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# Persistent gas emission originating from a deep basaltic magma reservoir of an active volcano: the case of Aso volcano, Japan

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## 35 **Abstract**

36 Volcanic gas emission is considered to reflect the degassing of magma beneath  
37 volcanoes. The combined observations of gas measurement and petrological study are  
38 expected to constrain the volatile concentrations and storage depths of the pre-eruptive  
39 and primitive magma. Aso volcano (Japan) is a constantly-monitored, persistently-  
40 degassing volcano, and an ideal site to acquire gas and petrologic data. We analyzed the  
41 melt inclusions and phenocryst minerals of Holocene basaltic eruption products, and  
42 reported their major and volatile element concentrations. The samples showed abundant  
43 evidence of magma mixing, such as reverse mineral zoning, and highly variable mineral  
44 and glass compositions. SiO<sub>2</sub> measured in melt inclusions varied from 46.0 to 65.8 wt. %.  
45 High-volatile concentration, S up to 3750 ppm, were only found in mafic melt inclusions  
46 hosted by high-Fo olivine phenocrysts (~Fo82). The pre-eruptive storage depths were  
47 determined from volatile concentrations: 2 km and 4 km depth for Strombolian eruption  
48 and sub-Plinian eruption, respectively. The volatile-rich primitive magma, one  
49 endmember of the mixed-magma, originated from a deeper level (>10 km) than these  
50 magma reservoirs. Initial volatile concentrations of the primitive magma were determined  
51 using multiple constraints: >4.68 wt. % H<sub>2</sub>O, 400 - 750 ppm CO<sub>2</sub>, 3750 ppm S, 716 ppm  
52 Cl, and 324 ppm F. The observed variation of volcanic gas composition was best  
53 explained by the mixing of the gas segregated from at least a depth of 10 km, with that  
54 from the shallow reservoirs. This study illustrated the method to identify the primitive  
55 mafic magma responsible for deep volatile flux in a mature volcano with complex  
56 magmatic processes.

## 57 **Keywords**

58 Melt inclusion, Volatile element, Magma mixing, Primitive magma, Storage  
59 depth, Excess degassing.

## 60 **Introduction**

61 Arc volcanoes are known for their explosive eruptions driven by abundant  
62 dissolved volatile elements in their magma. It has been recognized that gas exsolution is  
63 the key player triggering rapid magma ascent and eruption (Roggensack *et al.* 1997; Pioli  
64 *et al.* 2009; Edmonds and Wallace 2017). Therefore, volcanic gas emission is considered  
65 to reflect the degassing of magma beneath volcanoes, and is one of the commonly  
66 observed and monitored activities, even in the quiescent period. Such persistent degassing  
67 of active volcanoes has been seen in arc volcanoes of the world (e.g. Miyakejima,  
68 Shinohara *et al.* 2003; Stromboli, Burton *et al.* 2007). Typically, volatile element  
69 behavior in magmas is governed by their solubility, and the amounts of volatile elements  
70 issued from magma are closely related to their pre-eruptive abundance. Thus, it is critical  
71 to determine the depths at which magmas were present, and their initial compositions  
72 through the studies of volcanic products. To investigate pre-eruptive volatile  
73 concentration and possibly primitive concentration, the study of melt inclusions is a

74 powerful tool since the melt is trapped in a host crystal at depth, and isolated from  
75 interaction with surrounding magma (e.g. Anderson 1973; Sisson and Layne 1993;  
76 Wallace 2005). In fact, melt inclusions are not perfectly isolated from surrounding  
77 magmas since H<sup>+</sup> can diffuse through the host olivine (e.g. Gaetani *et al.* 2012) and CO<sub>2</sub>  
78 can be redistributed in shrinkage bubbles (e.g. Tucker *et al.* 2019). Therefore, H<sub>2</sub>O and  
79 CO<sub>2</sub> measurements in melt inclusions are regarded as minimum values indicating the  
80 condition of last equilibration. However, several studies have pointed out discrepancies  
81 between total masses of emitted gas measurements and melt inclusion-estimation of  
82 dissolved gas in glasses (e.g. Wallace 2005; Shinohara 2008). This is commonly referred  
83 to as “excess degassing”, and is observed in subduction zone volcanism (e.g. Métrich and  
84 Wallace 2008; Roberge *et al.* 2009; Wallace and Edmonds 2011; de Moor *et al.* 2013). It  
85 appears necessary to reconcile the melt inclusion-based volatile budget with the surface  
86 observation and this is the task we want to tackle in this study.

87 Aso volcano (Japan) is the ideal study site to compare gas emissions and  
88 petrological samples, including melt inclusions. It is a constantly monitored persistent  
89 degassing volcano since the 1970s, emitting more than 100 - 200 tons of SO<sub>2</sub> a day, even  
90 during the quiescent period (e.g. Mori *et al.* 2013; Shinohara *et al.* 2018; Japan  
91 Meteorological Agency (JMA) 2020). In addition, Holocene eruptions from Aso  
92 produced olivine-bearing tephra samples, ideal for finding quenched melt inclusions. For  
93 example, a recent melt inclusion study of historical Aso eruption products reported a  
94 shallow storage depth of erupted magma and a large excess of SO<sub>2</sub> degassing from Aso  
95 volcano (Saito *et al.* 2018). However, this previous study did not report volatile element  
96 concentrations of a primitive magma. Moreover, geophysical studies revealed multiple  
97 magma reservoirs with depths down to 24 km (below sea level: bsl) (Sudo and Kong  
98 2001; Sudo *et al.* 2006; Abe *et al.* 2010; Hata *et al.* 2016).

99 In this study, we analyzed the bulk tephra of less evolved erupted products,  
100 matrix glasses, melt inclusions (glass and host minerals) to obtain information on the pre-  
101 eruptive magma process, storage depth and characteristics of primitive magma of Aso  
102 volcano. This paper reports the first petrological description of the deep magma reservoirs  
103 of Aso volcano using melt inclusion data and corroborating gas composition model. Our  
104 petrological constraints on a magma plumbing system give us the primitive H<sub>2</sub>O, CO<sub>2</sub>, F,  
105 Cl, and S concentrations of Aso, a persistently-degassing active volcano.

## 106 **Samples and methods**

### 107 ***Geological setting of Aso volcano and its cones***

108 Aso volcano, located in central Kyushu Island of the Southwest Japan Arc, is  
109 one of the most active volcanoes of the volcanic front related to the Philippine sea plate  
110 subduction (Nakada and Kamata 1991; Kamata 1998; Miyoshi *et al.* 2008a, b). It has  
111 been active for at least 0.8 million years (e.g. Watanabe *et al.* 1989). Its activity is  
112 characterized by two different types of volcanisms: caldera-forming gigantic pyroclastic-  
113 flow eruptions (from 270 ka to 89 ka), and post-caldera extrusive activities (<89 ka) (Ono

114 and Watanabe 1985). In the post-caldera stage, various types of magma (basalt to rhyolite)  
115 were erupted from, at least, 17 observable vents in the caldera (Watanabe 2001; Miyoshi  
116 *et al.* 2005).

117 Holocene activities are well described in tephrostratigraphic studies  
118 (Miyabuchi and Watanabe 1997; Miyabuchi 2009, 2010, 2017). The tephra is mostly  
119 basaltic and limited to the ejections from central cones. The last silicic eruption deposited  
120 Aso central cone pumice 1 (ACP1; Takada 1989) at 4.0 ka (Miyabuchi and Watanabe  
121 1997; Hirata *et al.* 2020). ACP1 is the only silicic product in Holocene, which is dacitic  
122 banded pumice related to the effusion of Akamizu andesite lava flow (Miyabuchi 2017).  
123 Subsequently, volcanic activity of Kishimadake (3.7 ka), Ojodake (3.5 ka), Komezuka  
124 and Kamikomezuka cones (3.0 ka) was derived from basaltic to basaltic andesite magmas  
125 (Fig. 1; Miyabuchi and Watanabe 1997; Miyabuchi 2010; Hirata *et al.* 2020). Nakadake  
126 cone is formed in three stages: old volcanic (22 – 7.3 ka), young volcanic (7.3 – 3.7 ka)  
127 and youngest pyroclastic stages (<3.7 ka; Ono and Watanabe 1985; Miyabuchi 2009).  
128 Today, Nakadake is the only active central cone since the last eruption of Kamikomezuka  
129 (*ca.* 3.0 ka). All basaltic rocks in the post-caldera stage of Aso volcano evolved to low-  
130 Mg high-alumina basalt (MgO < 6.1 wt.%; Kuno 1960; Sisson and Grove 1993).

131 The magma chambers beneath Aso caldera have been described by geophysical  
132 surveys. Abe *et al.* (2010) reported a large low-velocity layer (LVL) from a depth of 11  
133 to 25 km (for consistency hereafter, all the depths in the paper are referred as depths from  
134 the vent of the edifice, adding 1 km to the depth below sea level), by receiver function  
135 tomography. Hata *et al.* (2016) identified the magma pathway from 21 km deep beneath  
136 the caldera by electromagnetic survey, and reported two anomaly centers (C1 and C2)  
137 separated by 3 km in horizontal distance. C1 is located 5 km beneath Kishimadake cone,  
138 which corresponds to the main magma chamber feeding present-day Nakadake eruptions.  
139 This is in good agreement with earlier studies that reported the C1 chamber between 4  
140 and 10 km depth, by deformation analysis of the volcano (Sudo *et al.* 2006) and seismic  
141 low-velocity anomaly (Sudo and Kong 2001). C2 is located 3 km beneath Nakadake cone.  
142 However, this C2 anomaly is geographically offsetted from the crater of Nakadake, and  
143 instead, a crack-like conduit extending from 0.3 to 2.8 km. A continuing passage was  
144 identified beneath this cracked-conduit, based on a region of lack of seismic reflectors at  
145 a depth from 2.5 to 4.5 km beneath the crater based on a 3-D seismic reflection analysis  
146 (Tsutsui and Sudo 2004). In this paper, we call this shallow magma passage as C2.

