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FULL PAPER

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Volatile (F and Cl) concentrations in Iwate olivine-hosted melt inclusions indicating low-temperature subduction

Estelle F Rose-Koga^{1*}, Kenneth T Koga¹, Morihisa Hamada², Thomas H  louis¹, Martin J Whitehouse³ and Nobumichi Shimizu⁴

Abstract

Investigation of olivine-hosted melt inclusions provides information about the abundance of volatile elements that are often lost during subaerial eruptions of lavas. We have measured the abundances of H₂O, CO₂, F, Cl, and S as well as Pb isotopes in 29 melt inclusions in the scoria of the 1686 eruption of the Iwate volcano, a frontal-arc volcano in the northeast Japan arc. Pb isotope compositions identify that Iwate magma is derived from a mixture of depleted mantle, subducted basalt, and sediment. Systematics of F in comparison to MORB and other arc magma indicates that (1) the slab surface temperature must be among the lowest on Earth and (2) hydrous minerals, such as amphibole, humites, and/or mica, must be present as residual phases during the dehydration of the slab.

Introduction

Japan arc lavas display a wide range of compositions mainly interpreted as involving aqueous fluids released from the subducting plates (e.g., Nakamura and Iwamori 2009 and references therein). Iwate volcano, is situated in the northeastern (NE) Japan arc (Figure 1), and mainly consists of Quaternary low-K basalts and andesites (e.g., Itoh and Doi 2005; Kuritani et al. 2013). The chemical variations of the Iwate lavas have been attributed to the different proportion of fluids coming from sediment and/or altered oceanic crust (Shibata and Nakamura 1997; Sano et al. 2001; Hanyu et al. 2006). Both velocity tomography measurements in the mantle wedge above the slab and mapping of earthquake size distribution within the slab point towards a fluid source located right above of the slab at depths of 140 to 150 km (Wyss et al., 2001). The pre-eruptive volatile content of these magmas is of fundamental importance to understand the chemistry of the fluids after their interaction with the mantle wedge. Fluid chemistry will elucidate melting processes and characterize the source mantle. Since major volatile elements (H₂O, CO₂,

SO₂) will have degassed from the erupted lava, melt inclusions trapped in early crystallizing olivine carry the best chance of retaining dissolved volatiles of magmas at depth (e.g., Sobolev 1996).

However, a note of caution is sounded because melt inclusions are leaky containers for H₂O (Chen et al. 2011; Gaetani et al. 2012). CO₂ solubility in silicate melt is significantly lower than that of H₂O and therefore CO₂ degassing begins earlier during magma ascent, i.e. from a deeper depth (e.g. Shishkina et al. 2010). Therefore, the CO₂ concentration measurements in melt inclusions is at best considered as minimum values. Sulfur concentrations measured in melt inclusions also display a degassing trend (e.g. Spilliaert et al. 2006). In the present olivine-hosted melt inclusion study we use F and Cl concentrations to assess the pre-degassed volatile compositions of the basaltic melt underneath Iwate volcano, Japan. We report that the volatile contents of Iwate magma are (i) solely due to fluids coming from the slab and (ii) the source region of the mantle wedge undergoing partial melting is highly depleted.

Samples and methods

The Iwate samples are some of the least differentiated rocks on the volcanic front of the Japanese island arcs (Figure 1). The olivine-hosted melt inclusions of the scoria

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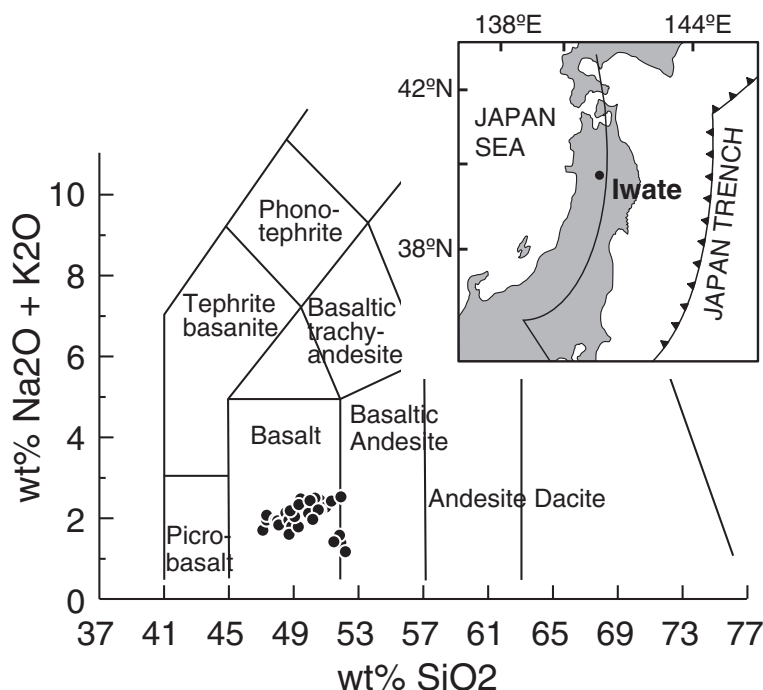


Figure 1 Diagram reporting SiO_2 as a function of K_2O and Na_2O (wt.%) of the Iwate glassy melt inclusions. The all fall in the basalt domain with few of them touching the basaltic-andesite domain. Inset: map of NE Japan arc. The volcanic front is indicated by a line. Iwate volcano is a front arc volcano.

of the 1686 eruption of Iwate volcano are basaltic to basaltic-andesitic in composition (Figure 1). Olivine crystals are separated from the rock and 34 glassy melt inclusions were selected for major elements among which 29 were also analyzed for volatiles and Pb-isotopes. The hand sample is olivine, plagioclase, and rare pyroxene-phyric. The olivine phenocrysts range from <0.5 to 2 mm in size. Primary melt inclusions are easily found in the olivines, but only the glassy (much scarcer) inclusions were selected. No secondary melt inclusions were observed in the olivines, but some were observed in the pyroxenes.

Volatile concentrations (H_2O , CO_2 , F, S, and Cl, Table 1) were measured by SIMS at WHOI (Woods Hole, MA, USA) and EMP at LMV (Clermont-Ferrand, France). The mono-collection SIMS settings are detailed in other studies (Rose-Koga et al. 2008; additional file 2: table S5 in Helo et al. 2011; Rose-Koga et al. 2012) and summarized in the following: we used a Cs+ primary beam with a current of 1.5 nA. The electron gun was used to avoid charging of the sample surface. The sample was pre-sputtered for 3 min with a $30 \times 30 \mu\text{m}$ square raster, and analyses were performed on the central $15 \times 15 \mu\text{m}$ area, centered in the rastered area by a mechanical aperture placed at the secondary ion image plane. The instrument was operated with a mass resolving power of 6,600, allowing complete discrimination of interferences ($^{34}\text{S}^{1}\text{H}$ on ^{35}Cl , ^{17}O on $^{16}\text{O}^{1}\text{H}$, $^{29}\text{Si}^{1}\text{H}$ on ^{30}Si , and $^{31}\text{P}^{1}\text{H}$ on ^{32}S).

We used a critical illumination method. A $400 \mu\text{m}$ contrast aperture was used and the energy slit opening was $\pm 30 \text{ eV}$. Including pre-sputter time, each measurement lasted approximately 15 min. The maximum uncertainties, taking into account the reproducibility over 10 cycles of analyses and the uncertainties on the regression of the calibration line, were less than 5% for H_2O , 15% for CO_2 , and 10% for Cl, F, and S.

