

Supplementary Table S1. Results of each experiment and sample-by-sample averages of volatile concentrations

Sample name	Frozen Crushing (vesicle)								Heating (solid)		
	Weight (g)	⁴ He/ ²⁰ Ne	³ He/ ⁴ He (10 ⁻⁵)	³ He (10 ⁻¹⁵ mol/g)	S (10 ⁻⁹ mol/g)	F (10 ⁻⁹ mol/g)	Cl (10 ⁻⁹ mol/g)	Br (10 ⁻⁹ mol/g)	Weight (g)	³ He/ ⁴ He (10 ⁻⁵)	³ He (10 ⁻¹⁵ mol/g)
418R002											
(Mid Atlantic Ridge Basalt (MARB))											
1	0.398	> 2048	1.23 ± 0.01	10.1 ± 0.6	< 50*	< 3800**	209 ± 9	< 10 (N.D)	0.151	1.07 ± 0.13	1.5 ± 0.2
2	0.418	> 520	1.19 ± 0.01	9.5 ± 0.6	< 48*	< 2400**	204 ± 3	31 ± 24	0.152	1.11 ± 0.08	2.7 ± 0.2
3	0.117	> 106	1.16 ± 0.01	29.1 ± 1.7	< 170*	4620 ± 339	434 ± 11	45 ± 2	0.078	1.26 ± 0.25	2.0 ± 0.4
4	0.501	> 952	1.18 ± 0.01	6.0 ± 0.3	422 ± 2	< 4500*	< 200 (N.D)	143 ± 10	0.198	1.07 ± 0.22	1.0 ± 0.2
5	0.475	> 1195	1.18 ± 0.01	6.5 ± 0.3	245 ± 2	3777 ± 469	< 200 (N.D)	27 ± 10	0.201	1.10 ± 0.07	2.1 ± 0.1
Average(MARB)			1.19 ± 0.02	12.2 ± 4.7	333 ± 104	4198 ± 711	282 ± 80	61 ± 34		1.12 ± 0.11	1.9 ± 0.4
RY380-R03b											
(East Pacific Rise Basalt (EPRB))											
1	0.107	> 521	1.27 ± 0.03	6.9 ± 0.4	< 83*	< 6900**	< 200 (N.D)	< 10 (N.D)	0.050	1.26 ± 0.15	5.2 ± 0.6
2	0.087	> 30	1.25 ± 0.02	8.7 ± 0.5	< 100*	< 7500**	< 200 (N.D)	< 10 (N.D)	0.045	1.28 ± 0.19	4.8 ± 0.7
3	0.355	> 22	1.31 ± 0.01	6.7 ± 0.4	< 56*	< 3500**	< 200 (N.D)	346 ± 4	0.046	1.35 ± 0.11	8.7 ± 0.7
4	0.528	> 156	1.33 ± 0.01	5.9 ± 0.4	–	–	–	–	0.199	1.31 ± 0.03	7.2 ± 0.2
5	0.515	> 5075	1.37 ± 0.01	3.7 ± 0.2	27 ± 8	< 2400**	165 ± 4	< 10 (N.D)	0.202	1.32 ± 0.03	9.2 ± 0.2
6	0.416	> 110	1.41 ± 0.02	4.0 ± 0.2	87 ± 1	2459 ± 19	< 200 (N.D)	649 ± 19	0.148	1.19 ± 0.09	2.9 ± 0.2
7	0.310	> 18	1.36 ± 0.02	4.6 ± 0.3	192 ± 1	2965 ± 39	< 200 (N.D)	665 ± 26	0.103	1.22 ± 0.18	2.2 ± 0.3
Average(EPRB)			1.33 ± 0.03	5.8 ± 0.8	102 ± 59	2712 ± 275	165 ± 4	553 ± 115		1.28 ± 0.07	5.7 ± 1.2
ST14-DT6											
(North Fiji Basin Basalt (NFBB))											
1	0.221	> 52	1.22 ± 0.01	13.3 ± 0.8	< 90*	144 ± 10	< 200 (N.D)	1272 ± 5	0.099	1.27 ± 0.16	2.6 ± 0.3
2	0.608	> 35	1.24 ± 0.01	5.9 ± 0.4	< 33*	71 ± 12	< 200 (N.D)	148 ± 2	0.054	1.44 ± 0.21	4.7 ± 0.7
Average(NFBB)			1.23 ± 0.02	9.6 ± 4.1		108 ± 45		710 ± 565		1.35 ± 0.22	3.7 ± 1.5
Air (Ozima and Podosek, 2002)											
	0.318	0.140 ± 0.001									

*: The peaks of spectrum were smaller than blank solution.