### 147 148 149 ***Tephra and scoria descriptions***

150 We collected tephra deposits of eruptions from Kishimadake (*ca.* 3.7 ka),  
151 Ojodake (*ca.* 3.5 ka), Kamikomezuka (*ca.* 3.0 ka) and Nakadake (the youngest pyroclastic  
152 cone; <3.7 ka). Tephra samples were specifically collected to avoid the diffusive volatile-  
153 loss and daughter mineral crystallization within melt inclusions (Danyushevsky *et al.*  
154 2002; Lloyd *et al.* 2013). Furthermore, we focused on recent (3.7 ka to present) eruption

155 activities of basalt to basaltic andesite composition involving olivine phenocrysts. The  
156 sample set was therefore used to obtain information about crustal magma evolution and  
157 the volatile characteristics of magmas. All tephra samples contain phenocrysts of  
158 plagioclase, olivine, clinopyroxene, orthopyroxene, and opaque minerals. Detailed  
159 descriptions of these tephra are found in Miyabuchi (2009, 2010).

160 Five tephra samples used in this study correspond to the eruptions of four cones:  
161 Kishimadake scoria (KSS), Ojodake scoria (OJSU and OJSL), Kamikomezuka scoria  
162 (KKO), and Nakadake scoria (NKD14). Specifically, KSS, OJSU, and OJSL were  
163 collected from a tephra deposit site (A9418 section reported by Watanabe 1991;  
164 Miyabuchi and Watanabe 1997; Fig. 1). KSS was collected from the lowest unit of sub-  
165 Plinian scoria-fall deposits in the N6 stage. It is a vesicular basaltic black scoria with a  
166 maximum size of 4.8 cm (Miyabuchi and Watanabe 1997). OJSU and OJSL were  
167 collected from the uppermost and lowest unit of sub-Plinian scoria-fall deposits in the N4  
168 stage, respectively. OJSU and OJSL are reddish-brown, weakly altered basaltic scoria,  
169 with a maximum size of 4.6 cm (Miyabuchi and Watanabe 1997). These deposits  
170 represent sub-Plinian eruption events, which are the biggest eruption events in Holocene  
171 (each with approximately 0.06 km<sup>3</sup>, VEI 3, Miyabuchi 2009). KKO was collected from  
172 Kamikomezuka cone, because the tephra deposit away from the edifice has not been  
173 identified presently. We sampled the black scoria from the outer part of the edifice from  
174 a road-cut outcrop, avoiding oxidized reddish scoria. KKO is poorly sorted, non-welded,  
175 vesicular scoria, ranging from cm-size to cow-dung bomb (tens of cm). NKD14 was  
176 collected from the crater rim of Nakadake cone's first crater, immediately after the  
177 eruption of Nov. 27, 2014. The samples were cm-size, well-vesiculated, black scoria. The  
178 eruption of Nakadake cone in the period 2014 - 2015 is the first magmatic eruption in 24  
179 years (Ikebe *et al.* 2008; JMA 2016). In the most violent phase, Strombolian eruptions  
180 occurred with ejections of scoria lapilli and bombs at Nakadake cone's first crater (Yokoo  
181 and Miyabuchi 2015).

### 182 ***Melt inclusion preparation***

183 These 5 tephra samples were used for petrological observation and chemical  
184 analysis (*i.e.* bulk rock, phenocrysts and melt inclusions). Samples were washed in an  
185 ultrasonic bath, and only lapilli-size scoria (up to 6 cm) were chosen. Five grams of each  
186 sample were powdered using a ball milling machine with an alumina cup and a ball, for  
187 bulk rock X-ray fluorescence analysis (XRF). Olivine, pyroxene, and plagioclase crystals  
188 were handpicked under an optical microscope from scoria crushed by hand and sieved  
189 (from 0.25 to 1 mm). The picked crystals were mounted in resin and polished until the  
190 melt inclusion was exposed. They were then measured using an electron probe micro-  
191 analyzer (EPMA) and a reflectance Fourier transform infrared spectroscopy (FTIR).  
192 Selected olivine crystals hosting melt inclusions of KSS and NKD14 were removed from  
193 the resin after EPMA and FTIR analyses, then mounted together in indium for secondary  
194 ion mass spectrometry (SIMS).  
195

196

197 *Analytical methods*

198 Bulk rock major and trace elements

199 Bulk rock major and trace elements were determined by XRF on flux-fused  
200 disks using a Philips PANalytical MagiX PRO spectrometer at the Kitakyushu Museum  
201 of Natural History and Human History. The detailed analytical procedures are described  
202 by Mori and Mashima (Mori and Mashima 2005). Accuracy was reported as  $\pm 0.1\%$   
203 relative for  $\text{SiO}_2$  and for trace elements, and varied from 5 to 25 ppm (Table 1).

204 Major and volatile elements in melt inclusion, mineral and glass

205 Major elements, S, and Cl concentrations were determined in melt inclusions,  
206 host minerals and groundmass glasses using a JXA8800R electron probe microanalyzer  
207 at the Earthquake Research Institute (ERI), University of Tokyo. The analytical settings  
208 were 15kV acceleration voltage, 12 nA beam current, with counting times for Na, Al, K,  
209 Fe, Mg, Si, Ti, V, Mn, and Ca at 20 s, and 30 s for Ni, Cr in mafic minerals and for S and  
210 Cl in hydrous melt. Beam diameter was set at 10  $\mu\text{m}$  for glass and plagioclase, and  
211 focused (1  $\mu\text{m}$ ) for other minerals. During melt inclusion and groundmass glass analysis,  
212 Na and K were always measured in the first analytical cycle to minimize alkali loss  
213 (Devine *et al.* 1995). All analyses applied oxide ZAF correction. Analytical uncertainties  
214 are  $<0.7\%$  relative for Si,  $<1\%$  relative for Al, Fe, Mg, and Ca,  $<5\%$  relative for Ti and  
215 Na,  $<10\%$  relative for K, and  $<20\%$  relative for Mn, V, Ni, Cr, S, and Cl.

216 In addition to the EPMA analysis, the core and rim compositions of phenocrysts  
217 were analyzed by energy-dispersive X-ray spectroscopy (EDS, AZtec system; Oxford  
218 Instruments) connected to a JEOL JSM-7001F field-emission electron microscope (FE-  
219 SEM) at Kumamoto University, using 15 kV acceleration voltage and 1 nA beam current.  
220 The beam diameter settings were the same as the EPMA setting at the University of Tokyo  
221 for all the host minerals. Analytical uncertainties are typically 0.5% relative for  $\text{SiO}_2$ , 1%  
222 relative for  $\text{Al}_2\text{O}_3$ , 1.5% relative for FeO, 0.5% relative for MgO, 2% relative for  $\text{TiO}_2$   
223 and  $\text{Na}_2\text{O}$ , 1.5% relative for  $\text{K}_2\text{O}$ , 1% relative for CaO. Other minor elements such as  
224 MnO and  $\text{P}_2\text{O}_5$  can be as high as 20% relative.

225 Water concentrations in melt inclusions were determined by FTIR micro-  
226 reflectance spectroscopy using a JASCO FT-IR-660 plus, equipped with an IRT-30VC  
227 analytical microscope at the ERI, following the procedures described by Yasuda (2014).  
228 The IR spectra were obtained using  $15 \times 15$  to  $60 \times 60 \mu\text{m}^2$  rectangular apertures, and by  
229 accumulating 220 to 1500 scans, over a range of 400 to  $7800 \text{ cm}^{-1}$ . A gold mirror was  
230 used as a reflectance reference. Water concentrations were quantified using an empirical  
231 linear relationship for basalt to rhyolite compositions for total  $\text{H}_2\text{O}$  concentrations,  
232 measured in the  $3650 \text{ cm}^{-1}$  wavelength region. The analytical uncertainty ( $2\sigma$ ) of the FTIR  
233 reflectance spectroscopy was  $< 0.3 \text{ wt. } \%$  (Yasuda 2014). While the detection limit of the