Cerium concentration measurements were performed with the WHOI 1280 SIMS. The duoplasmatron with a $10 \text{ nA } ^{16}\text{O}^-$ primary beam was used. A 10 kV secondary voltage was placed on the sample to accelerate the secondary ions and an energy filtering of -90 V was applied (e.g., Shimizu and Hart, 1982). A $20 \mu\text{m}$ contrast aperture is used, and a mass resolution power of 1,250 is achieved by setting the entrance slit at 405 mm and exit slit at 972 mm. A deadtime correction of 28 ns is applied. Prior to the analysis, a pre-sputter of 240 s was performed and the total analysis time is $\sim 20 \text{ min}$. During each analysis cycle, intensities for the following isotopes were collected: ^{30}Si (5 s), ^{88}Sr (10 s), ^{89}Sr (10 s), (10 s), ^{90}Zr (10 s), ^{93}Nb (10 s), ^{138}Ba (10 s), ^{139}La (10 s), and ^{140}Ce (10 s). The beam size (nA) and duration of each measurement were adjusted (decreased and increased, respectively) to accommodate the measurement of small-diameter melt inclusions. We used the basaltic reference glasses KL2-G and ALV519-4-1, to generate the working curves and

Table 1 major element concentrations (in wt%), corrected from olivine addition, of 34 glassy olivine-hosted melt inclusions from Iwate volcano

	Olivine addition (%)	SiO ₂	MgO	FeO	MnO	K ₂ O	CaO	Na ₂ O	Al ₂ O ₃	TiO ₂	P ₂ O ₅
ITM1.1	4.8	48.9(4)	7.89(10)	11.2(3)	0.15(7)	0.070(1)	8.45(6)	1.75(7)	16.2(2)	0.71(1)	0.040(2)
ITM1.2	3.2	48.9(4)	7.37(9)	10.5(3)	0.2(1)	0.100(2)	8.33(6)	1.87(7)	16.1(2)	0.84(1)	0.120(7)
ITM2	7.0	48.9(4)	7.59(10)	12.9(4)	0.19(9)	0.090(2)	8.14(6)	2.08(8)	14.5(2)	0.87(1)	0.080(5)
ITM5.1	6.0	48.6(4)	7.51(10)	11.1(3)	0.16(7)	0.090(2)	9.07(7)	2.02(8)	16.8(2)	0.72(1)	n.d.
ITM5.2	10.2	50.7(5)	8.14(10)	12.1(4)	0.18(8)	0.110(2)	8.90(7)	2.33(9)	15.3(2)	0.85(1)	0.030(2)
ITM6	6.8	48.8(4)	8.02(10)	12.0(4)	0.18(9)	0.140(3)	9.12(7)	1.71(7)	16.2(2)	0.76(1)	n.d.
ITM8	10.4	48.2(4)	9.26(12)	12.2(4)	0.11(5)	0.150(3)	8.87(7)	1.67(7)	15.3(2)	0.80(1)	0.070(4)
ITM10.1	10.6	50.6(5)	7.55(10)	10.4(3)	0.2(1)	0.150(3)	8.81(7)	2.03(8)	15.2(2)	0.81(1)	0.040(2)
ITM10.2	13.6	50.3(5)	7.32(9)	10.1(3)	0.18(8)	0.070(2)	8.65(7)	1.88(8)	16.8(2)	0.65(1)	0.050(3)
ITM11	6.8	51.0(5)	7.56(10)	10.9(3)	0.17(8)	0.170(4)	8.15(6)	2.08(8)	15.2(2)	0.73(1)	0.150(9)
ITM12	4.8	48.3(4)	7.97(10)	11.0(3)	0.2(1)	0.100(2)	8.97(7)	1.78(7)	16.1(2)	0.70(1)	0.150(9)
ITM13	8.0	48.0(4)	8.31(11)	10.9(3)	0.14(6)	0.110(2)	9.34(7)	1.83(7)	16.4(2)	0.70(1)	0.080(5)
ITM14	9.4	52.0(5)	8.25(11)	11.4(3)	0.17(8)	0.220(5)	8.71(7)	1.32(5)	15.2(2)	0.86(1)	0.150(9)
ITM15.1	8.0	50.5(5)	6.91(9)	12.0(4)	0.17(8)	0.190(4)	8.18(6)	2.29(9)	14.9(2)	1.01(1)	0.040(2)
ITM15.2	7.8	51.5(5)	7.02(9)	12.2(4)	0.3(1)	0.140(3)	8.20(6)	2.25(9)	14.4(2)	0.97(1)	0.140(8)
ITM15.3	9.0	51.1(5)	7.36(9)	12.8(4)	0.2(1)	0.150(3)	7.66(6)	2.11(8)	14.4(2)	0.75(1)	0.110(6)
ITM16	9.2	48.8(4)	7.48(10)	11.9(4)	0.12(6)	0.050(1)	9.70(7)	1.54(6)	15.8(2)	0.74(1)	n.d.
ITM17	13.6	51.2(5)	8.76(11)	13.8(4)	0.3(1)	0.250(5)	7.76(6)	2.01(8)	12.5(2)	0.98(1)	0.040(2)
ITM19.1	11.0	49.1(4)	7.40(9)	10.9(3)	0.15(7)	0.110(2)	9.21(7)	1.89(8)	16.7(2)	0.80(1)	0.17(1)
ITM19.2	13.6	44.3(4)	7.90(10)	11.7(3)	0.09(4)	0.140(3)	7.84(6)	1.73(7)	20.9(3)	0.85(1)	0.29(2)
ITM20	10.8	47.5(4)	8.17(10)	12.1(4)	0.19(9)	0.080(2)	9.34(7)	1.85(7)	16.0(2)	0.71(1)	0.110(6)
ITM23	8.6	51.6(5)	7.57(10)	12.2(4)	0.2(1)	0.170(4)	8.86(7)	1.23(5)	15.8(2)	1.01(1)	0.150(9)
ITM24	8.0	52.4(5)	7.38(9)	12.1(4)	0.20(9)	0.140(3)	9.18(7)	1.03(4)	15.4(2)	1.00(1)	0.28(2)
IEM26	11.0	48.5(4)	7.94(10)	12.0(4)	0.19(9)	0.130(3)	9.17(7)	1.84(7)	16.5(2)	0.64(1)	0.030(2)
IEM28.1	10.0	47.4(4)	8.74(11)	10.8(3)	0.18(8)	0.130(3)	8.32(6)	1.92(8)	15.9(2)	0.90(1)	0.100(6)
IEM31.2	9.2	52.1(5)	6.08(8)	12.8(4)	0.2(1)	0.220(5)	7.43(6)	2.29(9)	14.4(2)	0.89(1)	0.100(6)
IEM34.1	16.8	50.7(5)	9.84(13)	13.8(4)	0.20(9)	0.230(5)	7.49(6)	1.96(8)	13.2(2)	0.90(1)	0.18(1)
IEM34.3	9.8	47.2(4)	8.46(11)	11.9(4)	0.20(9)	0.110(2)	10.02(8)	1.59(6)	16.3(2)	0.71(1)	0.18(1)
IEM34.4	17.8	50.0(5)	9.98(13)	14.0(4)	0.13(6)	0.170(4)	7.78(6)	1.92(8)	13.0(2)	0.90(1)	0.140(8)
IEM35	9.8	49.5(4)	6.83(9)	10.3(3)	0.16(7)	0.140(3)	8.51(6)	2.30(9)	16.5(2)	0.62(1)	0.110(6)
IEM36	13.2	49.4(4)	7.56(10)	11.9(4)	0.16(7)	0.100(2)	8.45(6)	2.23(9)	16.0(2)	0.66(1)	0.19(1)
IEM37	12.2	52.0(5)	8.47(11)	12.2(4)	0.14(7)	0.170(4)	8.90(7)	1.22(5)	15.2(2)	0.82(1)	0.070(4)
IEM38	15.0	50.2(5)	8.04(10)	13.6(4)	0.18(9)	0.150(3)	8.41(6)	2.26(9)	14.0(2)	1.09(2)	0.140(8)
IEM39	10.4	49.4(4)	7.20(9)	10.3(3)	0.12(6)	0.100(2)	9.16(7)	1.67(7)	16.7(2)	0.92(1)	0.030(2)

