

LETTER

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Water content in arc basaltic magma in the Northeast Japan and Izu arcs: an estimate from Ca/Na partitioning between plagioclase and melt

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Abstract

The variation in water content of arc basaltic magmas in the Northeast Japan arc and the Izu arc was estimated using a simple plagioclase phenocryst hygrometer. In order to construct a plagioclase phenocryst hygrometer optimized for arc basalt magmas, we have conducted high-pressure melting experiments of relatively primitive basalt from the Miyakejima volcano, a frontal-arc volcano in the Izu arc. As a result of the experiments, we found that the Ca/Na partition coefficient between plagioclase and hydrous basaltic melt increases linearly with an increase in H₂O content in the melts. We then selected from literature geochemical data sets of relatively primitive basaltic rocks with no evidence of magma mixing and the most frequent Ca-rich plagioclase phenocrysts from 15 basaltic arc volcanoes including both frontal-arc and rear-arc volcanoes. In the 15 volcanoes studied, plagioclase phenocrysts of high anorthite content (An > 90) were commonly observed, whereas plagioclase phenocrysts in rear arc volcanoes usually had a lower anorthite content (90 > An > 80). In all volcanoes studied, the estimated H₂O content of basaltic magma was at least 3 wt.% H₂O or higher. The magmas of volcanoes located on the volcanic front have about 5 wt.% H₂O in magma whereas those from the rear-arc side are slightly lower in H₂O content.

Keywords: Island arc basalt; Water content; Plagioclase; Hydrous melting experiments; Across-arc geochemical variation

Findings

Introduction

Primary arc basalt magma is generated by partial melting of the sub-arc mantle with the addition of dehydrated aqueous fluid from the subducting slab. H₂O has profound effects on the melting temperature of the mantle, crystallization pathways of generated magmas, and the explosivity of magmas. Therefore, magmatism in an island arc along a subduction zone is controlled by H₂O released by the dehydration of a subducting slab. Precise estimation of H₂O content in arc basalt magma is important for evaluating the effect of H₂O on generation, differentiation, and eruption of magmas in subduction zones. However, estimation of pre-eruptive H₂O content on arc magmas is difficult owing to degassing from erupting magmas.

Sakuyama (1979) discussed lateral variation of the H₂O content of basalt magmas across the Northeast Japan arc. Based on the systematic differences in phenocryst mineral assemblages in andesite and dacite and the assumption that these rocks are derived from basalt magmas by fractional crystallization, Sakuyama (1979) concluded that primary magmas from a volcanic front have a lower H₂O concentration than those of a rear arc. This conclusion was further extended by Tatsumi et al. (1983) in constructing a magma genesis model for arc basalt magmas. However, this assumption is not always true because magma mixing and melting of crustal components may occur frequently to derive silicic magma. More recently, Nakamura and Iwamori (2009) estimated lateral variation of the fluid component in the source mantle based on the systematics of trace elements and isotopic compositions of arc volcanic rocks. According to their estimates, rear-arc volcanoes in central Japan are enriched in the fluid component of their mantle source, whereas the basaltic volcanoes on the volcanic front in the Izu arc are depleted in the fluid component of their source.

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In experimental research on arc basalt magmas, many studies (e.g. Sisson and Grove 1993a; Kawamoto 1996; Pichavant and Macdonald 2007; Hamada and Fujii 2008) proposed that fractional crystallization of hydrous basalts yields calc-alkaline trends whereas relatively low H₂O content in basaltic magma yields tholeiitic fractionation trends. On the contrary, recent melt inclusion studies (e.g. Sisson and Layne 1993; Newman et al. 2000; Saito et al. 2005, 2010; Ikehata et al. 2010) indicated that the H₂O content in tholeiitic basaltic melt is larger than 3 wt.% at the volcanic front.

The anorthite content of plagioclase is shown to increase with an increase in H₂O content of coexisting melt in all experimental studies (e.g., Johannes 1978; Sisson and Grove 1993a; Takagi et al. 2005; Hamada and Fujii 2007). Based on this relation, several plagioclase phenocryst hygrometers have been proposed (Housh and Luhr 1991; Putirka 2005; Hamada and Fujii 2007; Lange et al. 2009). High anorthite content of plagioclase phenocrysts (An > 90) is often reported in basaltic rocks from frontal-arc volcanoes (e.g. Kimata et al. 1995; Amma-Miyasaka and Nakagawa 2002), suggesting that magmas erupting from frontal-arc volcanoes are H₂O-rich such as >3 wt.% H₂O in basaltic melt from the Izu-Oshima volcano (Hamada and Fujii 2007). A major disadvantage of the plagioclase phenocryst hygrometers proposed thus far, however, is that they can be used only when the pressure, temperature, and compositions of coexisting plagioclase and melts are known.

In this study, we conducted high-pressure and high-temperature melting experiments on arc basalt from the Miyakejima volcano at an H₂O-undersaturated condition and at 100 to 200 MPa, which is the pressure corresponding to a shallower crustal magma chamber. In order to discuss the variation of H₂O content in arc basalts, a simple plagioclase phenocryst hygrometer is proposed. Using this simple hygrometer, the H₂O content in basalt magma occurring in the Northeast Japan and Izu arcs is estimated.