234 method strongly depended on the sample, we estimated the detection limits as 0.5 wt. %.  
235 H<sub>2</sub>O, CO<sub>2</sub>, F, S, and Cl concentrations in selected melt inclusions were  
236 determined by a SIMS (Cameca IMS-1280HR of Kochi Institute, JAMSTEC, Japan)  
237 following the procedure described by Shimizu *et al.* (2017). This analytical method favors  
238 a weaker primary current (up to 0.5 nA) than in previous studies (1-1.5 nA primary  
239 current; Le Voyer *et al.* 2010; Helo *et al.* 2011; Rose-Koga *et al.* 2012, 2014; 20 nA  
240 primary current in Hauri *et al.* 2002) and uses in-house standards covering the  
241 concentration range found in the natural samples in this study. In short, a 20 keV (10 keV  
242 at the ion source and 10 keV at the sample surface) Cs<sup>+</sup> ion beam of 300–500 pA was  
243 defocused to be 10-15 μm in diameter. Secondary ions were accelerated at 10 kV. A -10  
244 keV electron beam with a diameter of ~100 μm was applied for electrostatic charge  
245 compensation of the analysis area. The field aperture was set at the size corresponding to  
246 5 x 5 μm on the sample surface. Mass resolving power was set at ~6000 to separate mass  
247 interferences (for example, to separate <sup>34</sup>S<sup>1</sup>H interference on <sup>35</sup>Cl requires 5120 MRP;  
248 Burdo and Morrison 1971). Negative secondary ions of <sup>12</sup>C, <sup>16</sup>OH, <sup>19</sup>F, <sup>30</sup>Si, <sup>31</sup>P, <sup>32</sup>S, and  
249 <sup>35</sup>Cl and the mass position of 11.9 amu were measured by an axial electron multiplier  
250 using the peak switching method. An analysis consisted of 10 cycles, and the total  
251 measurement time for each analysis was ~6 min. Repeated analysis of a secondary  
252 basaltic glass standard from East Pacific Rise, EPR-G3 (Shimizu *et al.* 2017), yielded a  
253 relative standard deviation (1σ) for H<sub>2</sub>O, CO<sub>2</sub>, F, Cl, and S of 1.4, 3.2, 1.5, 2.5, and 0.9%,  
254 respectively. This SIMS analysis was conducted after EPMA analysis. Therefore,  
255 although we were careful to polish the sample with alumina powder again before SIMS  
256 analysis, measured CO<sub>2</sub> concentrations were reported in the supplementary material but  
257 not used due to possible carbon contamination. The measured values for S and Cl agreed  
258 within 25% between EPMA and SIMS analysis. So when volatile elements were  
259 measured by two methods, we adopted SIMS values rather than EPMA because analytical  
260 uncertainty of these elements by SIMS are generally lower (e.g. Rose-Koga *et al.* 2020).  
261 In this study, we did not find satisfactory agreement between H<sub>2</sub>O determined by FTIR  
262 and by SIMS (uncertainty of FTIR measurements is close to 40%) so in the following,  
263 we only consider H<sub>2</sub>O concentrations measured by SIMS, based on the better detection  
264 limit and precision of the method.

### 265 Sulfur speciation analysis

266 Selected inclusions were analyzed for SKα peak positions to constrain the *f*O<sub>2</sub>  
267 conditions of the glasses. The wavelength of SKα radiation [ $\lambda$ (SKα)] for melt inclusions  
268 was measured using the EPMA following the procedures described by Yasuda *et al.*  
269 (2001), based on the method of Wallace and Carmichael (1994) and Carroll and  
270 Rutherford (1988). Sulfur speciation as the proportion of S<sup>6+</sup> over a total S content was  
271 determined by measuring a relative shift from the peak position of an anhydrite mineral  
272 (Carroll and Rutherford 1988), with an assumption that S<sup>2-</sup> and S<sup>6+</sup> are the only two



273 relevant species in silicate melt (Jugo *et al.* 2010). To avoid sulfur oxidation due to  
274 prolonged beam exposure (*i.e.* Rowe *et al.* 2007), we conducted a measurement with  
275 multiple spots for each melt inclusion. Therefore, we added up the wavelength scan data  
276 of individual spots to determine the precise peak position. A Gaussian curve fitting was  
277 used for sulfur peak deconvolution. Analytical uncertainty ranges from 5 to 10% relative  
278 (Table S1).

279

### 280 **Data processing: Post-entrapment crystallization correction**

281 Major and volatile element data for all melt inclusions hosted in olivine were  
282 corrected for the effects of post-entrapment crystallization (PEC; e.g. Danyushevsky *et al.*  
283 *et al.* 2000) by incremental calculation of equilibrium olivine (0.1 wt. % step) adding into  
284 the residual melt, until the melt reaching equilibrium with host olivine (Toplis 2005),  
285 following the procedures described in Danyushevsky *et al.* (2000). The melt  $\text{Fe}^{2+}/\text{Fe}^{\text{Total}}$   
286 ratios were calculated from the empirical equation of Kilinc *et al.* (1983), assuming a  
287 constant  $fO_2$  ( $\Delta\text{FMQ} + 1.0$ ) based on measured  $SK\alpha$  peak shifts of all eruptions (Jugo *et al.*  
288 *et al.* 2010). Temperatures of olivine-melt equilibrium were calculated using the olivine-  
289 saturated melt thermometer of Sugawara (2000), corrected for the effect of water on  
290 olivine liquidus temperature according to Médard and Grove (2008). We assumed an  
291 average  $\text{H}_2\text{O}$  concentration of NKD14 and KSS analyzed by SIMS for melt inclusions  
292 that were not analyzed individually for  $\text{H}_2\text{O}$ . Note,  $K_D$  and temperature were recalculated  
293 at each increment of olivine addition. The melt inclusions were not corrected for post-  
294 entrapment diffusive Fe-loss (Danyushevsky *et al.* 2000), as total FeO concentrations in  
295 melt inclusions are either the same or higher than those in bulk rocks. Corrected volatile  
296 concentrations were adjusted assuming volatiles are perfectly incompatible to host  
297 minerals and corrected values are used in the figures and reported in Table 2 (raw  
298 uncorrected data in supplementary material S1).

299 Plagioclase-, clinopyroxene-, and orthopyroxene-hosted melt inclusions were  
300 not corrected for PEC because there is no universally accepted procedure, although some  
301 attempts have been reported (Yasuda *et al.* 2001; Neave *et al.* 2017; Hartley *et al.* 2018).  
302 Among these samples, only the melt inclusions in equilibrium with host minerals were  
303 used for the magmatic temperature calculation, in which the exchange coefficients were  
304 within expected range of basaltic composition:  $(K_D(\text{Fe-Mg})_{\text{cpx-liq}} = 0.28 \pm 0.08$ ,  $K_D(\text{Fe-}$   
305  $\text{Mg})_{\text{opx-liq}} = 0.29 \pm 0.06$ , and  $K_D(\text{An-Ab})_{\text{pl-liq}} = 0.27 \pm 0.01$  or  $0.1 \pm 0.05$  (depending on  
306 the calculated temperature; Putirka 2008).

## 307 **Results**

308 The analysis of five scoria from Aso edifices produced data of major elements  
309 for bulk rocks along with 204 melt inclusion data (major and volatile element  
310 concentrations) and host mineral compositions. More than 890 point-analyses were made  
311 to determine major element compositions of groundmass glasses and phenocrysts (core  
312 and rim; Tables 1 – 3 and S1 – S3).

313

314 ***Petrography***

315 All samples were porphyritic with approximately 40 vol.% crystals. Plagioclase,  
316 clinopyroxene, olivine plus minor orthopyroxene, and opaque minerals were present. In  
317 all samples, plagioclase phenocrysts commonly showed dusty zone and honeycomb  
318 texture (Fig. 2a), and all orthopyroxene phenocrysts had reaction rims of olivine and  
319 clinopyroxene (Fig. 2b). Aggregates of phenocrysts (i.e. plagioclase, clinopyroxene,  
320 olivine, and opaque minerals) were frequently found in all samples (Fig. 2c). The  
321 groundmass consisted of microlites of plagioclase, clinopyroxene, olivine, and magnetite.  
322 The groundmass of KSS occasionally showed the heterogeneous mingling texture where  
323 crystals are relatively abundant (Fig. 2d).

324

325 ***Host mineral compositions***

326 The olivine phenocrysts were grouped into two types based on the core Fo  
327 contents [ $\text{Mg} / (\text{Fe} + \text{Mg}) \times 100$  in mole], as low-Fo (62 to 72) and high-Fo (72 to 82)  
328 (Fig. 3a). High-Fo olivine was observed only in KKO, OJSU, OJSL, and KSS. Low-Fo  
329 olivine was found in all samples. As Fo content of the phenocryst rims ranged from 68 to  
330 78 in KKO, OJSU, OJSL, and KSS, and from 64 to 67 in NKD14 (Table S3), generally  
331 low-Fo olivine phenocrysts were reversely zoned, whereas olivine phenocrysts in NKD14  
332 were homogeneous. All high-Fo olivine phenocrysts were normally zoned.  
333 Clinopyroxene and orthopyroxene phenocryst cores in all samples were in the range of  
334 65-76 and 61-71, respectively ( $\text{Mg\#} = [\text{Mg} / (\text{Fe} + \text{Mg}) \times 100$  in mole]). The majority of  
335 clinopyroxene phenocrysts in KKO, OJSU, OJSL, and KSS were reversely zoned. Mg#  
336 of NKD14 varied little, at ~70. The compositions in the rims of olivine and clinopyroxene  
337 phenocrysts were the same as the minerals found in reaction rims of orthopyroxene  
338 phenocrysts. The plagioclase phenocrysts were also divided into two types based on the  
339 normal and reverse zoning patterns. The An content [ $\text{Ca} / (\text{Ca} + \text{Na}) \times 100$  in mole] of  
340 the plagioclase phenocryst cores was within the 55-93 range.

341

342 ***Major and volatile elements in melt inclusions***

343 The tephra samples of this study were basaltic to basaltic andesite with  $\text{SiO}_2$   
344 ranging from 51.5 to 54.3 wt. % (large circles in Fig. 3c). Melt composition in inclusions  
345 varied significantly more than that of the bulk tephra composition:  $\text{SiO}_2$  ranging from  
346 46.0 to 65.8 wt. %. This range mostly overlapped with the compositional variation of  
347 post-caldera volcanic products (grey circles in Fig. 3c). The melt inclusion compositions  
348 of NKD14 were generally similar to evolved matrix glass and varied little, and were  
349 distinguished from the melt inclusions of KKO, OJSU, OJSL, and KSS.

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351 In our sample set, the core composition of host olivines were clearly divided  
352 into two groups with Fo72 representing the divide (Fig. 3a). We interpreted that olivines  
353 have grown from two distinctively different lavas, which we called mafic and felsic.  
Based on the anti-correlation of  $\text{SiO}_2$  concentration in olivine-hosted melt inclusions with

354 host Fo content, we concluded that magma composition can be divided into two groups  
355 above and below 55 wt.% SiO<sub>2</sub>. The high SiO<sub>2</sub> corresponded to low-Fo olivine and low  
356 SiO<sub>2</sub> corresponded to high-Fo olivine. Thus, in the following we grouped all the melt  
357 inclusions lower than 55 wt.% SiO<sub>2</sub> in a mafic group, and others in a felsic group. S  
358 concentration in melt inclusions correlates well with host Fo content and is anti-correlated  
359 with SiO<sub>2</sub> (Fig. 3a) and K<sub>2</sub>O, indicating a mafic volatile-rich (with S up to 3750 ppm and  
360 Cl up to 1311 ppm) magma, and a felsic volatile-poor magma (Fig. 3b). NKD14  
361 inclusions were notably different, with less than 434 ppm S, indicating extensive  
362 degassing. Melt inclusion Cl concentrations varied from 530 ppm to 1311 ppm (Table 2  
363 and Table S1; SIMS value is preferred to EMP value when both are reported). F  
364 concentrations were determined only on a subset of olivine-hosted melt inclusions and  
365 therefore there are fewer data. F concentrations varied between 258 and 853 ppm (Table  
366 2 and Table S1). F and Cl correlates with SiO<sub>2</sub> and K<sub>2</sub>O and anti-correlate with host Fo  
367 content. H<sub>2</sub>O concentrations measured in melt inclusions vary between 0.47 to 2.89 wt.%  
368 (NKD14-Ol-e3-4-m4 and KSS-Olivine-4-m11, respectively; Table 2 and Table S1).

## 369 **Discussion**

### 370 *Evidence for magma mixing*

371 Zoning in magmatic minerals characterizes their crystallization in a magma  
372 chamber and can trace the history of magma cooling (e.g. Costa *et al.* 2008), and magma  
373 mixing (e.g. Sakuyama 1979). Among the tephra samples in this study, phenocrysts in  
374 every sample indicate textural evidence of magma mixing that could have been produced  
375 by the introduction of hotter magma: for example, the reverse zoning and disequilibrium  
376 texture, such as the honey-comb texture of plagioclase and reaction rim of orthopyroxene  
377 (Fig. 2a and 2b; Tsuchiyama 1985). The coexistence of normally zoned plagioclase and  
378 olivine phenocrysts with these disequilibrium phenocrysts suggests that magma mixing  
379 of at least two distinct components has occurred (e.g. Sakuyama 1979). This  
380 interpretation is supported by the compositional variation of the melt inclusions; felsic  
381 melt inclusions are hosted in reversely zoned phenocrysts such as plagioclase,  
382 clinopyroxene, orthopyroxene, and low-Fo olivine. Mafic melt inclusions are hosted in  
383 high-Fo olivine and some of plagioclase (Fig. 3).

384 The presence of orthopyroxenes surrounded by reaction rims and An-rich zones  
385 in the middle part of plagioclases in NKD14 (Fig. 2f) indicates that the 2014 magma is  
386 also the result of mixing. Such petrological features are found in magmatic products of  
387 every Nakadake eruption (Miyoshi *et al.* 2005). Thus, we consider that all the Nakadake  
388 eruption products are the result of magma mixing, rather than the derivatives from a single  
389 parental magma. In addition, Miyoshi *et al.* (2005) also show that the compositional  
390 variation in the trace elements in the post-caldera basaltic rocks of Aso volcano is  
391 consistent with a magma mixing model and not with a fractional crystallization process.  
392 Therefore, at least two types of magmas are present beneath the Nakadake cone, and feed  
393 its eruption.

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395 ***Characterization of mixing endmembers***

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We conducted a two-component mixing model calculation of two distinct magmas, one silicic endmember and one mafic endmember, based on the major element variation of melt inclusions (see Supplementary document S4). The model used the most primitive basaltic melt (Melt ID: 2-m1 hosted in an olivine) and the most SiO<sub>2</sub>-rich dacitic melt (Melt ID: d2-m1 hosted in a plagioclase) of KSS as the mafic and silicic endmember, respectively. The major element variation was well reproduced with the mixing model for melt inclusions of all host minerals (Fig. S4-5). While the major variance of concentration variations was explained by a simple mixing process, in close inspection of trends, it is likely that crystal fractionation contributed to the dispersion from the mixing model. It should also be noted that the mixing model required the presence of independent mixing endmembers, it does not constrain their origin.

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It is important to note that there is a surface expression of this silicic endmember in the Aso eruption products, while the mixing endmember is set by a melt inclusion. Major element compositions of ACP1 dacitic pumice (Takada 1989), the only Holocene felsic product erupted three hundred years before that of KSS, are similar to the endmember, and this indicates the presence of the silicic magma. Furthermore, the presence of a banded pumice was reported in ACP1 prior to KSS (Miyabuchi 2017). This banded texture is evidence of magma mingling and therefore the mixing trend is unlikely a result of assimilation and crystal fractionation of single parental magma. The cores of reversely zoned phenocrysts of KSS scoria samples were formed at equilibrium conditions with the silicic endmember. Therefore, the temperature of silicic endmember magma was determined with a two-pyroxene thermometer by pairing core compositions (Putirka 2008). The estimated temperatures of the silicic endmember are 1010-1025°C for KKO, OJSU, OJSL, and KSS ( $\pm 13^\circ\text{C}$ ,  $1\sigma$  for samples, while standard error of the thermometer is  $\pm 38^\circ\text{C}$ ). We adopted this estimated range as the temperature of the silicic endmember magma. These estimated temperatures are higher than those of typical dacitic magma with 4 - 5 wt. % H<sub>2</sub>O (e.g. 770 – 915 °C for Mount St. Helens, Gardner *et al.* 1995) and lower than that estimated for a completely anhydrous dacitic magmas, for example Puna Geothermal Venture Wellfield, Hawaii have the highest temperature estimates 1050°C (Teplow *et al.* 2009). This silicic endmember magma (T between 1010-1025°C) will mix with the mafic endmember (most likely hotter) magma at temperature presumably higher than 1010-1025°C.

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Basaltic lava corresponding to the pure mafic endmember is absent among the eruption products of the entire post caldera stage (Miyoshi *et al.* 2005), while it is found in olivine-hosted melt inclusion. Many tephra of this study also contain normally zoned phenocrysts with high-Fo and high-An cores. As for the case of the silicic endmember, it is possible to assume the equilibrium of these cores with the mafic endmember melt. We therefore calculated the magmatic temperature of the pure mafic endmember, using Sakuyama's method (Sakuyama *et al.* 2014), which involves the combined application of

435 a plagioclase-melt hygrometer (Lange *et al.* 2009) and an olivine-saturated melt  
436 geothermometer (Sugawara 2000; Médard and Grove 2008). This method accounts for  
437 the H<sub>2</sub>O-dependency of the olivine thermometer by simultaneously solving for H<sub>2</sub>O and  
438 temperature using an additional constraint from the plagioclase hygrometer. The resulting  
439 temperature of mafic endmember magma varies between 1051 – 1063 °C, depending on  
440 the assumed pressure condition of the magma chamber (0.1 - 0.5 GPa, respectively). In  
441 addition, the entrapment temperature of the endmember inclusion (KSS-2-m1) is 1092  
442 °C based on the olivine-liquidus thermometer (Table S1, Sugawara 2000; Médard and  
443 Grove 2008). Considering the uncertainties of the thermometry methods, these  
444 temperature estimates are likely representing the range for the mafic magma. By taking  
445 the high temperature result (~1090 °C), the temperature difference between mafic and  
446 felsic (1010-1025 °C) endmembers is at least 65 °C.