Concentrations are given after normalization to 100% on an anhydrous basis. Units in parenthesis represent 1 standard deviation of least units cited; thus, 48.8(4) should be read as 48.8 ± 0.4.

calculate the concentrations of the elements in our samples. For the purpose of this study, only the Ce concentrations are reported in Table 2.

The Pb isotope compositions were measured by multi-collection SIMS 1280 at the NORDSIMS (Stockholm, Sweden). In summary, samples were sputtered with a focused O₂⁻ primary beam of 8 nA, with a primary accelerating voltage of 13 kV. After a 90 s pre-sputter with a

25 × 25 μm raster, the raster was stopped during analyses. We used a Kohler illumination method. This resulted in a 15 μm flat-bottomed pit centered in the 25 μm square raster-cleaned area. Secondary positive ions were accelerated at 10 kV and analyzed at mass resolution of 4,800 to resolve all mass interferences. The size of the field aperture was set at 4,000 μm, and the contrast aperture at 400 μm. The centered energy

Table 2 Volatile elements and cerium concentrations corrected from olivine addition and Pb isotopes in 29 olivine-hosted glassy melt inclusions from Iwate volcano

	CO ₂ ppm	H ₂ O wt%	F ppm	S ppm	Cl ppm	Ce ppm	H ₂ O/Ce	²⁰⁷ Pb/ ²⁰⁶ Pb	²⁰⁸ Pb/ ²⁰⁶ Pb	²⁰⁶ Pb/ ²⁰⁴ Pb	²⁰⁷ Pb/ ²⁰⁴ Pb	²⁰⁸ Pb/ ²⁰⁴ Pb					
ITM1.1	368(140)	2.82(3)	123(2)	1505(20)	408(7)	5.4(2)	5200(700)	0.836	0.002	2.084	0.004	19.20	0.13	16.05	0.12	40.30	0.31
ITM2	315(320)	2.80(1)	164(6)	1071(30)	414(10)	4.7(2)	5900(400)	0.840	0.002	2.081	0.004	18.49	0.10	15.53	0.09	38.75	0.21
ITM5.1	331(5)	2.80(3)	178(1)	1952(10)	394(3)	4.0(1)	7100(300)	0.844	0.004	2.094	0.007	18.31	0.18	15.45	0.13	38.61	0.38
ITM5.2	5(1)	0.77(1)	171(2)	258(2)	350(3)	5.4(3)	1480(90)	0.843	0.001	2.084	0.003	18.56	0.10	15.65	0.08	38.95	0.20
ITM6	28(8)	2.65(1)	155(1)	1527(2)	401(1)	n.d.	n.d.	0.838	0.004	2.084	0.008	18.50	0.30	15.49	0.28	38.80	0.67
ITM8	175(41)	2.74(3)	200(30)	951(14)	423(20)	4.7(4)	5700(1100)	0.840	0.001	2.085	0.004	18.82	0.14	15.81	0.12	39.52	0.30
ITM10.1	479(23)	2.71(1)	159(4)	1273(20)	463(10)	5.1(2)	5300(400)	0.837	0.001	2.081	0.003	18.99	0.12	15.90	0.10	39.81	0.23
ITM11	224(3)	2.57(1)	189(1)	932(5)	387(2)	5.1(2)	5100(200)	0.836	0.002	2.083	0.004	19.23	0.20	16.07	0.16	40.32	0.40
ITM12	6446(200)	2.85(2)	143(1)	1515(8)	405(1)	3.8(1)	7600(300)	0.841	0.002	2.082	0.004	18.86	0.16	15.85	0.13	39.55	0.34
ITM13	3835(590)	2.44(1)	131(3)	1478(40)	372(9)	3.7(2)	6400(600)	0.842	0.003	2.074	0.007	17.74	0.24	14.94	0.19	37.04	0.44
ITM14	32(1)	1.00(1)	138(1)	456(1)	337(2)	4.9(2)	2000(100)	0.839	0.002	2.079	0.003	18.70	0.11	15.68	0.10	39.15	0.23
ITM15.1	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.843	0.005	2.091	0.011	19.35	0.21	16.28	0.23	40.70	0.54
ITM15.2	177(110)	2.38(1)	178(1)	644(2)	409(3)	4.1(3)	5900(400)	0.836	0.002	2.084	0.005	18.82	0.12	15.73	0.09	39.49	0.27
ITM16	546(68)	2.28(2)	144(1)	1671(20)	385(3)	4.4(2)	5200(300)	0.841	0.003	2.085	0.007	18.40	0.21	15.46	0.19	38.62	0.40
ITM17	4348(640)	0.62(1)	230(1)	198(1)	427(2)	4.7(2)	1310(90)	0.850	0.002	2.097	0.004	18.43	0.15	15.66	0.11	38.93	0.30
ITM19.1	205(71)	0.41(4)	178(16)	464(40)	406(40)	6.0(3)	690(100)	0.840	0.001	2.084	0.002	18.79	0.07	15.77	0.06	39.44	0.14
ITM23	1071(190)	0.22(1)	196(1)	204(1)	361(2)	5.9(2)	370(30)	0.839	0.001	2.083	0.003	18.84	0.12	15.80	0.10	39.52	0.25
ITM24	77(10)	0.27(1)	204(1)	263(1)	372(2)	6.0(3)	450(40)	0.842	0.001	2.082	0.003	18.63	0.09	15.68	0.07	39.06	0.19
IEM26	755(62)	2.37(3)	173(1)	1805(6)	339(2)	2.9(3)	8200(800)	0.844	0.003	2.089	0.007	18.13	0.18	15.29	0.14	38.13	0.37
IEM28.1	380(6)	3.65(3)	147(1)	1705(10)	352(3)	10(2)	3600(800)	0.843	0.002	2.087	0.004	18.53	0.16	15.61	0.12	38.95	0.32
IEM31.2	4714(440)	0.77(1)	60(1)	287(3)	515(8)	6.9(4)	1120(70)	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
IEM34.1	46(4)	0.82(1)	216(1)	288(1)	393(2)	6.2(2)	1330(70)	0.839	0.001	2.079	0.002	18.63	0.11	15.63	0.09	39.00	0.22
IEM34.3	326(11)	2.44(1)	121(1)	1518(5)	366(2)	2.5(2)	9700(1000)	0.839	0.001	2.091	0.003	19.13	0.11	16.04	0.10	40.27	0.24
IEM34.4	55(4)	0.69(1)	192(1)	951(5)	348(2)	6.4(4)	1070(80)	0.839	0.002	2.089	0.003	18.77	0.08	15.75	0.06	39.47	0.15
IEM35	164(4)	2.94(2)	174(1)	1071(4)	381(3)	4.4(3)	6600(400)	0.837	0.002	2.082	0.004	18.64	0.16	15.60	0.14	39.08	0.34
IEM36	n.d.	n.d.	74(58)	1298(30)	397(30)	3.7(2)	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
IEM37	n.d.	n.d.	158(11)	48(25)	448(30)	n.d.	n.d.	0.839	0.001	2.077	0.003	18.34	0.08	15.40	0.07	38.38	0.18
IEM38	n.d.	n.d.	143(96)	407(37)	514(26)	n.d.	n.d.	0.841	0.001	2.076	0.003	18.34	0.10	15.41	0.09	38.34	0.18
IEM39	n.d.	n.d.	n.d.	1058(60)	411(37)	n.d.	n.d.	0.840	0.003	2.069	0.007	18.24	0.20	15.31	0.17	38.01	0.41

Analytical uncertainties are indicated in parenthesis with last significant digits (1 standard deviation of analysis). Errors for Pb-isotope compositions represent 2 standard error of the mean. n.d., not determined. No vapor bubble correction was applied on the volatile concentration measurements in the melt.

window was adjusted to ± 45 eV. The multicollection system was composed of five ion-counting electron multipliers set on moveable trolleys (L2, L1, C, H1, and H2). The detection array was configured to measure ²⁰⁴Pb + in the electron multiplier set on the trolley position L2, ²⁰⁶Pb + in L1, ²⁰⁷Pb + in C (placed on the ion optical axis) and ²⁰⁸Pb + in H1. The measurements were conducted over 40 to 120 cycles with a 20 s counting time for each mass and a deadtime correction of 60 ns. A measurement lasted approximately 20 min for 40 cycles. With this setting, the sensitivity on ²⁰⁸Pb is 19 cts/ppm/nA. Details of Pb measurements are described in Rose-Koga et al. (2012).

Results

Major elements

Major element concentrations in melt inclusions are corrected from olivine post-entrapment crystallization and reported in Table 1. They were measured at Laboratoire Magmas et Volcans in Clermont-Ferrand, France, on a Cameca SX 100 using standard procedures outlined in Le Voyer et al. (2008). The Iwate melt inclusions have corrected MgO concentrations between 6.83 and 9.98 wt.% which reach higher values than the range described for the whole rock (3.98 to 7.50 wt.%; Sano et al. 2001). The forsterite values for the olivines range from 70.0 to 80.0 which denotes an evolved magma (Table 3). The melt

inclusions were plotted in a total alkali versus silica (TAS) diagram which classifies them as mainly basaltic with a few exceptions close to the basaltic-andesite domain (Figure 1).

Cerium and volatile (H₂O, CO₂, F, S, Cl) concentrations of melt inclusions

All concentrations are recalculated for olivine post-entrapment crystallization (Table 2). Cerium (Ce) concentrations in Iwate melt inclusions vary between 2.5 and 6.9 ppm with one higher value at 10 ppm. This represents one of the lowest ranges of Ce concentrations among arc melt inclusions. Other arc melt inclusions with low Ce concentration also extend to higher Ce concentrations, for example up to 35 ppm in Lesser Antilles (Bouvier et al. 2010) and up to 30 ppm in Central America (Sadofsky et al. 2008). Iwate thus represents the low end of the range of Ce concentrations in arc melt inclusions.

Iwate melt inclusions have variable concentrations of H₂O, S, and CO₂ which range from 0.2 to 3.7 wt% H₂O, 258 to 1,952 ppm S, and 5 to 6,446 ppm CO₂ (Table 2). The volatile contents (H₂O, S) reach values comparable to other Pacific rim melt inclusions such as Izu Oshima (up to 3.4 wt% and 1,500 ppm, respectively; Ikehata et al. 2009). F concentrations vary between 40 and 230 ppm and Cl concentrations between 337 and 515 ppm (Table 2). The average F and Cl concentration in the melt inclusions is 156 ± 44 ppm and 399 ± 45 ppm, respectively, demonstrating that F and Cl concentrations are relatively low and clustered compared to other Pacific rim melt inclusions (Figure 2). The low F concentrations are similar to those of melt inclusions from oceanic basalts from EPR (125 ± 43 ppm; Wanless and Shaw 2012), Juan de Fuca (117 ± 12 ppm; Wanless and Shaw 2012), and Gakkel ridge (150 ± 6 ppm; Shaw et al. 2010). The Cl concentrations of Iwate melt inclusions are enriched compared to that of melt inclusions from

Table 3 Major element concentrations (in wt.%) of the 26 Iwate olivines in which glassy melt inclusions where exposed

Olivine name	Melt incl. name	SiO ₂		CaO		FeO		MgO		MnO		NiO	
ITO1	ITM1.2	39.1	(0.7)	0.14	(0.04)	20.85	(0.73)	40.42	(0.61)	0.38	(0.19)	0.04	(0.021)
ITO2	ITM2	38.2	(0.7)	0.14	(0.05)	23.37	0.82	37.93	0.57	0.38	0.19	0.07	0.032
ITO4	ITM5.1; 5.2	38.5	(0.7)	0.17	(0.05)	21.19	0.74	39.67	0.60	0.30	0.15	0.08	0.039
ITO5	ITM6	38.5	(0.7)	0.12	(0.04)	21.22	(0.74)	39.40	(0.59)	0.30	(0.15)	0.09	(0.042)
ITO6	ITM8	39.1	(0.7)	0.10	(0.03)	19.73	0.69	41.52	0.62	0.26	0.13	0.06	0.028
ITO8	ITM10.1; 10.2	38.9	(0.7)	0.12	(0.04)	20.20	0.71	40.66	0.61	0.26	0.13	0.02	0.008
ITO10	ITM11	38.9	(0.7)	0.12	(0.04)	20.92	0.73	39.99	0.60	0.25	0.12	0.05	0.022
ITO11	ITM12	39.0	(0.7)	0.14	(0.04)	20.24	0.71	40.77	0.61	0.29	0.14	0.07	0.031
ITO12	ITM13	39.1	(0.7)	0.15	0.05	19.57	0.68	41.30	0.62	0.32	0.16	0.08	0.039
ITO13	ITM14	38.9	(0.7)	0.14	0.04	20.33	0.71	40.71	0.61	0.31	0.16	0.07	0.034
ITO15	ITM15.1; 15.2	38.5	(0.7)	0.14	0.05	23.67	0.83	37.78	0.57	0.36	0.18	0.01	0.007
ITO16	ITM16	38.2	(0.7)	0.17	0.05	22.08	0.77	38.61	0.58	0.34	0.17	0.05	0.022
ITO17	ITM17	38.7	(0.7)	0.16	0.05	22.15	0.78	38.89	0.58	0.38	0.19	0.02	0.007
ITO19	ITM19.1; 19.2	38.7	(0.7)	0.15	0.05	21.22	0.74	39.68	0.60	0.32	0.16	0.07	0.032
ITO20	ITM20	38.2	(0.7)	0.15	0.05	21.21	0.74	39.34	0.59	0.36	0.18	0.11	0.052
ITO24	ITM23	38.4	(0.7)	0.16	0.05	22.62	0.79	38.63	0.58	0.38	0.19	0.02	0.010
ITO25	ITM24	38.8	(0.7)	0.15	0.05	22.77	0.80	38.27	0.57	0.32	0.16	0.03	0.012
IEO26	IEM26	38.5	(0.7)	0.15	0.05	21.67	0.76	39.51	0.59	0.32	0.16	0.10	0.046
IEO28	IEM28.1	39.0	(0.7)	0.12	0.04	18.92	0.66	42.19	0.63	0.29	0.15	0.08	0.037
IEO31	IEM31.2	37.6	(0.6)	0.15	0.05	26.92	0.94	35.45	0.53	0.40	0.20	0.06	0.028
IEO34	IEM34.1; 34.3; 34.4	38.7	(0.7)	0.16	0.05	20.42	0.71	40.29	0.60	0.28	0.14	0.07	0.035
IEO35	IEM35	38.2	(0.7)	0.15	0.05	21.47	0.75	39.41	0.59	0.34	0.17	0.05	0.023
IEO36	IEM36	38.5	(0.7)	0.15	0.05	22.32	0.78	39.01	0.59	0.32	0.16	0.06	0.027
IEO37	IEM37	38.7	(0.7)	0.18	0.06	20.65	0.72	39.98	0.60	0.33	0.17	0.09	0.044
IEO38	IEM38	37.8	(0.6)	0.13	0.04	23.30	0.82	38.00	0.57	0.34	0.17	0.05	0.024
IEO39	IEM39	39.1	(0.7)	0.12	0.04	21.00	0.74	40.39	0.61	0.38	0.19	0.04	0.021