**:: Could not be determined due to interferences of other elemental spectrums.

(N.D): Could not be determined due to low concentration. The upper limits were estimated as 10 nmol/g for Br and 200 nmol/g for Cl from fluctuation of base line.

Note: Sample-by-sample averages were calculated using well determined concentrations. Uncertainties of each experimental result are 1σ, and errors of

sample-by-sample averages were calculated as the combinations of the uncertainties from the reproducibility of each sample and of the individual measurement.

Especially, corrections of seawater contaminations are included in the errors of the average S concentrations. Net Cl concentrations may be lower than those listed because they may be affected by seawater contaminations.

Supplementary Table S2. Retention times of the peaks of anions and interferences by sodium hydroxide in two IC measurements

Peak name	Measurement-A	Measurement-B
	(7/6/11)	(9/1/11)
	Retention time [min]	Retention time [min]
Interference-1	1.12-1.13	1.13-1.14
Fluoride	1.93-1.97	1.78-1.83
Interference-2	2.01-2.07*	1.96-2.00*
Interference-3	2.86-3.59	2.78-3.18
Chloride	3.98-4.10	3.32-3.45
Bromide	6.49-6.58	5.84-5.98
Interference-4	7.90-8.07	7.01-7.80
Sulfate	10.13-10.25	9.33-9.39

*: could not be separated from fluoride peaks when the concentration of sodium hydroxide is 4mol/L.

Note: Since retention times of interferences are obviously different from those of fluoride, chloride, bromide and sulfate, we could mostly separate the interferences from the peaks of the anions and measure F, Cl, Br and S concentrations.

Supplementary Table S3. $X/{}^3\text{He}$ ratios in vesicles of samples collected at each location where X is a volatile element

Sampling site	$\text{S}/{}^3\text{He}$ ($\times 10^6$)	$\text{F}/{}^3\text{He}$ ($\times 10^6$)	$\text{Cl}/{}^3\text{He}$ ($\times 10^6$)	$\text{Br}/{}^3\text{He}$ ($\times 10^6$)
Mid Atlantic Ridge	27 \pm 13	343 \pm 144	23 \pm 11	5 \pm 3
East Pacific Rise	18 \pm 11	469 \pm 82	28 \pm 4	96 \pm 24
North Fiji Back-Arc Basin		11 \pm 7		74 \pm 67
Average of $X/{}^3\text{He}$ ratios	22 \pm 13	274 \pm 192	26 \pm 9	58 \pm 51

Note: Please see Table 1 for the calculations.

Supplementary Table S4. Element concentrations and $X/{}^3\text{He}$ ratios in the bulk of samples collected at each location where X is a volatile element

Sampling site	${}^3\text{He}$ (10^{-15}mol/g)	S (10^{-6}mol/g)	S/ ${}^3\text{He}$ ($\times 10^9$)	F (10^{-6}mol/g)	F/ ${}^3\text{He}$ ($\times 10^9$)	Cl (10^{-6}mol/g)	Cl/ ${}^3\text{He}$ ($\times 10^9$)	Br (10^{-6}mol/g)	Br/ ${}^3\text{He}$ ($\times 10^9$)
Mid Atlantic Ridge	14.1 \pm 4.9	0.3 - 10.3	0.02 - 0.73	9.4 - 532	0.7 - 37.7	0.6 - 35.8	0.04 - 2.54	0.1 - 7.8	0.01 - 0.55
East Pacific Rise	11.5 \pm 1.6	0.1 - 3.1	0.01 - 0.27	6.1 - 344	0.5 - 29.8	0.4 - 20.9	0.03 - 1.81	1.2 - 70.1	0.11 - 6.08
North Fiji Back-Arc Basin	13.2 \pm 3.4			0.2 - 14	0.02 - 1.0			1.6 - 90.0	0.12 - 6.81
Average of $X/{}^3\text{He}$ ratios			0.02 - 0.50		0.4 - 22.9		0.04 - 2.18		0.08 - 4.48

Note: Please see Table 1 and text for the calculations.