447 The mixing trend among volatile elements is present and generally consistent  
448 with the trend of major elements. However, in detail, there are systematic disparities from  
449 the mixing curve (Supplementary document S4). Notably, abundances of H<sub>2</sub>O, S, and Cl  
450 for NKD14 sample, are depleted compared to the mixing trend traced by KSS melt  
451 inclusions (Fig. S4-4). This is best explained by significant degassing occurring  
452 during/after magma mixing. Furthermore, there is no single melt inclusion uniquely  
453 representing suitable mafic endmember volatile concentrations. The sample KSS-2-m1 is  
454 selected as the major element endmember, but its H<sub>2</sub>O contents are not the highest values.  
455 From the inspection of the trend, we inferred that the mafic endmember must have higher  
456 volatile content thus most likely lost water after its entrapment. If melt inclusions formed  
457 at a deeper depth, and were stored in a shallower magma chamber, it is expected that H<sup>+</sup>  
458 diffusion through the olivine would equilibrate the melt inclusion with the surrounding  
459 magma (Portnyagin *et al.* 2008; Chen *et al.* 2011; Gaetani *et al.* 2012; Bucholz *et al.*  
460 2013; Ferriss *et al.* 2018). Complete re-equilibrated melt inclusion would have erased the  
461 mixing trend. It is not the case here, we have found suitable endmember volatile element  
462 concentrations that satisfy the general trend. Because the mafic endmember magma is  
463 expected to be volatile-rich, its H<sub>2</sub>O concentration has to be higher than the H<sub>2</sub>O of the  
464 mixed-magma (e.g. that of the melt inclusions). The maximum estimated H<sub>2</sub>O  
465 concentration (4.68 wt. %) based on the hygrometer discussed above is therefore taken as  
466 the concentration for the volatile-rich mafic endmember. This value of 4.68 wt.% H<sub>2</sub>O is  
467 higher, by about 2 wt. %, than the highest H<sub>2</sub>O concentration measured in the melt  
468 inclusion. Sakuyama's method implicitly ignores the CO<sub>2</sub> activity in magma, and predicts  
469 lower H<sub>2</sub>O content when considering CO<sub>2</sub>-bearing system (by 0.9 wt. % less H<sub>2</sub>O,  
470 assessed from an experimental result of Melekhova *et al.* 2017). Furthermore, this volatile  
471 content is the value at the condition of olivine - plagioclase crystallization, most likely of  
472 the cooling magma in the crust. At this point, we have no other constraint on the H<sub>2</sub>O  
473 concentration of the mafic endmember and the primary magma could have an even higher  
474 H<sub>2</sub>O content.  
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476 ***Volatile concentrations of primitive basaltic melt***

477 The melt inclusions similar to the mafic endmember of the mixing model  
478 showed element concentrations with primitive character, such as low incompatible  
479 element concentrations (low K<sub>2</sub>O, Cl, and F), and higher volatile element such as S.  
480 However, H<sub>2</sub>O concentration is likely equilibrated to a lower pressure conditions, and  
481 CO<sub>2</sub> concentration in melt inclusion appears to be low. While these melt inclusions are  
482 hosted in high Fo olivine, the H<sub>2</sub>O and CO<sub>2</sub> abundances are not of primitive character.  
483 On the contrary, as F, S, and Cl are not expected to diffuse through the host olivine, and  
484 at the time of entrapment, they retain the value closest to the primitive magma. In an  
485 attempt to constrain the mafic endmember composition, compositional trends are  
486 examined with ratios of S, Cl, and F over K<sub>2</sub>O. These ratios are less affected by  
487 crystallization within melt inclusions and before entrapment, assuming the strong  
488 incompatibility of K<sub>2</sub>O in magmatic minerals near basaltic liquidus (Fig. 4). Because the  
489 composition of the mafic endmember points towards that of the primitive magma, the  
490 maximum values are taken from Fig. 4a, b, and c, as the primitive volatile ratios: S/K<sub>2</sub>O  
491 = 0.711, Cl/K<sub>2</sub>O = 0.170, and F/K<sub>2</sub>O = 0.047.

492 Dissolved S concentrations, up to 3750 ppm in the mafic group melt inclusions,  
493 are higher than many melt inclusions from subduction-like hydrous basalt (Fig. 5a; S  
494 mostly between 900 and 2500 ppm; e.g. Wallace 2005), while high S content appears to  
495 occur in oxidized magmas (Roggensack 2001; Webster *et al.* 2010). Our primitive magma  
496 S estimate is therefore 3750 ppm, the highest measured concentration in a melt inclusion  
497 of the mafic group. We also noted that S concentration of half of the melt inclusions are  
498 supersaturated in sulfide, plotting above the sulfur concentration at sulfide saturation  
499 (SCSS, Fig. 5b), while no sulfide is found in the melt inclusions or in the groundmass.  
500 These sulfide supersaturated melt inclusions must be at high  $fO_2$  condition, so that the  
501 concentration of sulfide-precipitating S<sup>2-</sup> in magma is lower than SCSS. In fact, it has  
502 been shown that arc basalt magmas have higher oxygen fugacities than MORB magmas  
503 (e.g. Wallace 2005; Kelley and Cottrell 2009). In addition to this, the redox state of the  
504 melt inclusions was determined and ranges from FMQ + 0.68 to FMQ + 1.41 (Average =  
505 1.05, 1 $\sigma$  = 0.17, corresponding to S<sup>6+</sup>/S<sub>total</sub> = 0.14 - 0.85, Table S1). Such oxidizing  
506 conditions allow for (i) higher S solubility in the mafic magma (Carroll and Rutherford  
507 1985; Jugo *et al.* 2010) and (ii) the presence of both S<sup>2-</sup> and S<sup>6+</sup> in the melt. Fig. 5b also  
508 shows a dashed line expected for the SCSS corrected for abundance of S<sup>2-</sup> species  
509 (S<sup>6+</sup>/S<sub>total</sub> = 0.85). Such first order correction puts all of our melt inclusion to be below  
510 sulfur saturation, consistent with our observation. Also, S concentrations of all the melt  
511 inclusions are under-saturated with respect to anhydrite saturation (SCAS was from 5300  
512 to 6000 ppm, Li and Ripley 2009; Baker and Moretti 2011). The elevated S concentrations  
513 in the undersaturated oxidized melt are therefore considered to represent, less-degassed,  
514 non-sulfide-fractionated, S concentrations, at least at the condition of the shallow storage  
515 depth.

516 Similarly, we also consider that the mafic endmember Cl and F concentrations

517 are representative of primary concentrations, as the exsolution pressure of Cl and F is at  
518 shallower depths (~100 MPa for Cl and ~10 MPa for F; Spilliaert *et al.* 2006) than for  
519 other volatile species. Cl concentration in the Aso primitive magma was 716 ppm (took  
520 the value of the mafic end member melt inclusion: KSS-2-m1) which is higher than that  
521 of MORB (Fig. 6; max 500 ppm Cl, Le Voyer *et al.* 2015), as it is interpreted as the  
522 addition of Cl to the subarc mantle from the subducting slab (e.g. Straub and Layne 2003).  
523 On the contrary, F concentration in the primitive magma was 324 ppm, which is the same  
524 order of magnitude as that of MORB (max 500 ppm, Le Voyer *et al.* 2015). This suggests  
525 that F addition to the subarc mantle was insignificant.

526 The Cl/F ratios of melt inclusions globally reflect the composition of the slab-  
527 agent added to the mantle source as F fractionate from Cl depending on the physical  
528 character of the flux leaving the slab (Le Voyer *et al.* 2010; Van den Bleeken and Koga  
529 2015; Narvaez *et al.* 2018). Observed Cl/F ratio of melt inclusions from Aso volcano are  
530 relatively high (from 1 to 2,5, Fig. 6) compared with that of MORB (Saal *et al.* 2002;  
531 Wanless and Shaw 2012; Wanless *et al.* 2014, 2015; Le Voyer *et al.* 2017; Shimizu *et al.*  
532 2019). Because Cl/F values in arc magma are characterized by several parameters such  
533 as the composition of slab, nature of the fluid and fractionating minerals in residual slab,  
534 and the degree of melting of arc magma, the physical character of slab derived flux is  
535 determined with a set of assumptions. In any case, the primitive magmas with MORB-  
536 like F concentration and a high Cl/F value (2.2) are found among the melt inclusions of  
537 calc-alkaline arc magmas in a typically “cold” subduction setting (Straub and Layne  
538 2003; Rose-Koga *et al.* 2014). The halogen characteristics of our Aso melt inclusions  
539 were in the range of typical arc magma, and the value of the primitive magma was close  
540 to the melt inclusions of Iwate volcano, a prototypical example of the arc magma derived  
541 from aqueous fluid driven from a cold oceanic crust (Rose-Koga *et al.* 2014). This  
542 indication is consistent with studies based on high bulk B/Nb ratios indicating aqueous  
543 fluid additions from the subducting Philippine sea plate in the source of the Aso basalt-  
544 basaltic andesite rocks (Miyoshi *et al.* 2008a, b). Other studies based on Sr, Nd, and Pb  
545 isotope variations of Quaternary lavas in the northern Kyushu area also support the model  
546 of slab-derived aqueous fluid addition from the subducted Philippine sea plate beneath  
547 Aso volcano (Shibata *et al.* 2014).

548

#### 549 ***Storage depth of the magma mixture and mafic endmember magma***

550 Solubility of volatiles in magma strongly depends on pressure and temperature  
551 conditions, and the chemical composition of the magma (e.g. Dixon *et al.* 1995). In this  
552 section, we discuss the storage depth of the mixed-magma based on H<sub>2</sub>O concentration.  
553 For example, measured volatile concentrations of melt inclusions are considered to be re-  
554 equilibrated to a condition of a magma storage. Alternatively, it is also possible to  
555 determine H<sub>2</sub>O concentrations estimated from plagioclase rim and groundmass glass pairs

556 (geo-hygrometer, Lange *et al.* 2009), likely corresponding to the value of the mixed  
557 magma during the plagioclase rim growth. The H<sub>2</sub>O concentrations of such mixed  
558 magmas are, ~2 wt.% for NKD14 and KKO, and ~3 wt.% for OJSU, OJSL and KSS  
559 (Table 4). Here, the hygrometer calibrated for the total fluid pressure is equal to  $P(\text{H}_2\text{O})$ ,  
560 neglecting the role of CO<sub>2</sub>. Yet, these values are comparable to those measured in the  
561 melt inclusions, and indicate a magma chamber pressures of 0.5 kbar for NKD14 and  
562 KKO, and 1.0 kbar for OJSU, OJSL, and KSS (*i.e.* 2.1 and 4 km deep, respectively, Fig.  
563 7, note the depth is calculated using a density of 2200 kg/m<sup>3</sup> for the first 1 km and 2700  
564 kg/m<sup>3</sup> for the crust at greater depths (Komazawa 1995), by solving for the depth,  $h$  [m],  
565  $P = g \{2200 (1000) + 2700 (h-1000)\}$ ). Because CO<sub>2</sub> concentrations in the felsic group  
566 melt inclusions are notably low (0 - 77 ppm, Table S1), it makes negligible changes to  
567 the calculated equilibrium pressure (~20 MPa, using RhyoliteMELTS/MagmaSat,  
568 Ghiorso and Gualda, 2015). These pressures correspond to the depths of magma storage  
569 after mixing, since (1) the last growth rim of plagioclase must have occurred after mixing,  
570 as both normal and reverse zoning plagioclase have the same rim composition. (2) It is  
571 less likely that plagioclase rim significantly grew during sub-Plinian and Strombolian  
572 eruption. Melt inclusion H<sub>2</sub>O concentrations range from 0.47 to 2.89 wt. %, indicating  
573 the minimum equilibrium pressure in agreement with the plagioclase-rim method. Two  
574 magma chambers, C2 and C1, detected by geophysical studies are also located at similar  
575 depths, 1 - 4 and 3 - 10 km, respectively (Tsutsui and Sudo 2004; Sudo *et al.* 2006; Hata  
576 *et al.* 2016; Fig. 7). At the depths, because the solubility of CO<sub>2</sub> is so low, the conclusion  
577 presented here would remain the same even if the hygrometer calculation does not  
578 account for CO<sub>2</sub>.

579 The magma of the mafic endmember is expected to have more than 4.68 wt. %  
580 H<sub>2</sub>O, and it is therefore derived from a greater depth than these C1 and C2 storage depths.  
581 The corresponding equilibrium pressure is approximately 2.7 kbar (SolEx, Witham *et al.*  
582 2012; note MagmaSat give 2.3 kbar, Ghiorso and Gualda 2015; with CO<sub>2</sub> of 340 ppm  
583 reported in a melt inclusion from Nakadake by Saito *et al.* 2018) that is about 10 km deep  
584 below the edifice. As gas bubbles were seen in melt inclusions, large amounts of CO<sub>2</sub>  
585 incorporated in such shrinkage bubbles significantly increases the entrapment pressure  
586 estimation (e.g. Moore *et al.* 2015). Therefore, our pressure estimations are minimum and  
587 true entrapment pressures of the basaltic magma certainly occurred at a depth greater than  
588 10 km. Recent melt inclusion studies reported more than 40-90 % of the initial CO<sub>2</sub> that  
589 was dissolved in the melt at the time of entrapment was lost to shrinkage bubbles, with  
590 an average loss of 75-80 % (Hartley *et al.* 2014; Moore *et al.* 2015; Wallace *et al.* 2015).  
591 If 90 % of initial CO<sub>2</sub> is present in the shrinkage bubble and a maximum CO<sub>2</sub> value of  
592 340 ppm is assumed in the melt (from Saito *et al.* 2018), then the expected initial value  
593 of the melt would reach 3400 ppm. This value is in the same order of magnitude as the  
594 initial CO<sub>2</sub> concentration in a typical primary arc magma (Aster *et al.* 2016), and in this  
595 case the saturation pressure would exceed 5 kbar (~19 km depth equivalent). However,  
596 at the time of this study, bubble sizes were not documented with impossibility to go back



597 to measuring them *a posteriori* since most are now polished away. In this case, we chose  
598 not to use the CO<sub>2</sub> data of the melt inclusions. Hata *et al.* (2016) also reported a magma  
599 pathway from depths deeper than 11 km, feeding the deeper C1 reservoir. Also, the  
600 presence of a seismic low-velocity layer at 11 - 25 km depth was reported and attributed  
601 to a magma ponding location beneath the Aso caldera (Abe *et al.* 2010). These  
602 observations are in good agreement with our petrological implication for the presence of  
603 volatile-rich basaltic magma beneath C1 magma reservoir at a depth greater than 10 km.

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#### 606 ***Persistent degassing from a deeper magma reservoir (>10km)***

#### 607 Excess degassing of SO<sub>2</sub> and CO<sub>2</sub> from Nakadake 2014 eruption

608 Decomposition of magma mixing endmembers and the subsequent  
609 identification of the primitive magma composition provide critical information for  
610 investigating volcanic emission of gas and its mass balance. For the 2014 eruption of Aso  
611 volcano, the mass of erupted magma is insufficient to account for the mass of the observed  
612 SO<sub>2</sub> gas emissions (Saito *et al.* 2018), which is commonly reported in many active  
613 volcanoes and is called “excess degassing”. For Aso, approximately 90 – 140 times more  
614 magma than the erupted tephra mass is needed to account for the observed total SO<sub>2</sub>  
615 emission of  $1.4 - 2.2 \times 10^5$  tons for the period of 70 days from Nov. 2014 to Feb. 2015  
616 (Table 5 and; also Saito *et al.* 2018). As demonstrated in the sections above, NKD magma  
617 is a mixed magma, so the gas phase must have come from the degassing of mafic and  
618 felsic endmember magmas. For this reason, we have used the highest volatile element  
619 concentrations determined for Nakadake samples (*i.e.* mixed magmas) for the sulfur  
620 budget calculation (Supplemental material S4). This calculation simply demonstrates that  
621 the mixed magmas are inadequate sources of the observed volcanic gas flux. The most  
622 commonly proposed explanation for this excess is the existence of a gas/fluid phase in  
623 the magma, possibly containing C-O-H-S and co-existing with the magma prior to the  
624 eruption (e.g. Anderson 1973; Wallace 2001; Scaillet and Pichavant 2003; Shinohara  
625 2008). The magmatic volatile component must originate from the volatile-rich magma  
626 corresponding to the basaltic/mafic endmember. This magma must degas at a deeper  
627 depth than that indicated by the equilibrium solubility depth of NKD samples (~2 km at  
628 the depth of C2). Therefore, the degree of S excess should be re-assessed with a deeper,  
629 volatile-rich, basaltic magma. It should be noted that the observation of S-excess is  
630 common (*e.g.* Wallace and Edmonds 2011) and we simply point out here that the eruption  
631 of Nakadake edifice of Aso also shows such excess.

632 There are geophysical observations indicating that degassing is fed by volcanic  
633 activities corresponding to depths deeper than ~2 km. The SO<sub>2</sub> gas emission of Aso  
634 volcano is continuously monitored during the eruptive and the quiescent periods. The  
635 amount of SO<sub>2</sub> emission during the quiescent period (alternatively, “persistent

636 degassing"; Shinohara 2008) is more than 100 – 200 tons/day from 1975 to 2006 (Mori  
637 *et al.* 2013). Ground deformation measurements since 1937 revealed deformation  
638 (deflation) of the deeper C1 magma reservoir synchronous to gas emission from  
639 Nakadake cone during the quiescent period (Sudo *et al.* 2006). This C1 deformation,  
640 located approximately 3 km West of Nakadake, causes no significant deformation on the  
641 surface around the crater. This degassing is therefore interpreted as open-system  
642 degassing of the C1 magma reservoir through stable conduit passages to Nakadake crater  
643 (Yamamoto *et al.* 1999; Sudo *et al.* 2006). Furthermore, deflation of the C1 reservoir was  
644 seen to slow down from 2008 to early 2014, before acceleration of inflation of the C1  
645 reservoir from July 2014, shortly before the eruption in November (Ohkura *et al.* 2015;  
646 JMA 2016). These changes were accompanied by a sudden increase in persistent SO<sub>2</sub> gas  
647 emission in September 2013, considered as a precursor to the 2014 - 2015 eruptive period  
648 (JMA 2016). These temporal and spatial relationships of geophysical observation and  
649 degassing activities strongly indicate the role of the deeper C1 reservoir during degassing.  
650 However, to sustain replenishment of the C1 reservoir, the magma must be supplied from  
651 an even deeper depth (greater than 10 km; Sudo *et al.* 2006; Hata *et al.* 2016). For example,  
652 the slowing down of deflation in 2008 and subsequent inflation in 2014 can be interpreted  
653 as events relating to magma replenishment of the C1 reservoir (Ohkura *et al.* 2015, 2017).

654 We also found excess degassing of CO<sub>2</sub> for this eruption period. Using the  
655 maximum pre-eruptive CO<sub>2</sub> concentration of 340 ppm measured in a melt inclusion of  
656 Nakadake eruption products (Saito *et al.* 2018), and assuming there is no pre-eruptive  
657 vapor phase approximately 1700 – 2700 times more dissolved CO<sub>2</sub> is required to account  
658 for the observed 0.77 – 1.2 × 10<sup>6</sup> tons of CO<sub>2</sub> emission (Table 5). In the case of Aso, the  
659 gas phase must be derived from a depth greater than that of the C1 reservoir.

#### 660 Modelling degassing of the deep magma reservoir

661 In the sections above, the equilibrium depths of volatile concentration in  
662 magma were used to indicate magma storage depth, where stagnated-magma and gas  
663 remain at equilibrium. Equilibrium gas composition at that depth should be the source of  
664 the gas composition observed at the surface, only if the gas segregated and travelled to  
665 the surface without precipitating solids or re-dissolving gas in between. In detail, the  
666 speciation of mixed gas is a function of pressure, temperature, and oxygen fugacity, but  
667 the gas system composition can be considered closed. With this hypothesis, it is possible  
668 to determine the depth at which the observed gas composition is in equilibrium with a  
669 magma (e.g. Burton *et al.* 2007; Allard 2010).

670 Fig. 8 is a result of such a calculation using SolEx (Witham *et al.* 2012),  
671 showing the variations in H<sub>2</sub>O/S, CO<sub>2</sub>/S, and Cl/S molar ratios of gas in equilibrium with  
672 a magma as a function of pressure. The observed H<sub>2</sub>O/S gas composition of the 2014  
673 eruption (shown as horizontal dotted red lines) intersects with the equilibrium gas  
674 composition (plain lines Fig. 8) between 2.6 to 3.3 kbars. These pressures correspond to

675 10 - 13 km depth below the edifice. The family of curved lines represent the results of the  
676 model with varying initial CO<sub>2</sub> concentration (400, 750, and 1000 ppm), as the initial CO<sub>2</sub>  
677 concentration was unknown (but for sure higher than 340 ppm). For the range of  
678 degassing trajectory, the observed Cl/S values (horizontal dotted blue line) intersect with  
679 the model calculations between 2.6 and 3.8 kbars. H<sub>2</sub>O/S and CO<sub>2</sub>/S are less sensitive to  
680 the variation of the initial CO<sub>2</sub> concentration. It should be noted that the closed system  
681 degassing path here is used to find a unique pressure condition at which the observed high  
682 temperature gas composition is at the equilibrium with a mafic magma. The systematics  
683 shows that only CO<sub>2</sub> concentrations between 400 and 750 ppm can produce the observed  
684 gas composition at pressures between 2.6 to 3.3 kbars, satisfying the observed ranges of  
685 H<sub>2</sub>O/S, CO<sub>2</sub>/S, and Cl/S all at once.

686 Therefore, the observed gas composition must have been derived from the  
687 depth of about 10 to 13 km (~2.6 to 3.3 kb) at which it was in equilibrium with a magma  
688 with the initial magmatic CO<sub>2</sub> concentration between 400 to 750 ppm (Fig. 8). The  
689 determination of initial CO<sub>2</sub> is strongly tied to the Cl and S partitioning into H<sub>2</sub>O-CO<sub>2</sub>  
690 fluid, and we think that these values may need to be revised once we have a better  
691 understanding of Cl and S element partitioning. Lastly, this conclusion is highly model  
692 dependent. Here, we chose to use SolEx for its agreement with the variation of S in the  
693 melt inclusions. However, one must be cautious with the use of SolEx, because it is shown  
694 to fail to reproduce degassing trajectories of other volcanoes (e.g., Werner et al. 2020).

695 Fig. 9 shows the observed gas composition emitted from Aso volcano in the  
696 quiescent period and eruptive period. The gas data shown in Fig. 8 correspond to the  
697 eruptive period data (orange). While Fig. 8 demonstrated the occurrence of deep gas  
698 segregation, it does not explain the range of gas composition observed before and, even,  
699 during the eruption of the Nakadake cone. We modelled the variation in gas composition  
700 using (1) a degassing trend (solid line), the same model as that used in Fig. 8, and it fits  
701 the data with some scatter and (2) a mixing line (dotted line) between a deep reservoir,  
702 for example at the storage depth around 10 km, and a shallow reservoir (C2 or top of C1).  
703 Both explain the data (Fig. 9). Thus, on the one hand, it is possible to consider the  
704 variation in gas composition as the variation in gas segregation depth. On the other hand,  
705 the observed gas composition variation potentially represents the different degrees of  
706 mixing between discharged gas from NKD14 magma at shallow depth and segregated  
707 gas from a deep reservoir (*i.e.* gas segregation depth at 10 km; Fig. 8 and 10). Case 1 is  
708 less likely to apply here. Because of the closed-system degassing model (solid line), the  
709 ascending primitive magma would have been expected to be in equilibrium with gas all  
710 the time until it reaches near the surface. Therefore, the gas composition representing the  
711 low pressure must have been in equilibrium with the mafic magmas at the shallowest  
712 depth. However, no volcanic product of primitive composition was found during the  
713 eruption of Nakadake cone. Instead, observed volcanic products were all the result of  
714 mixing. As case 1 is unlikely, we conclude that the mixing of gases (case 2) is the process  
715 explaining the observed compositional variation. The gas mixing could have been

716 possible, for example, by bubble accumulation at the roof of a magma chamber (Jaupart  
717 and Vergnolle 1988, 1989).

718

### 719 *Implications for understanding current Aso volcanic activity*

720

721 This study identified the role of a volatile-rich, primitive magma during  
722 volcanic degassing (Fig. 10). During ascent, the volatile-rich primitive magma started to  
723 degas at a depth between 11 and 13 km (2.8 to 3.5 kbars, SolEx) based on the initial  
724 volatile concentration. The gas and magma reached 10 km depth maintaining equilibrium.  
725 At that depth, the gas segregated from the magma and moved to the surface without any  
726 significant modification. The separation of gas from magma was potentially caused by  
727 stopping of magma ascent, which contributed to overpressuring of C1 observed by the  
728 deformation of the edifice. The magma then moved upwards and mixed with silicic  
729 magma in the C1 magma reservoir. While the exact depth of mixing was undetermined,  
730 the mixed magma was eventually stored at depths of C1 and C2 (for example 2 km (C2)  
731 for NKD14 and KKO samples, and 4 km (C1) for OJSU, OJSL, and KSS samples).

732 The segregated deep gas mixed with the shallow gases upon its ascent at  
733 shallower levels. We were unable to constrain the exact mechanism of the deep gas  
734 transport to the shallow depth without significant interaction. One possibility is the  
735 presence of porosity providing connected gas passages. However, this mechanism implies  
736 the rigid structure at a temperature lower than magmatic conditions, and this contradicts  
737 with the observation of the high temperature gas indicating magmatic degassing.  
738 Alternatively, a convection within C1 and C2 potentially serves a “gas pump”. In this  
739 scenario, a shallow mixed magma (devoid of volatile elements) descends by convection  
740 to the bottom of C1 where gas segregation is occurring. Upon the return of such magma  
741 to shallow depth, it drags the gas component with it. Such parcel of magma may further  
742 pull gas bubbles from the shallow depth and result in a bulk system of mixed gas. If the  
743 magma degases completely, the resulting gas composition will be identical to the result  
744 of the gas mixing. This scenario would fail if some batch of magma do not degas and  
745 remain in some depth; it requires complete degassing. Shinohara (2018) proposed a model  
746 which suggested that the observed chemical variation in gases during eruption was caused  
747 by mixing of gases derived from magmas at different depths. This model is consistent  
748 with our gas mixing model. Our model furthered understanding of Aso activity, by (1)  
749 quantifying the volatile concentration of primitive mafic magma, and (2) by determining  
750 its degassing, segregation, and storage depths.

### 751 **Summary and conclusion**

752 Petrological analyses of melt inclusions in basaltic tephra from the late  
753 Holocene eruption of Aso volcano provided us with insights into the process of the  
754 persistent degassing in a magma plumbing system.

755 (1) The compositions of melt inclusions in phenocrysts from recent eruption

756 products (Nov. 2014) of Nakadake cone were homogeneous, evolved and  
757 degassed ( $S < 434$  ppm), while compositions of melt inclusions in  
758 phenocrysts from Kishimadake and other related cones (ca. 3.0 – 3.7 ka)  
759 showed variable compositions with two extreme endmembers. One was  
760 mafic ( $\text{SiO}_2 < 55\text{wt.}\%$ ) and volatile-rich, and the other was felsic ( $\text{SiO}_2 >$   
761  $55\text{wt.}\%$ ) and relatively degassed. Mafic melt inclusions were hosted in  
762 high-Fo olivine and some plagioclase, whereas felsic melt inclusions were  
763 hosted in plagioclase, clinopyroxene, orthopyroxene and low-Fo olivine.

- 764 (2) Reverse mineral zoning, and highly variable mineral and glass  
765 compositions indicated a mixing process during which volatile-rich basaltic  
766 magma was injected in a degassed shallower magma.
- 767 (3) The estimated depths of pre-eruptive magma were approximately 2 km (C2  
768 magma reservoir) for Strombolian eruption of Nakadake cone (Nov. 27,  
769 2014) and Kamikomezuka cone (3.0 ka) and approximately 4 km (top of  
770 C1 magma reservoir) for sub-Plinian eruption of Ojodake and Kishimadake  
771 cones in 3.5 – 3.7 ka. These depths corresponded to the partially melted  
772 zones characterized by geophysical investigations (e.g. Hata *et al.* 2016).  
773 Furthermore, volatile-rich primitive magma originated from a deeper level  
774 (greater than 10 km) than these two magma reservoirs.
- 775 (4) Even with the highest S concentration of NKD14, it was impossible to  
776 account for the excess S gas emission. This suggests an addition of gas  
777 derived from the deep magma reservoir (greater than 10 km depth).
- 778 (5) Initial volatile concentrations of the volatile-rich primitive magma were  
779 determined from the systematics of melt inclusion, plagioclase  
780 compositions, and degassing model:  $>4.68$  wt.%  $\text{H}_2\text{O}$ , 3750 ppm S, 716  
781 ppm Cl and 324 ppm F.
- 782 (6) Emitted gas compositions constrained the initial  $\text{CO}_2$  concentration ranges  
783 in the magma, between 400 and 750 ppm.
- 784 (7) The variation observed in volcanic gas composition was best explained by  
785 the mixing of the gas segregated at 10 km depth with those from the shallow  
786 reservoirs.

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1174

## 1175 **Figure caption**

1176

1177 **Fig. 1** Locations of volcanic cones and sampling sites of the study. Main map shows  
1178 topography of Aso volcano with a 50 m contour interval. Volcanic cones are indicated by  
1179 solid triangles. Sampling sites are indicated by solid circles. Location of Aso volcano is  
1180 indicated in an inset in the top left corner, showing Japanese islands with plate boundaries:  
1181 solid line, active subduction plate boundary, and dashed line, diffused plate boundary.  
1182 Stratigraphic log shows corresponding eruption stages and nature of volcanic deposits in  
1183 A9418 site (Miyabuchi and Watanabe 1997). Kishimadake scoria (KSS) and Ojodake  
1184 scoria (OJSL and OJSU) were collected at the indicated units. Kamikomezuka scoria  
1185 (KKO) and Nakadake scoria (NKD14) were collected at the location indicated by field  
1186 symbols. KKO in parenthesis indicates the relative eruption age (top of N4 stage).  
1187

1188 **Fig. 2** Microscope photographs (a-d) and backscatter electron images (e, f) of  
1189 representative phenocrysts and textures from Holocene basaltic tephra products in Aso  
1190 central cones. (a) Plagioclase (Pl) phenocrysts with honeycomb texture in KKO, under  
1191 cross-polarized light. Clinopyroxene (Cpx) and olivine (Ol) phenocrysts also present. (b)  
1192 An orthopyroxene (Opx) phenocryst with reaction rim consisted of clinopyroxene and  
1193 olivine in KSS, under cross-polarized light. (c) An aggregate of phenocryst consisted of  
1194 plagioclase, clinopyroxene and olivine in KSS, under cross-polarized light. (d) A  
1195 microlite-rich part of groundmass in KSS with abundant crystals, under plain-polarized  
1196 light. (e) High-Fo olivine phenocryst and its melt inclusions observed in KSS. (f) A  
1197 texture of plagioclase phenocryst with (optically) dusty zone, in the middle part of the  
1198 crystal, observed in NKD14.

1199

1200 **Fig. 3** Major element and volatile concentrations in melt inclusions from Holocene  
1201 basaltic tephra products in Aso central cones. Melt inclusion data from minerals other  
1202 than olivine are shown as open symbols. The melt inclusion data are compared to bulk  
1203 tephra and average groundmass glass compositions, solid circles, and solid squares,  
1204 respectively. The variation of Fo content in all olivine cores is also shown as relative  
1205 density estimates on the left side of (a). Dashed line in Figure 3a and 3b indicates Fo72.  
1206 Bulk lava compositions of late-Holocene volcanic products, reported previously, are also  
1207 shown as solid grey circles in (c) (Miyoshi *et al.* 2005; Miyabuchi 2010, 2017; Saito *et*  
1208 *al.* 2018). All oxide concentrations were normalized to 100 wt. %, excluding volatile  
1209 element abundance.

1210

1211 **Fig. 4** Ratios of volatiles/K<sub>2</sub>O vs. K<sub>2</sub>O in melt inclusions and groundmass glasses (the  
1212 same symbols as Fig. 3). All volatile elements showed a systematic decrease (a,

1213 H<sub>2</sub>O/K<sub>2</sub>O; b, S/K<sub>2</sub>O; c, Cl/K<sub>2</sub>O) with respect to increasing K<sub>2</sub>O. High volatile/K<sub>2</sub>O  
1214 corresponds to high volatile concentration as well as low K<sub>2</sub>O.

1215

1216 **Fig. 5** (a) Variations in S and total FeO concentrations of melt inclusions and groundmass  
1217 glasses. Symbols are the same as Fig. 3. Melt inclusions hosted in olivine show abrupt  
1218 decrease in S content, and the change lacks correlation with FeO. (b) Measured S  
1219 concentration in olivine-hosted melt inclusions as a function of sulfur concentration at  
1220 sulfide saturation (SCSS). In this figure, the samples at the saturation can be seen on the  
1221 solid grey line (1:1 line). The dashed line shows the SCSS corrected for abundance of S<sup>2-</sup>,  
1222 which was 0.85, the maximum value. All data plots below the corrected SCSS. SCSS is  
1223 calculated according to Fortin *et al.* (2015) using the trapping temperature ( $T_{\text{trap}}$ ) of the  
1224 melt inclusion (Table S1) at 0.05 GPa (for NKD14) and 0.1 GPa (for KKO, OJSU, OJSL,  
1225 and KSS) based on estimated volatile saturation pressure of the pre-eruptive magma  
1226 reservoir. For melt inclusions without measured H<sub>2</sub>O concentration, we attributed average  
1227 H<sub>2</sub>O concentrations (by SIMS) of NKD14 and KSS.

1228

1229 **Fig. 6** Variations in Cl and F concentrations of melt inclusions (KSS and NKD14) plotted  
1230 in decadic log scale. The KSS and NKD14 data are compared with the volatile rich mafic  
1231 primitive magma, represented by a large blue diamond. The two areas are indicated to  
1232 show the Cl and F concentrations in primitive arc basalt and MORB, which were  
1233 measured in olivine-hosted melt inclusions (taken from Van den Bleeken and Koga 2015).  
1234 MORB melt inclusions were plotted as solid light-grey circles (Saal *et al.* 2002; Wanless  
1235 and Shaw 2012; Wanless *et al.* 2014, 2015; Le Voyer *et al.* 2017; Shimizu *et al.* 2019)  
1236 including Cl-rich data of assimilated MORB samples by seawater derived component.  
1237 Melt inclusions from Iwate volcanoes, one of low F concentration arc lava from Japan  
1238 arc, is shown as an example of a cold subduction zone (red plus sign, Rose-Koga *et al.*  
1239 2014). The lines show the constant Cl/F values, and the values are indicated above the  
1240 line.

1241

1242 **Fig. 7** Pre-eruptive temperatures and water concentrations calculated from the  
1243 compositions of plagioclase rim and groundmass glass. The squares represent the  
1244 estimated range of temperature and H<sub>2</sub>O concentration. The calculation was based on a  
1245 combination of a plagioclase-olivine-liquid hygrometer (Lange *et al.* 2009) and olivine-  
1246 saturated liquid geothermometer (Sugawara 2000; Médard and Grove 2008) according to  
1247 Sakuyama *et al.* (2014), assuming the crustal conditions (0.1 – 0.5 GPa). Standard  
1248 deviations ( $1\sigma$ ) of the difference in temperature and H<sub>2</sub>O concentration between  
1249 estimation and expectations using this method were 23 °C and 0.6 wt. %, respectively.  
1250 The dashed lines represent water saturation isobars for groundmass glass composition of  
1251 KSS calculated according to Moore *et al.* (1998); the corresponding pressures (kbar) and  
1252 the depths (km) are indicated to the left of the line. The depth in the crust was calculated  
1253 assuming a density of 2400 kg/m<sup>3</sup> for less than 1 km depth and a density of 2700 kg/m<sup>3</sup>

1254 for depth greater than 1 km (Komazawa 1995). The red and blue colors show the range  
1255 of the magma storage depths: red, C1 from 3 to 9 km depth, and blue, C2 from 1 to 3 km  
1256 depth beneath Nakadake's first crater (Sudo and Kong 2001; Sudo *et al.* 2006; Hata *et al.*  
1257 2016).

1258

1259 **Fig. 8** Evolution of H<sub>2</sub>O/S, CO<sub>2</sub>/S, and Cl/S molar ratios of magmatic gases in equilibrium  
1260 with the primitive basaltic melt plotted against pressure. Horizontal dotted lines indicate  
1261 measured gas compositions of H<sub>2</sub>O/S = 22.5, CO<sub>2</sub>/S = 6, and Cl/S = 0.07 (S as total sulfur)  
1262 with the variation observed with propagated measurement uncertainties shown by shaded  
1263 regions. The values correspond to high-pressure gas component A reported in previous  
1264 studies (Shinohara 2013; Shinohara *et al.* 2018). SO<sub>2</sub> and H<sub>2</sub>S components were added to  
1265 calculate the total S. Horizontal colored bars at the top of the panel show pressure ranges  
1266 corresponding to the C1, C2, and LVL. To evaluate the evolution of the gas composition  
1267 of the basaltic magma from a great depth, we modelled the equilibrium compositions of  
1268 gas and magma for a closed system magma ascent using SolEx (Witham *et al.* 2012).  
1269 Grey field indicates the pressure range of 2.3 – 2.9 kbars, at which conditions the modeled  
1270 gas compositions (solid and dashed curves) intersect with measured gas ratios. Thus, the  
1271 observed gas composition potentially originated at these depths. We chose the primitive  
1272 magma composition (discussed in the text) with values of FMQ+1.4 and 1090 °C based  
1273 on the individual melt analyses. We tested initial CO<sub>2</sub> contents of 400, 750 and 1000 ppm  
1274 for the calculation, as the CO<sub>2</sub> concentration was unknown to us. In combination with  
1275 H<sub>2</sub>O/S and Cl/S, the initial CO<sub>2</sub> concentration from 400 to 750 ppm produced the gas  
1276 composition evolution curves that agree at a similar depth.

1277

1278 **Fig. 9** Observed volcanic gas compositions emitted from Aso volcano compared with  
1279 modelled variation in CO<sub>2</sub>/SO<sub>2</sub> and H<sub>2</sub>O/CO<sub>2</sub> molar ratios. