.073		
(c)				0.842	2.075		
(d)				0.841	2.076		

T1-G (MPI-DING quartz-diorite, Pb 11.6 ppm)

T1-G_1	NIST612	JET-X	3 Hz	0.836	2.081	0.004	0.009
T1-G_2	NIST612	JET-X	3 Hz	0.840	2.075	0.004	0.009
T1-G_3	NIST612	JET-X	3 Hz	0.835	2.090	0.004	0.008
T1-G_4	NIST612	JET-X	3 Hz	0.835	2.078	0.004	0.009
T1-G_5	NIST612	JET-X	3 Hz	0.833	2.073	0.004	0.009
T1-G_1	NIST612	JET-X	5 Hz	0.834	2.075	0.004	0.008
T1-G_2	NIST612	JET-X	5 Hz	0.838	2.059	0.004	0.008
T1-G_3	NIST612	JET-X	5 Hz	0.838	2.086	0.004	0.009
T1-G_5	NIST612	JET-X	5 Hz	0.832	2.085	0.004	0.009
T1-G_6	NIST612	JET-X	5 Hz	0.837	2.066	0.003	0.008
Mean				0.836	2.077		
2 SD				0.005	0.019		
2 SD, %				0.55	0.91		
Accuracy, %				-0.19	-0.20		
Reference value (a)				0.837	2.081		
(b)				0.837	2.081		

GOR132-G (MPI-DING komatiite, Pb 19.5 ppm)

GOR132-G_1	BCR-2G	JET-X	5 Hz	0.821	2.008	0.003	0.005
GOR132-G_2	BCR-2G	JET-X	5 Hz	0.818	2.007	0.003	0.005
GOR132-G_3	BCR-2G	JET-X	5 Hz	0.817	2.008	0.002	0.005
GOR132-G_4	BCR-2G	JET-X	5 Hz	0.817	2.010	0.003	0.005
GOR132-G_5	BCR-2G	JET-X	5 Hz	0.817	2.010	0.002	0.005
GOR132-G_1	BCR-2G	JET-X	5 Hz	0.827	2.034	0.003	0.005
GOR132-G_2	BCR-2G	JET-X	5 Hz	0.819	2.016	0.003	0.005
GOR132-G_3	BCR-2G	JET-X	5 Hz	0.814	2.006	0.003	0.005
GOR132-G_4	BCR-2G	JET-X	5 Hz	0.816	2.012	0.003	0.005
GOR132-G_5	BCR-2G	JET-X	5 Hz	0.816	2.008	0.003	0.006
GOR132-G_1	NIST612	JET-X	2 Hz	0.820	2.011	0.004	0.009
GOR132-G_2	NIST612	JET-X	2 Hz	0.820	2.015	0.004	0.008
GOR132-G_3	NIST612	JET-X	2 Hz	0.822	2.025	0.004	0.009
GOR132-G_4	NIST612	JET-X	2 Hz	0.819	2.020	0.004	0.008
GOR132-G_5	NIST612	JET-X	2 Hz	0.819	2.017	0.004	0.008
Mean				0.819	2.014		
2 SD				0.006	0.015		
2 SD, %				0.74	0.76		
Accuracy, %				0.36	0.29		
Reference value (a)				0.816	2.008		
(b)				0.817	2.011		

GSD-1G (USGS doped basalt glass, Pb 48 ppm)

GSD-1G_3	BCR-2G	N-X	1 Hz	0.804	1.972	0.003	0.006
GSD-1G_4	BCR-2G	N-X	1 Hz	0.806	1.972	0.003	0.006
GSD-1G_5	BCR-2G	N-X	1 Hz	0.803	1.975	0.003	0.006
GSD-1G_6	BCR-2G	N-X	1 Hz	0.800	1.972	0.003	0.006
GSD-1G_7	BCR-2G	N-X	1 Hz	0.806	1.985	0.003	0.006
GSD-1G_1	NIST612	JET-X	1 Hz	0.807	1.991	0.003	0.007
GSD-1G_2	NIST612	JET-X	1 Hz	0.805	1.975	0.003	0.007
GSD-1G_3	NIST612	JET-X	1 Hz	0.810	1.988	0.003	0.007
GSD-1G_4	NIST612	JET-X	1 Hz	0.805	1.984	0.003	0.007
GSD-1G_5	NIST612	JET-X	1 Hz	0.801	1.983	0.003	0.007
Mean				0.805	1.980		
2 SD				0.006	0.015		
2 SD, %				0.74	0.73		
Accuracy, %				0.07	-0.38		
Reference value (a)				0.804	1.987		

Note: 2SE with each analysis is bracketing standards propagated in-run error. Pb concentration data are from Jochum et al. (2006) for MPI-DING glasses; Guillong et al. (2005) for GSD-1G; GeoReM (<http://georem.mpch-mainz.gwdg.de>), Application Ver. 15, 2013) for BHVO-2G. Accuracy compared with preferred values of calculated (Jochum et al., 2011). Reference values of Pb isotopes are (a) Jochum et al. (2011); (b) Jochum et al. (2005b); (c) Paul et al. (2011); (d) Souders and Sylvester (2007).