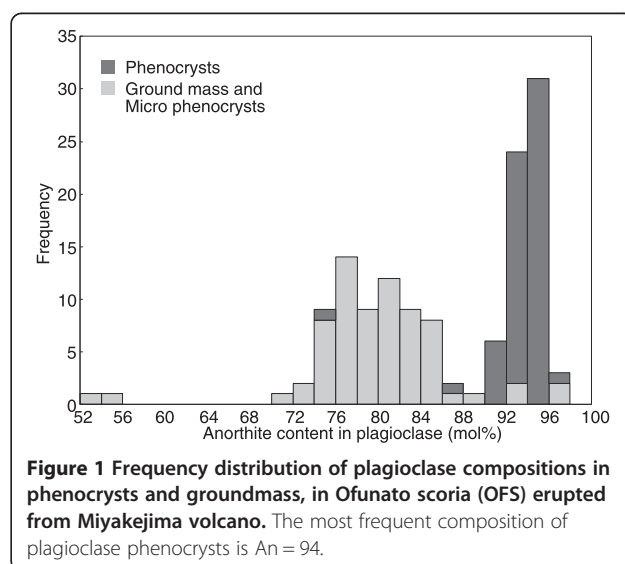
Methods

High-pressure and high-temperature experiments

Starting material In order to construct a plagioclase phenocryst hygrometer for arc basalt magma, high-pressure and high-temperature experiments were conducted on Ofunato scoria (OFS), which is a relatively primitive basalt from the Miyakejima volcano. Miyakejima is an active volcanic island located about 200 km south of Tokyo on the volcanic front of the Izu-Mariana arc. The OFS is one of the most primitive basalts to have erupted in the last 10,000 years in the Ofunato Stage 7,000 to 10,000 years ago (Tsukui et al. 2002; Niihori et al. 2003). The whole-rock composition of the OFS is as follows: 50.11 wt.% SiO₂, 0.92 wt.% TiO₂, 18.09 wt.% Al₂O₃,

11.06 wt.% FeO (total), 0.21 wt.% MnO, 5.78 wt.% MgO, 11.78 wt.% CaO, 1.85 wt.% Na₂O, 0.22 wt.% K₂O, 0.01 wt.% P₂O₅, and Mg# (=Mg/(Mg + Fe) × 100) = 48.2, which was determined through analysis of glass fused at 1 atm using an electron microprobe. OFS has 0.7 vol.% of olivine and 10.9 vol.% of plagioclase phenocrysts in which core compositions are Mg# = 78 to 82 and An (=Ca/(Ca + Na) × 100) = 90 to 96 (Figure 1). The most frequent core composition of plagioclase phenocrysts in OFS is An94. All phenocrysts have a large clear core and a narrow normal-zoned rim with a sharp boundary. The amount of magma mixing with the evolved magma is negligible. Hydrous OFS glasses with various H₂O contents were synthesized and were used as starting materials for high-pressure and high-temperature melting experiments in this study (Additional file 1: Table S1).

Experimental and analytical procedures We conducted experiments at pressures of 100, 150, and 200 MPa using SMC-2000 and SMC-5000 internally heated pressure vessels (IHPVs; Miyagi et al. 1997; Tatsumi and Suzuki 2009; Tomiya et al. 2010; Hamada et al. 2013) installed at the Magma Factory, Tokyo Institute of Technology, Tokyo, Japan. These IHPVs use argon gas as the pressurizing medium and can be pressurized up to 200 and 480 MPa, respectively. Pressure was measured by a strain-gauge pressure transducer. For SMC-2000, a single W5Re-W26Re thermocouple near the capsule was used to monitor temperature. For SMC-5000, two W5Re-W26Re thermocouples were used that were spaced 10 mm apart vertically; the observed temperature gradient across the capsule was less than 10°C. Experiments were performed in the temperature and pressure ranges of 1,050°C to 1,150°C and 100 to 200 MPa, respectively. Run durations were 12 h for experiments at