Units in parenthesis represent 1 standard deviation.

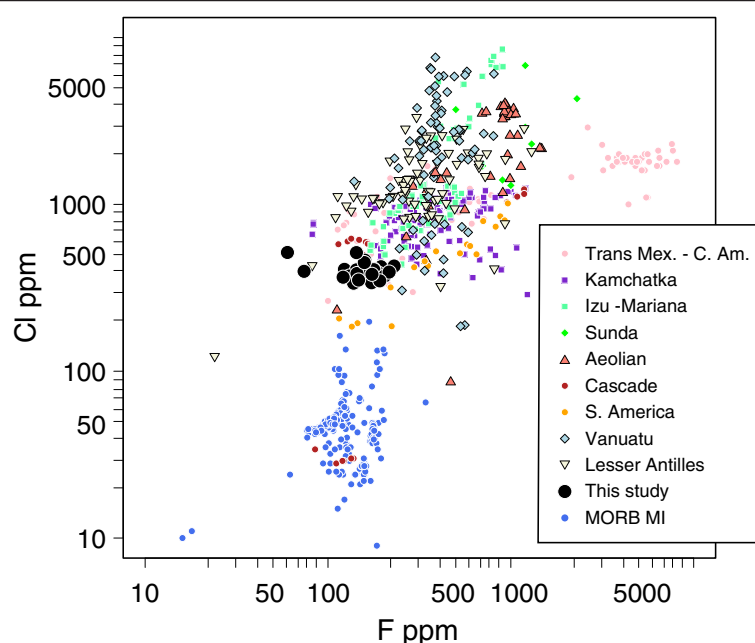


Figure 2 F concentrations versus Cl concentrations of Iwate melt inclusions (black filled circles). The Iwate melt inclusions are compared to other arc melt inclusions from the literature: Trans Mexican, Vigouroux et al., 2008; Central America, Sadofsky et al., 2008; Kamchatka, Portnyagin et al., 2007; Izu, Straub and Layne, 2003; Sunda, Elburg et al., 2007; Aeolian, Rose-Koga et al., 2012; Le Voyer unpublished data; Cascades, Le Voyer et al., 2010; S. America, Le Voyer et al., 2008; Vanuatu, Sorbadere et al. 2011; and Lesser Antilles, Bouvier et al. 2008, 2010. For MORB melt inclusions, data are from: Gakkel ridge, Shaw et al., 2010; and EPR and Juan de Fuca, Wanless, and Shaw, 2012.

oceanic basalts (EPR, 44 ± 10 ppm; Juan de Fuca, 61 ± 32 ; Gakkel ridge, 40 ± 45 ppm; Wanless and Shaw 2012; Shaw et al. 2010).

Pb isotope compositions of melt inclusions

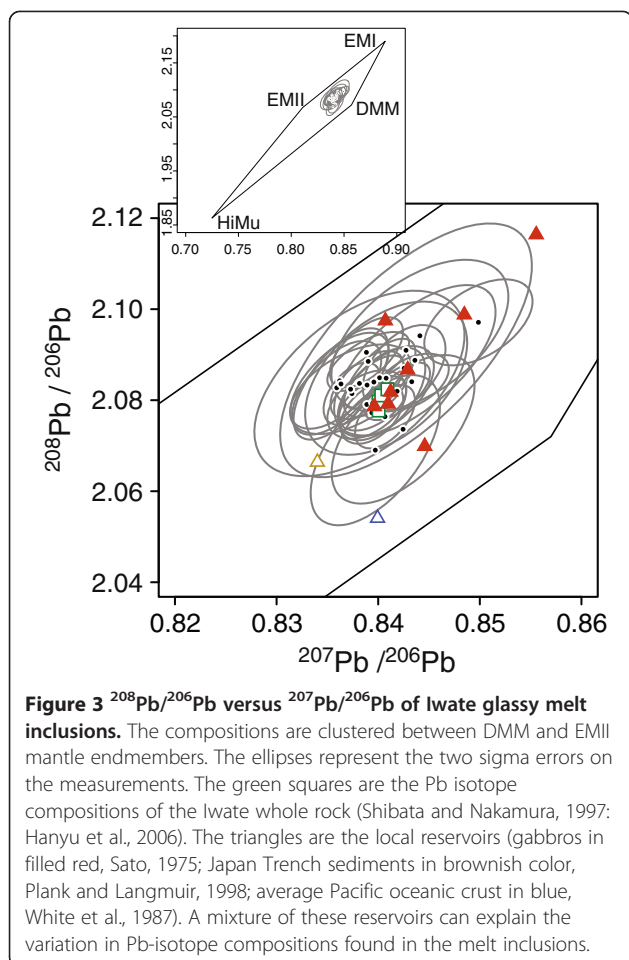
The Pb-isotope compositions of Iwate melt inclusions show more variability than that of their whole rock (Figure 3). This observation is commonly seen among relative abundances of trace elements in melt inclusions because the whole rock often represents an average of separate batches of magma that are trapped in the melt inclusions (e.g., Sobolev and Shimizu 1993; see review by Schiano 2003). The Iwate melt inclusions have Pb isotope compositions $^{207}\text{Pb}/^{206}\text{Pb}$ between 0.836 to 0.850 and $^{208}\text{Pb}/^{206}\text{Pb}$ between 2.069 ± 0.007 and 2.097 ± 0.004 (Table 2). These $^{208}\text{Pb}/^{206}\text{Pb}$ values are equal or higher than that of the neighboring contributing reservoir, the average Pacific oceanic crust (2.055, White et al. 1987), and the average sediments from the Japan Trench (2.067, Plank and Langmuir 1998). The range of $^{207}\text{Pb}/^{206}\text{Pb}$ variation is within the two endmember values: DMM (0.8570, Hart et al. 1992) and sediments (0.8341, Plank and Langmuir 1998). The melt inclusions' Pb-isotope compositions overlap the literature data of whole rock lower crustal material from NE Japan (Sato 1975; Figure 3).

Discussion

Magmatic degassing

H₂O concentrations measured in Iwate melt inclusions are at best considered as minimum water concentrations because melt inclusions are leaky containers for H₂O (Chen et al. 2011, 2012; Gaetani et al. 2012). An indication of water loss in Iwate melt inclusions is the weak anti-correlation between H₂O and TiO₂. Since availability of H₂O is the principal cause of subarc mantle partial melting, H₂O abundance often correlates with that of other incompatible elements, such as TiO₂, among primitive arc magmas (e.g., Kelley et al. 2006; Langmuir et al. 2006). Lack of such anti-correlation suggests the loss of the volatile, incompatible element, hydrogen, from melt inclusions.

The positive correlation between H₂O and S in Iwate melt inclusions then shows degassing of both elements (Figure 4). Sulfur degasses after CO₂ prior to H₂O saturation, and it exsolves at pressure of about 140 MPa (Spilliaert et al. 2006). CO₂ abundance is less than 1,000 ppm in nearly all the melt inclusions (i.e., five inclusions excluded). These melt inclusions recorded significant pre-entrapment degassing. Therefore, the positive linear correlation (slope 2.32 and correlation coefficient of 0.65) between CO₂ (for CO₂ < 1,000 ppm) and S indicates melt degassing prior to entrapment. The

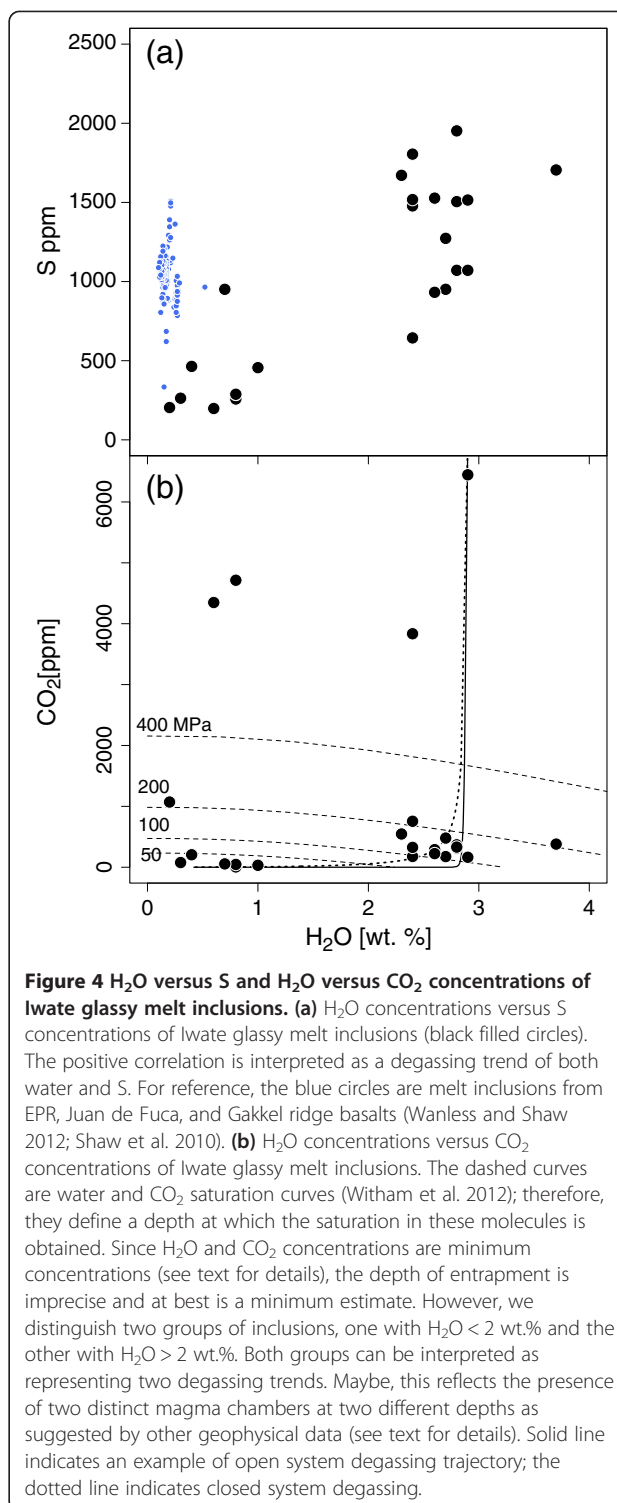


primitive CO_2 , S, and H_2O concentrations are indeterminate with the current data set. The maximum values we measured are 2,000 ppm for S and ≈ 4 wt.% for H_2O which represent the minimum values for primitive S and H_2O contents for Iwate melt inclusions.

F and Cl concentrations in Iwate volcano are clustered (relative standard deviations of F and Cl are 28 and 11%, respectively) and lower than most of other arc melt inclusions (Figure 2). There is no correlation between these halogens and the degassed volatile elements (H_2O , CO_2 , and S) which implies that the abundances of F and Cl are likely characterized by processes other than degassing and magma chamber crystallization. The halogens can be used to decipher additional information about the source of the Iwate magma. It should be noted, however, that Cl content is constant over the entire range of K_2O variations (0.07 to 0.25) and TiO_2 abundance does not correlate with F and Cl.

Magma chamber depth and H_2O - CO_2 saturation pressure

The degassing of CO_2 from the magma is obvious when reporting H_2O versus CO_2 in melt inclusions on a graph along with the saturation curves (SolEx: Witham et al.



2012, Figure 4). There are two groups, one with $\text{H}_2\text{O} < 2$ wt.% and the other with $\text{H}_2\text{O} > 2$ wt.%. Each trend individually has similar water content and could be classically interpreted as degassing trends in which significant variation of CO_2 is observed while H_2O remain relatively

constant (Figure 4 degassing trend). During magma ascent, CO₂ degasses first, but this degassing can be sluggish which causes aliquots of melt to be entrapped in the olivine crystals (basalt liquidus forming mineral) prior to complete degassing of CO₂. This could be the case of the higher CO₂ concentration on both trends. Then H₂O loss is observed at the end of the trends. The pressure indicated along the curve of Figure 4 cannot be used to estimate precisely the depth of entrapment, because the H₂O and CO₂ measured concentrations are minimum values. However, the fact that two separate trends coexist could be coincidentally considered as in agreement with the presence of two magma chambers (with slightly different water concentrations) suggested by geophysical data (tomography study and seismic swarms) showing the location of two magma chambers by S-wave velocity slowdowns under Iwate volcano, a crustal magma chamber around 2 km deep and a deeper one around 30 km (Nishida et al. 2008). Long-frequency volcanic swarms are also reported around 30 km depths immediately below the arc crust (Nakajima et al. 2001).

Halogen transport

F concentrations of Iwate melt inclusions are in the same range as those reported for MORB melt inclusions (Gakkel from 146 to 166 ppm, Shaw et al. 2010; EPR from 18 to 342 ppm; and Juan de Fuca from 62 to 123 ppm, Wanless and Shaw 2012). Cl concentrations in the Iwate melt inclusions are systematically higher than MORB melt inclusions by approximately an order of magnitude (Figure 2). The enrichment of fluid-mobile elements has been interpreted as a mixing of slab input to the melting region (e.g., Gill 1981; Tatsumi and Eggins 1995). Cl enrichment in arc melt inclusions is observed in multiple studies (Hamada and Fujii 2007; Shaw et al. 2008; Kelley et al. 2010; Rose-Koga et al. 2012; and see review by Wallace 2005). Iwate melt inclusions have a much lower Cl concentration compared to other arc melt inclusions. Other arc volcano melt inclusions rarely plot at lower Cl concentrations than Iwate samples except a few Ecuadorian melt inclusions (Le Voyer et al. 2008), a handful of Mariana melt inclusions (Kelley et al. 2010; Figure 2), and Shasta melt inclusions from dry high-aluminum olivine tholeiite (HAOT; Le Voyer et al. 2010).

Straub and Layne (2003) report significant F and Cl enrichment in clinopyroxene (cpx) and plagioclase-hosted melt inclusions and glass shards from the Izu arc volcanoes. Enrichment of Cl and absence of enrichment of F in Iwate magmas are contrasting characteristics compared to those of Izu and other Cl- and F-rich arc magmas. Simultaneous enrichment of F and Cl in arc lavas is interpreted as the evidence of element addition by aqueous fluid transport from slab to sub-arc mantle (Straub and Layne 2003). However, there is no F

enrichment of Iwate melt inclusions with respect to MORB melt inclusions. Furthermore, Iwate melt inclusions are the Cl- and F-poor endmember of arc melt inclusions. Arc magmas with elevated Cl, with the absence of concomitant F enrichment, therefore enforce the necessity of refining the existing halogen transport model. While the exact cause of the variations seen on the F versus Cl plot is unknown, some magmatic processes are less likely to cause these variations. First, we have argued that degassing of magma before and after entrapment of the inclusion would not be the cause of F and Cl variation. Second, because olivine is the liquidus mineral of basalt in crustal conditions, magma chamber crystal fractionation would not be recorded in primitive olivine melt inclusions. Element fractionation during partial melting and compositional variation of source mantle, perhaps due to metasomatism, are the remaining potential causes of the variation. While element fractionation during partial melting does occur, and F and Cl partition differently in mantle minerals (Dalou et al. 2011 and Table 4), the observed order of magnitude change in F and Cl abundances (between melt inclusions from MORB and Iwate melt inclusions; Figure 3) can only be derived from a highly variable degree of melting of the mantle. Because heterogeneity of non-metasomatized depleted mantle below arcs should have relatively similar composition worldwide (e.g., there are only 40% abundance difference of F between BSE (Lyubetskaya and Korenaga 2007) and DMM (Workman and Hart 2005), the large F and Cl abundance variations can only be derived from the compositional variation of arc metasomatic agents.

The low F and Cl magma from Iwate contains a significant quantity of H₂O, consistent with the typical arc magma characteristics. Magma genesis beneath Iwate volcano must be derived from partial melting of a depleted mantle wedge triggered by a water-rich metasomatic agent, typical in arc magmatism. Variation of F and Cl among arc primitive magmas should therefore indicate (1) fractionation processes during transfer of water-rich flux from slab and/or (2) source compositions of the water-rich metasomatic agent.

What are the magma sources?

We measured Pb-isotope compositions of the Iwate melt inclusions to decipher the source(s) and/or source mixing in these magmas. The isotopic compositions cluster around a single value (²⁰⁷Pb/²⁰⁶Pb = 0.840 and ²⁰⁸Pb/²⁰⁶Pb = 2.084), and 81% (22 out of 27) of inclusions are indistinguishable. While the limit of SIMS precision is a partial cause of this homogeneity, the mean Pb composition of Iwate magma is significantly different from depleted mantle (DMM 0.8570 and 2.072; Hart et al. 1992), they actually plot above the tie line between DMM

Table 4 F and Cl partition coefficients

D (Mineral/liquid)	F	Cl
(Olivine/basalt)	0.0015 to 0.0047 ^a 0.0007 to 0.0033 ^b 0.00024 to 0.00054 ^c	<0.0005 ^a
(Clinopyroxene/basalt)	0.039 to 0.067 ^a 0.043 to 0.153 ^c 0.086 to 0.153 ^d 0.0052 to 0.087 ^e	<0.01 ^a 0.0017 to 0.021 ^c 0.0086 to 0.0145 ^d
(Orthopyroxene/basalt)	0.015 to 0.0448 ^a 0.031 to 0.037 ^b 0.0158 to 0.1233 ^c	0.0017 to 0.028 ^c
(garnet/basalt)	0.0039 to 0.0174 ^a 0.0123 ^c 0.166 ^d	<0.007 ^a 0.0029 ^c 0.087 ^d
(Clinopyroxene/rhyolite)	0.048 to 0.73 ^f	0.065 to 0.158 ^f
(Garnet/rhyolite)	0.028 to 0.12 ^f	0.031 to 0.492 ^f
(Amphibole/basalt)	0.85 to 1.19 ^a 0.36 to 0.63 ^d 0.96 ^g	0.038 to 0.046 ^a 0.12 to 0.38 ^d
(Amphibole/rhyolite)	0.95 to 2.7 ^f	0.079 to 1.87 ^f
(Amphibole/fluid)	7.7 ^h	0.0049 ⁱ
(Olivine/fluid)	0.11 to 1.4 ^j	0.9 to 2×10^{-4i} 0.19 to 1.6×10^{-4j}
(Clinopyroxene/fluid)		1.4 to 2×10^{-4i}
(Garnet/fluid)	0.014 to 0.26 ^j	0.13 to 13.8×10^{-4j}
(Orthopyroxene/fluid)	0.0019 to 0.13 ^j	1.5 to 3.7×10^{-4i} 0.2 to 6.3×10^{-4j}

^aNatural system: Hauri et al. (2006). ^bCMAS and NCMS system: Beyer et al. (2012). ^cNatural system: Dalou et al. (2011). ^dNatural system: Dalou et al. (manuscript in preparation), Dalou (2011). ^eNatural and Al-free system: O'Leary et al. (2010). ^fNatural system: Van den Bleeken and Koga (submitted to EPSL). ^gNatural system: Adam and Green (2006). ^hNatural system: Wu J, Koga (2013). ⁱNatural system: Fabbriozzi et al. (2013). ^jMAS system: Bernini et al. (2012).

and EMII (0.81088 and 2.0674; Workman et al. 2004) in a plot of Pb-isotopes normalized to ²⁰⁶Pb (Figure 3). Mixing of several crustal and oceanic materials containing Pb can explain these compositions, even the range of composition of the basement rocks of NE Japan can alone explain the melt inclusion Pb-isotope compositions (see points on Figure 3). Also, Pb composition of average Pacific oceanic crust and Japan trench sediments loosely bracket the variation (Figure 3). Therefore, isotopic variations recorded in melt inclusions from Iwate volcano show a consistent interpretation. There are three potential sources of Pb identified for Iwate magma: DMM, oceanic crust, and subducted sediments. While Pb isotopes cannot decipher the presence or absence of Pb signature of the basement rock, we argue that melt inclusions hosted in olivine (the first mineral to

crystallize) would unlikely record assimilation process during magma transport through the basement.

The halogens can be used to further constrain the potential sources. F concentration in DMM is 11 ppm (Salters and Stracke 2004), which should produce primitive MORB with F average concentration around 110 ± 30 ppm (an average derived from published melt inclusion data, Figure 2), which is close to the average F in Iwate melt inclusions (i.e., 160 ppm). While F concentrations of other mantle lithologies (EMI, EMII, FOZO, HIMU) are unknown, they are not considered here since variation of Pb isotopes shows none of their presence in the source. As far as F abundance is concerned, any addition of F to the source would produce magmas that are inconsistent with that of Iwate. On the contrary, it is abundantly clear from H₂O and Cl abundances and Pb isotopes that a volatile-rich component was added from the subducted oceanic crust to the magma source.

The abundances of F in different lithologies of subducted oceanic crust should be higher than DMM, while the exact values are yet to be determined. Recent studies of serpentinization at ridges show the increase of F in peridotite sections of oceanic lithosphere up to several 100 s of ppm (Orberger et al. 1999; John et al. 2011; Debret et al. 2013). Fresh MORB contains approximately 100 ppm F, and given that F is insoluble to seawater (1.3 ppm), F content in the basalt section of the oceanic lithosphere must be at least 100 ppm and higher. Sediments are expected to have high F abundance, in the high 100 s (average continent 250 ppm F, pelagic clays 1,300 ppm F, and shale 740 ppm F; Li 1991), while the volume of subducted sediment is probably insignificant compared to that of basalt and peridotite. In any case, it is clear that the metasomatic agent must discriminate F against H₂O, Cl, and Pb.

The presence of significant serpentinite in subducted oceanic lithosphere favors the release of F during prograde metamorphism (Debret et al. 2013). This would lead to increase of F in the subarc mantle. Therefore, we conclude that there is no significant quantity of serpentinites in the subducted lithosphere below Iwate volcano. While present-day seismic tomography imaging may not be directly applicable to magma genesis that took place in the past, no significant quantity of serpentinites is indeed detected below NE Japan (Reynard et al. 2010). The presence of dry slab mantle is also identified by the observation of a strong S-wave velocity jump at the bottom subducted ocean crust detected by receiver function tomography of NE Japan (Kawakatsu and Watada 2007).

The metasomatic agent that is deprived of F must be derived from the basalt and sediment portion of the oceanic lithosphere. F partitions favorably into (dry or wet) silicate melt rather than anhydrous mantle minerals (olivine, clinopyroxene, orthopyroxene, plagioclase,

Dalou et al. 2011; Table 4). Furthermore, partial melt of hydrous oceanic crust also incorporates more F than anhydrous minerals (garnet, cpx), and shows compatibility for amphibole near unity (Van den Bleeken and Koga, personal communication about submitted manuscript). Therefore, any silicate melt generated in/from the slab would transport similar or higher concentration of F to the magma source region. Therefore, there is no silicate melt below Iwate volcano.

On the contrary, F partition coefficient between hydrous minerals and fluid favors F remaining in the mineral; therefore, aqueous fluid coming from the slab cannot transport significant amounts of F to the sub-arc mantle (Wu and Koga 2013). Because Cl systematically favors fluids over solids (Bernini et al. 2012; Fabbri et al. 2013), aqueous fluid is an ideal metasomatic agent that fractionate F from Cl. It should be noted that the salinity of the fluid significantly influences the compatibility of F and Cl. Further refinement of the model is needed to quantify the role of salinity on F/Cl fractionation. The role of supercritical fluid in the source region of Iwate volcano is indeterminable. We speculate that supercritical fluid would favor the transport of high-ionic-strength anions such as F. For Iwate volcano, as it was the case for slab melt, supercritical fluid is unnecessary for the metasomatism of its mantle source.

To summarize, the nature of the slab flux added to the Iwate magma source is an aqueous fluid (in accordance with Hanyu et al. 2006) that must be in equilibrium with hydrous minerals such as amphibole, humite group minerals, and/or mica. This indicates that the temperature at which the aqueous fluid is generated in the slab must not exceed the transition to supercritical fluid or partial melting. For example, it cannot exceed the melting temperature of sediment which is around 700°C for the lowest reported (Nichols et al. 1994). Furthermore, hydrous minerals must be present at the depths of fluid generation, to retain some of the volatile elements (F). For example, amphibole breaks down at pressure above 2.3 to 2.5 GPa (Poli 1993; Van den Bleeken and Koga, submitted) and temperature of 650°C (Poli 1993). On the basis of F concentration variations, we suggest that Iwate volcano represents the low end of the spectrum of slab surface temperatures. This low temperature primarily controls the physical nature of the fluxing agent that originates from the subducted slab and metasomatizes the sub-arc mantle.

Low slab surface temperature

Iwate slab surface temperature must be low in order to explain the presence of hydrous phases retaining F in the slab, and aqueous fluid. It must be lower than the melting point of subducted oceanic crust (650 to 700°C after Schmidt and Poli 1998). A recently developed slab

surface thermometer (Plank et al. 2009) is applied to these melt inclusions using H₂O/Ce ratio. For the melt inclusions with the highest H₂O concentrations (2.9 wt% in ITM12 and IEM35 and 3.7 wt% in IEM28.1), H₂O/Ce values predict 720, 740, and 790°C slab surface temperatures, respectively. These predicted temperatures are higher than the solidus. Vice versa, if temperatures lower than the solidus are needed to explain our data (i.e., temperature lower than 650 to 700°C, after Schmidt and Poli 1998), then the measured H₂O/Ce values of the three most water-rich melt inclusions (7,600, 6,600, and 3,600, respectively) are much lower than the predicted H₂O/Ce ratio of aqueous fluid (10⁵ to 10⁶, Plank et al. 2009). Therefore, we conclude that metasomatism of subarc mantle by aqueous fluid, is in disaccord with melt forming temperature inferred from H₂O/Ce.

It should be noted that the loss of H₂O through degassing can drive the H₂O/Ce to lower values. Similarly, crystal fractionation can increase Ce concentration. While these Iwate melt inclusions hosted in olivine are glassy and sampled from scoria, they may have somehow failed to retain the H₂O and Ce source character predicted by the thermometer of Plank et al. (2009). Geochemical indicators involving highly volatile elements, such as H₂O/Ce, may in fact be applicable to a limited number of samples that can meet stringent criteria proving non-degassing. Furthermore, the thermometer calibration is potentially limited to a narrow range of slab compositions at a 4-GPa condition. Indiscriminate application of the slab surface thermometer to arc melt inclusions would lead to an erroneous temperatures.

Competing interests

The authors declare that they have no competing interests.

Authors' contribution

ERK prepared the samples, participated to the measurements, and drafted the manuscript. KTK participated to the isotope measurements and drafted the manuscript. MH provided the scoria samples and initiated ERK's interest in Iwate and edited the manuscript. TH performed most of the electron probe measurements during his short internship as a 'L3-student' in the lab. NS and MW helped a great deal in fine-tuning the SIMS to appease our wish of precision and participated to the edition of the manuscript. All authors read and approved the final manuscript.

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