1,150°C and 24 h for the other experiments. The double-capsule method was used to buffer oxygen fugacity with a solid powder Ni-NiO (10:1, wt.) assemblage. A metal tube with a composition of either Au₇₅Pd₂₅ (2.5/2.2φ), Au₇₅Pd₂₅ (2.5/2.3φ) or Ag₅₀Pd₅₀ (2.3/2.0φ) was used as an inner sample tube that was welded on one end and weighed. Hydrous glasses synthesized at high pressure as a starting material was inserted into the inner capsule and weighed, then welded on the other end and weighed. The weight loss during welding was generally <0.00005 g. We also prepared a metal tube (Pt, 3.0/2.8φ) that was welded on one end. Ni-NiO powder and distilled water were then inserted into the tube, and the other end was only crimped rather than welded. Prepared sample capsules and a buffer capsule were placed into the outer capsule (Au₈₀Pd₂₀; 6.0/5.6φ, Au₇₅Pd₂₅; 8.0/7.8φ or Au₈₀Pd₂₀; 8.0/7.6φ), and the outer capsule was then welded. The entire capsule was kept at 110°C in an oven for 20 to 30 min and was weighed to ensure that the seal on the capsule was effective. After each run and quenching of the products, experimental charges were weighed to measure volatile loss and were punctured to verify the presence of liquid H₂O. In each buffered capsule, the presence of two phases of Ni and NiO was confirmed. The phase assemblage of the run products and compositions of minerals and glass were determined by using a JEOL JXA-8530 F electron microprobe (JEOL Ltd., Tokyo, Japan) for plagioclase; JXA-8800 was used for the others. To analyze small plagioclase 5- to 20-μm long, the acceleration voltages were 10 kV for plagioclase and 15 kV for the other phases. The beam current and time spent on each elemental peak, except for Na and the background, were 12 nA, 20 s and 10 s, respectively. Na was analyzed for 10 s (background: 5 s) without a peak search to avoid count loss. Minerals were analyzed using a focused electron beam, whereas a defocused electron beam 10 to 15 μm in diameter was employed for the glass. A loss of Na count was not detected for minerals and hydrous glasses during analyses. The H₂O content of hydrous glass as a starting material was analyzed using a 15 to 89 μm doubly polished wafer of glass and transmittance measurements of Fourier transform infrared spectroscopy (FTIR; Jasco FT/IR-6100 and IRT-5000, vacuum type, JASCO Corporation, Tokyo, Japan) with an aperture size of 100 μm. A Ge/KBr beam splitter and HgCdTe (MCT) detector were also used. The thicknesses of the doubly polished thin sections, 15 to 89 μm, were analyzed by a digital micrometer (Mitutoyo Corporation, Kanagawa, Japan) with ±1 μm precision. The dispersion of sample thickness was ±2 μm. Homogeneity of hydrous glasses was confirmed in analyses of several different points. Following the procedures of Yamashita et al. (1997), the absorbance peak heights at the 3,500 cm⁻¹ band were used to quantify the H₂O content. The density of the hydrous basaltic glass was calculated by applying

the equation of Ohlhorst et al. (2001). The H₂O content of the run products was analyzed by reflectance measurements using the same FTIR (Additional files 2 and 3). The calibration curve of H₂O in the melt versus the Δreflectance at peak near 3,650 cm⁻¹, shown in Additional file 2: Figure A1, was used to estimate the H₂O content in the hydrous glass in the run products.

Results and discussion

Experimental results and proposal for a plagioclase phenocryst hygrometer

Experimental conditions and results are listed in Table 1 and Table 2. All experiments except for C1839 were conducted near the liquidus of plagioclase (±magnetite); therefore, the composition of the melt was essentially same as the starting material. Oxygen fugacity was determined for C1839 using a plagioclase-olivine oxygen fugacity barometer (Sugawara 2001). That for C1839 was conducted at a low temperature in order to crystallize mafic minerals; the estimated *f*O₂ was ΔNNO + 0.8. Although *f*O₂ in the other run products was not estimated due to the lack of plausible mineral assemblages, the value would be close to that of NNO for three reasons. Ni and NiO coexisted in recovered buffer capsules in all runs, and similar *f*O₂ was estimated in previous works using the same double-capsule technique and similar apparatus. Moreover, the FeO content of plagioclase depends on *f*O₂ (Sugawara 2001) and the range of the FeO (total) content of plagioclase in all run products was narrow (1.3 ± 0.1 wt.%). Hamada and Fujii (2007, 2008) conducted high-pressure, high-temperature experiments using the double-capsule method with the NNO + H₂O buffer and an internally heated pressure vessel (IHPV) apparatus similar to SMC-5000 at the University of Tokyo. Hamada and Fujii (2007) estimated that the *f*O₂ condition was ΔNNO +0.8 to 2.0 by using the Co-Pd alloy redox sensor technique (Taylor et al. 1992), and Hamada and Fujii (2008) estimated that *f*O₂ was ΔNNO +1 (±0.8) by using the oxygen barometer of Sugawara (2001). We conducted experiments for a shorter duration (12 h) at 1,150°C compared to 24 h in the others in order to minimize the loss of diffusive H₂ from the capsule assemblage. Plagioclase was euhedral and rectangular and up to 15 μm. Magnetite was up to 10 μm in diameter. Both minerals were uniform in composition throughout the recovered run products, indicating that equilibrium was achieved. The H₂O content of the melt was calculated by mass balance calculation of all phases assuming that water was concentrated in only the melt. The H₂O content in the melt in run products relatively poor in crystals was measured by the reflectance infrared (IR) method (Additional files 2 and 3); the results agreed with mass balance calculations (Table 1). Figure 2a shows the relation between H₂O content in the melt and the anorthite

Table 1 Experimental conditions and results

Run#	Capsule#	Run durations (h)	T (°C)	Pressure (MPa)	Bulk H ₂ O (wt.%) ^a	H ₂ O in melt (wt.%) ^b	H ₂ O in melt (wt.%) ^c	Phase and modal proportions (wt.%) ^d	Calculated fO ₂ (ΔNNO) ^e	Relative Fe-loss (wt.%) ^f	K _D ^{pl-melt} _{Ca-Na}	An in plagioclase
HG442	C1838	12	1,150	100	0.77 (3)	0.91 (10)		Melt (85), pl (15)		-13	1.05 (17)	80.4
HG442	C1837	12	1,150	100	2.07 (9)	2.11 (13)	2.01 (19)	Melt (98), pl (2)		-6	2.21 (27)	88.4
G463	C1831	24	1,100	100	2.99 (12)	3.23 (17)	3.00 (25)	Melt (93), pl (5), mt (2)		0	3.14 (31)	91.3
G459	C1824	12	1,150	150	0.77 (3)	0.91 (10)		Melt (85), pl (15)		-11	0.93 (10)	76.9
G459	C1820	12	1,150	150	1.57 (11)	1.71 (16)	1.52 (14)	Melt (92), pl (8)		-8	1.56 (19)	84.7
G458	C1817	24	1,100	150	2.59 (9)	2.92 (10)		Melt (88), pl (10), mt (2)		-3	2.36 (33)	88.8
HG448	C1857	24	1,075	150	2.99 (12)	3.44 (13)		Melt (87), pl (10), mt (3)		1	2.78 (26)	90.2
G465	C1839	24	1,100	200	0.77 (3)	1.24 (11)		Melt (62), pl (32), ol (3), cpx (1), opx (2)	0.8	-14	1.06 (23)	79.4
G465	C1840	24	1,100	200	2.07 (9)	2.42 (10)		Melt (86), pl (12), mt (3)		5	2.11 (28)	87.7
G465	C1841	24	1,100	200	2.99 (12)	3.16 (13)	3.16 (27)	Melt (95), pl (3), mt (2)		6	2.42 (26)	89.9

^aH₂O content of starting materials analyzed by transmittance measurements using Fourier transform infrared spectroscopy (FTIR). Standard deviation is indicated in parentheses; ^bH₂O in melt calculated from mass balance. Standard deviation is indicated by parentheses; ^cH₂O in melt analyzed by Δreflectance measurements using FTIR. Standard deviation is indicated in parentheses; ^dphase proportion in weight percent indicated by parentheses. Abbreviation of phases: pl, plagioclase; mt, magnetite; ^efO₂ was calculated using the oxygen fugacity barometer (Sugawara 2001); ^fRelative Fe loss was calculated as (FeO_{after} - FeO_{starting material})/FeO_{starting material} × 100. FeO_{after} is the summation of FeO (total Fe as FeO) from all phase in run products.

Table 2 Volcanoes which were estimated by Equation 1

Volcano	Reference	Sample name	An content	Mode	CaO/Na ₂ O (bulk)	Al ₂ O ₃ /SiO ₂ (bulk)	$K_D^{pl-melt, Ca-Na}$	Estimated H ₂ O
Towada	Hunter and Blake (1995)	Zak13	91 to 94	92.5	4.71	0.36	4.74	5.6
Iwate ^a	Kuritani et al. (2014a)	VIW-18	85 to 92	92	6.11	0.36	3.41	3.9
Funagata ^a	Fujinawa (1982)	Type1	94.8		7.65	0.32	4.31	5.1
Zao	Tatsumi et al. (2008)	Za1013	80 to 96	92	4.94	0.35	4.21	5.0
Adataru	Fujinawa (1988; 1990)	2-1	78 to 95	90	4.10	0.39	3.98	4.7
Nasu (Minami-Gassan)	Ban (1991)	E-1 101408	86 to 92	89	4.58	0.35	3.20	3.7
Myoko	Sakuyama (1981)	MK278-4	80 to 94	88	3.65	0.36	3.63	4.2
Ueno	Nakano (1993, 1994)	SKS-3	82 to 87	87	3.81	0.31	3.18	3.7
Fuji	Kaneko et al. (2010)	FG1013	64 to 92	88	4.17	0.36	2.66	3.1
East-Izu	Kawamoto (1992)	Uchino	84 to 88	86	3.27	0.36	4.06	4.8
	Kikuchi and Takahashi (2004)	Takatsukayama, scoria1-2	84 to 88	87	3.84	0.37	2.53	3.7
Izu-Oshima	Ikehata et al. (2010)	O95	88 to 90	89	6.42	0.34	2.28	2.6
Niijima	Koyaguchi (1986)	WAK-10	71 to 92	91	4.28	0.34	4.28	5.1
Miyakejima	This study	OFS	90 to 96	94	6.37	0.36	4.45	5.3
	Amma-Miyasaka et al. (2005)	S-1	53 to 97	87	4.68	0.35	2.59	3.0
Hachijo-Higashiyama	Tsukui et al. (1993)	Nakanohana scoria	85 to 93	92	8.60	0.37	2.42	2.7
Sumisu-caldera	Tamura et al. (2005)	D2-R4	80 to 90	90	5.92	0.34	2.75	3.2
Torishima ^a	Tamura et al. (2007)	OP 35	95.9		10.15	0.37	4.17	4.9

^aReference had no frequency distribution of plagioclase phenocryst compositions.

content of plagioclase in our run products. In the range of experimental conditions, the anorthite content of plagioclase positively correlated with the H₂O content in melt. As shown in Figure 2a, $K_D^{Ca-Na, pl-melt}$ ($= (Ca/Na)_{pl} / (Ca/Na)_{melt}$) was proportional to the H₂O content in the melt (Figure 2b), and the effects of temperature and pressure

were not significant. A simple plagioclase hygrometer is expressed as a linear function as

$$K_D^{Ca-Na, pl-melt} = 0.74 X_{H_2O}(wt.) \times 100 + 0.36 \quad (1)$$

$$R^2 = 0.917 \quad \text{Standard error} = 0.23.$$

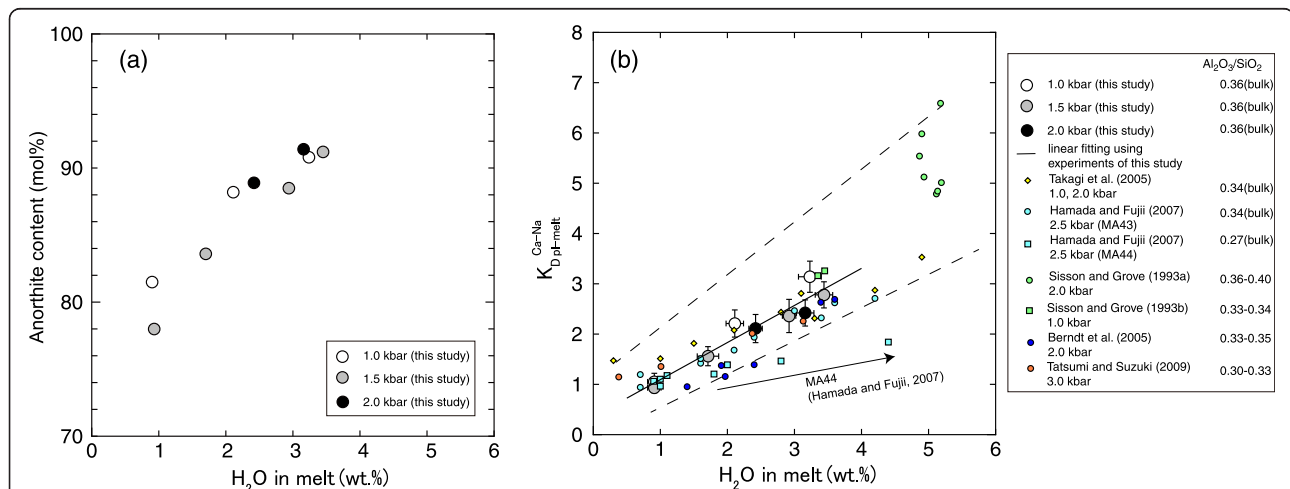


Figure 2 Relation between H₂O concentration in melt and (a) anorthite content of coexisting near-liquidus plagioclase or (b) $K_D^{Ca-Na, pl-melt}$ At 1.0, 1.5, and 2.0 kbar. (b) Dashed lines represent the range in typical basalt from the Northeast Japan and Izu arcs ($0.32 < Al_2O_3/SiO_2 < 0.39$). When the H₂O content was estimated by Equation 1, the expected error was within that indicated by the dashed lines. Source: Berndt et al. (2005) and Sisson TW et al. 1993b.

Strictly speaking, this equation is applicable for only arc basalts with compositions and pressure and temperature conditions similar to those in the present experiments. According to Hamada and Fujii (2007), $K_{D\text{ pl-melt}}^{\text{Ca-Na}}$ depends strongly on the $\text{Al}_2\text{O}_3/\text{SiO}_2$ ratio of the melt. In order to check the effect of the melt composition on $K_{D\text{ pl-melt}}^{\text{Ca-Na}}$, previous experimental data obtained at 100 to 300 MPa with various ratios of $\text{Al}_2\text{O}_3/\text{SiO}_2$ of basaltic melt were compiled, as shown in Figure 2b. Most previous experiments were conducted with melts in limited ranges of $\text{Al}_2\text{O}_3/\text{SiO}_2$ ratio ($\text{Al}_2\text{O}_3/\text{SiO}_2 = 0.31$ to 0.39). Lower $\text{Al}_2\text{O}_3/\text{SiO}_2$ ($=0.27$) experiments (MA44) by Hamada and Fujii (2007) show significantly lower $K_{D\text{ pl-melt}}^{\text{Ca-Na}}$ than that in other experiments. If Equation 1 was applied to estimate the H_2O content in relatively primitive arc basalts ($\text{Al}_2\text{O}_3/\text{SiO}_2 = 0.31$ to 0.39 ; Table 2), the error of the plagioclase phenocryst hygrometer would be approximately ± 1 wt.% H_2O (Figure 2b).

Contrary to the effect of dissolved H_2O in the melt, crystallized plagioclase becomes sodic with increasing pressure at a given melt composition (e.g. Takagi et al. 2005). Takagi et al. (2005) determined experimentally that anorthite-rich plagioclase ($\text{An} > 90$) phenocrysts can be crystallized at only a shallow-level crustal magma chamber (200 to 300 MPa) with 5 to 6 wt.% dissolved H_2O in the melt. Because H_2O solubility depends strongly on pressure, magma chambers that crystallize anorthite-rich plagioclase should not be too shallow. Therefore, we assume that the typical pressure is 200 MPa for anorthite-rich plagioclase phenocrysts in arc basaltic rocks. This assumption is supported by geophysical imaging of magma chambers beneath the Northeast Japan and the Izu arc volcanoes (e.g. Mikada et al. 1997; Murakami et al. 2001). In our experimental study, high-An plagioclase ($\text{An} = 94$) was not crystallized because the high H_2O content in the melt depressed the liquidus temperature of the plagioclase. Thus, the origin of highly Ca-rich plagioclase phenocrysts needs further experimental study.

In Equation 1, $K_{D\text{ pl-melt}}^{\text{Ca-Na}}$ was expressed as a function of only H_2O in the melt without regard to temperature and pressure. The present hygrometer can be applicable to arc basalt magmas under pressure and temperature conditions at least similar to those in present experiments (100 to 200 MPa, $1,075^\circ\text{C}$ to $1,150^\circ\text{C}$). Our starting material was slightly higher in Al_2O_3 than basaltic magmas from the Izu-Oshima volcano without plagioclase accumulation (Nakano and Yamamoto 1991). Nevertheless, our calibration can be applicable to arc basalts in a broad sense. Hamada and Fujii (2007) suggested that the Ca/Na partition between plagioclase and the melt depends on the $\text{Al}_2\text{O}_3/\text{SiO}_2$ ratio. We estimated the error of Equation 1 at ± 1 wt.% by comparison with previous experimental

data in terms of $\text{Al}_2\text{O}_3/\text{SiO}_2$ ratios; the range of arc basalts listed in Table 2 is 0.31 to 0.39.

Comparison to other hygrometers

Plagioclase phenocryst hygrometers have been proposed by many authors (Housh and Luhr 1991; Putirka 2005; Hamada and Fujii 2007; Lange et al. 2009). The results calculated by our simple hygrometer were compared with those by other hygrometers using experimental melting study data of arc basalts (see Figure 3 and Additional file 4: Table S2).

Calculation results using Equation 1 were generally higher than those reported by Lange et al. (2009). In particular, the discrepancy was large for low H_2O content. On the contrary, results using Putirka's calculations (2005) were similar to those with Equation 1. Furthermore, in order to examine various basaltic compositions, the calculated water contents based on Equation 1, Lange et al. (2009) and Putirka (2005) for previous experimental studies on alkali basalt, mid-ocean ridge basalt (MORB) and arc basalt are compared in Additional file 5: Table S3. For this calculation, experimental pressure and temperature conditions and the compositions of melt and plagioclase of the reported values were used. It is important to note that the calculated H_2O contents estimated by Equation 1 show good agreement (± 1 wt.% H_2O) with experimental values. The agreement is particularly good for low to medium K tholeiitic basalt, (i.e., those of Izu-Oshima (Hamada and Fujii 2007) and Iwate volcanoes (Takagi et al. 2005)). Considering the alkali

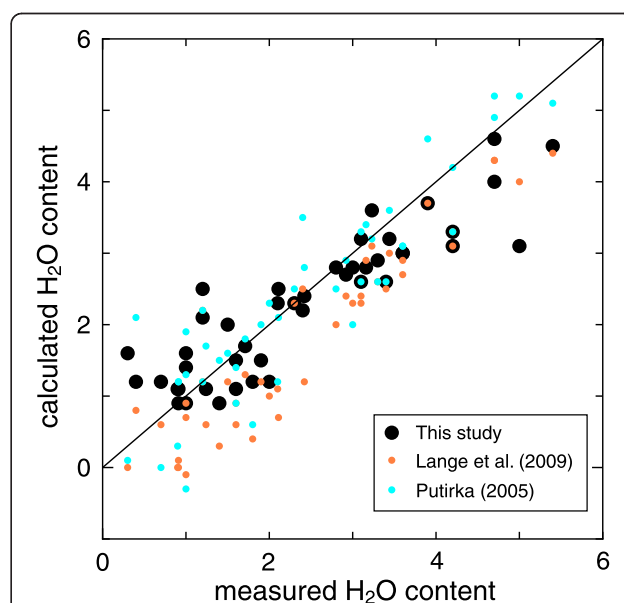


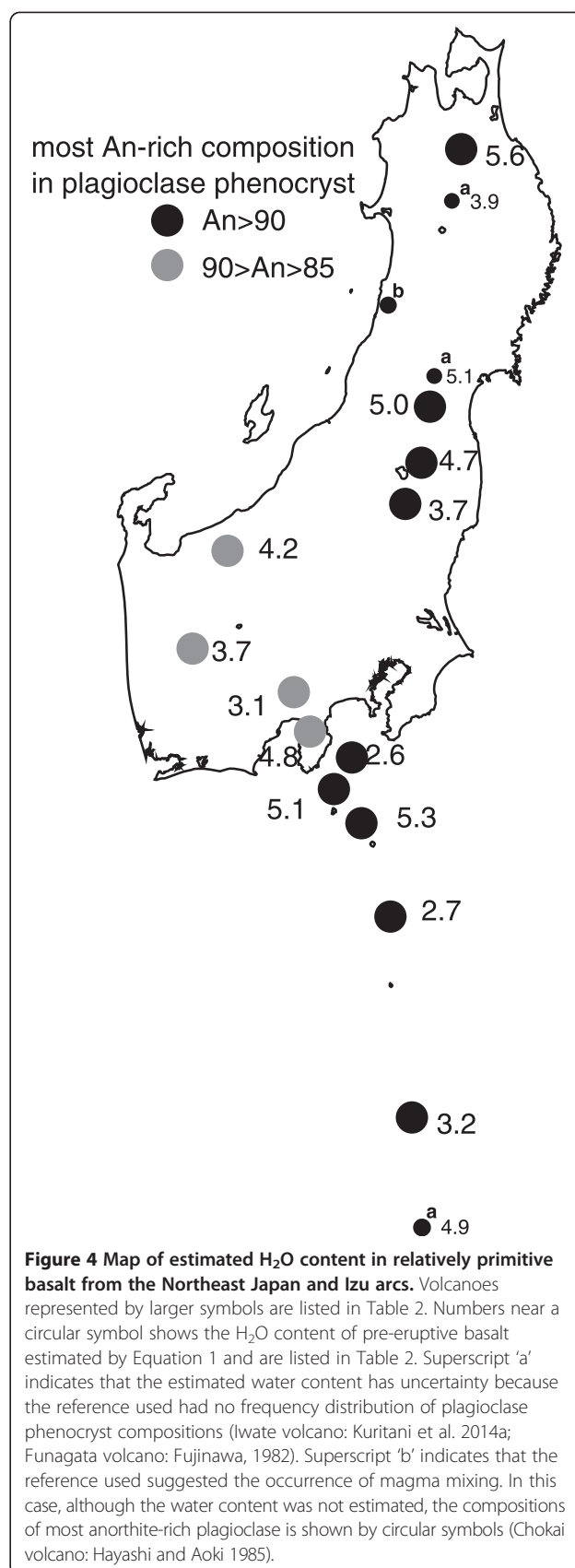
Figure 3 Relation between measured H_2O content in It and calculated H_2O content using hygrometers. Hygrometers used are of this and previous studies listed. Experimental data that was plotted in this figure is listed in Additional file 5: Table S3.

dependency noted by Honma (2012), Equation 1 may be less reliable for alkali basalts. As demonstrated in Figure 3, our simple hygrometer is useful for estimating the water content in natural magma, although its application is limited in melt composition and pressure and temperature conditions for the crystallization of arc basalts.

Pre-eruptive H₂O content in arc basalt magmas

Procedure of estimation of water content in arc basalt magma We used the bulk rock composition as the melt composition to be in equilibrium with the high-An plagioclase. Although bulk rock compositions generally do not represent melt compositions owing to selective phenocryst accumulation, trial calculations utilizing groundmass composition as the melt composition resulted in aberrant H₂O estimation of the melt and, therefore, selected the bulk rock composition to calculate the equilibrium H₂O contents of relatively primitive arc basalt magmas using Equation 1. The following discrimination rules in choosing samples were adopted: 1) Samples should come from volcanoes that have erupted basalt (SiO₂ < 53 wt.%); 2) among the basalt products, the frequency of plagioclase phenocryst composition is known and shows a unimodal compositional spectrum (Figure 1); 3) we selected the least fractionated rock that satisfies the first two constraints from each volcano peak composition of plagioclase phenocryst for the estimation of H₂O content of arc basaltic magma. If magma mixing occurred, estimation of melt composition that was equilibrated with plagioclase phenocrysts is difficult. Therefore, basalt without evidence of magma mixing had to be selected. According to Sakuyama (1981), the plagioclase composition frequency diagram is the best indicator for judging the occurrence of magma mixing.

Pre-eruptive H₂O content of arc basalt magma The estimated H₂O content of the basalt magma beneath each volcano is listed in Table 2. In magmas erupting from frontal-arc volcanoes, anorthite-rich plagioclase phenocrysts (An > 90) are commonly found. On the contrary, plagioclase phenocryst tends to be slightly sodic (An < 90) in magmas erupting from rear-arc volcanoes. We estimated the H₂O content of arc basalt magma using the most frequent composition of plagioclase phenocrysts using Equation 1 (Figure 4). The estimated H₂O content of arc basalt magmas is in the range of 3 to 5 wt.% at both frontal-arc and rear-arc volcanoes. In the rear-arc region, the H₂O content was estimated in only four volcanoes (Myoko, Ueno, Fuji, and Niiijima); therefore, systematic differences between volcanoes in a volcanic front and those in a rear arc with regard to pre-eruptive H₂O content were difficult to assess.



In some volcanoes, H₂O content in basaltic magma was estimated by melt inclusion analysis. In the case of the Fuji volcano, Yasuda (2011) analyzed melt inclusions of olivine phenocrysts in the Fuji products and estimated the H₂O content was 3.8 wt.%. Products of Takatsukayama and Sukumoyama volcanoes located in the Higashi-Izu monogenetic volcano field contain up to 3.4 wt.% H₂O in olivine phenocrysts (Nichols et al. 2012). In the case of Izu-Oshima volcano, the maximum H₂O content of melt inclusions in the olivine phenocrysts was 3.4 wt.% by analysis of products of the older Oshima group (Ikehata et al. 2010). In the case of Miyakejima volcano, Saito et al. (2005, 2010) analyzed melt inclusions of plagioclase and olivine phenocrysts from the AD 2000 eruption and estimated the H₂O content to be 3.3 wt.% in olivine-hosted melt inclusions. These analyses are consistent with the values of pre-eruptive H₂O content in melt estimated in this study.

Kuritani et al. (2014a) estimated the H₂O content of relatively primitive basalt (8.87 wt.% MgO) located at the volcanic front in Iwate volcano to be 4 to 5 wt.% based on petrologic study. Rose-Koga et al. (2014) also analyzed olivine-hosted melt inclusions from the AD 1686 products in Iwate volcano to determine a maximum H₂O content of 3.65 wt.%. These results are consistent with our new estimate. In the case of Sannomegata volcano, which is a rear-arc volcano in the Northeast Japan arc, Kuritani et al. (2014b) estimated the H₂O content of primitive basalt and source mantle and estimated H₂O content to be 6 to 7 wt.% in the primary melt. Their estimation is based on a magma genesis model with several assumptions. In contrast, our estimates of the pre-eruptive H₂O content are based on simple observation on plagioclase phenocrysts. The discrepancy between these studies can be a key issue for understanding magma differentiation processes from the upper mantle to the surface and is a topic for future investigation. Kimura and Yoshida (2006) and Kimura et al. (2010) analyzed trace element and isotope compositions of basalts in the Northeast Japan arc and the Izu-Mariana arc, respectively, and estimated the composition of primary melt, the amount of added aqueous fluid, and the H₂O content in primary melt based on forward modeling of magma genesis in a subduction zone. Their estimations of H₂O content in primary melts based on forward modeling are generally accordance with our estimates based on plagioclase phenocryst compositions. Sakuyama (1979) proposed across-arc lateral variation in H₂O content of basalt magma in the Northeast Japan arc based on systematic differences in phenocryst mineral assemblage in evolved rocks. This theory, such that magmas in the rear-arc are more enriched in H₂O than those along the volcanic front was supported by Uto (1986) and Kawamoto (1996) based on systematic changes in Al₂O₃ content during fractionation. Contrary to previous

belief, our observation showed that volcanoes on a volcanic front erupt the most H₂O-rich basalt magma (3 to 5 wt.%; Figure 4) or there is no measurable across arc difference in the water content of basalt magmas. In a detail study at Miyakejima volcano (Ushioda 2014), we showed that the water content in differentiated magmas strongly depends on the presence or absence of shallow-level magma chambers in a volcano. Therefore, comparison of phenocryst mineral assemblages in differentiated rocks (Sakuyama 1979) or that of the Al₂O₃ content among fractionation trends (Uto 1986; Kawamoto 1996) does not necessarily represent differences in water content in parental basalt magmas.

In other arc magmas, the H₂O content has been estimated by analysis of melt inclusions. For example, in the case of the Kamchatka arc, Portnyagin et al. (2007) indicated that primitive melts in a volcanic front contain an H₂O content equal to or slightly higher than those in a rear-arc. Similarly, arc basaltic melts along the Central American Volcanic Arc (Walker et al. 2003; Sadofsky et al. 2008) and the Michoacan-Guanajuato Volcanic Field of Central Mexico (Johnson et al. 2009) have slightly higher H₂O content in a volcanic front.

In subduction zones, volcanoes along the volcanic front produce the largest volume of magmas, and those from volcanoes along the rear-arc produce significantly less (Sugimura et al. 1963). Therefore, the fact that basaltic magma in a volcano on the volcanic front is H₂O-rich has fundamental importance in the consideration of mass flux of H₂O in subduction zones. Magma genesis models in subduction zones (e.g., Tatsumi et al. 1983) that assume nearly dry melting conditions beneath the volcanic front need to be reconsidered. Lateral variation of the fluid component in the source mantle based on the systematics of trace elements and isotopes reported by Nakamura and Iwamori (2009) may need reconsideration and revision since they show very small fluid addition in some volcanoes on the volcanic front.

Conclusions

In order to estimate the variation of H₂O content in relatively primitive basalt in the Northeast Japan and the Izu arcs, we constructed a simple plagioclase phenocryst hygrometer applicable for temperature and pressure of shallow crustal magma chamber beneath arc volcanoes. The H₂O content in representative basaltic rocks from 15 volcanoes in eastern Japan was estimated using our newly constructed simple hygrometer. High anorthite (An > 90) plagioclase phenocrysts were shown to be common in volcanoes along volcanic front whereas those along the rear arc were slightly lower (An > 80). We conclude that volcanoes on the volcanic front generally erupt H₂O-rich basalt magma (3 to 5 wt.%). As was suggested by Sakuyama (1979), across-arc variation in

H₂O content of basaltic magmas is not valid for pre-eruptive H₂O content of basaltic magmas inferred from plagioclase-melt equilibria.

Additional files

Additional file 1: Table S1. Synthesized conditions of hydrous Ofunato scoria (OFS) glasses as starting materials under non-buffered conditions. H₂O content of hydrous glasses was analyzed by Fourier transform infrared spectroscopy (FTIR) with transmitted light.

Additional file 2: Calibration line for reflectance infrared (IR) method. Figure A1 and Figure A2 are included. Source: Yasuda (2014).

Additional file 3: Table S4. Experimental conditions for reflectance infrared (IR) method.

Additional file 4: Table S2. Analyzed compositions of each phase in recovered run products. Using an electron probe microanalyzer (EPMA). Fe content is given as FeO* (total iron).

Additional file 5: Table S3. Comparison of the simple hygrometer (Equation 1) with previous hygrometers. Source: Parman et al. (2011).

Competing interests

The authors declare that they have no competing interests.

Authors' contributions

MU conducted the experimental petrological studies and drafted the manuscript. ET conceived of the study, participated in the design of the study, and contributed to drafting of the manuscript. MH participated in its design and coordination and contributed to drafting of the manuscript. TS contributed to performing the high-pressure experiments. All authors read and approved the final manuscript.

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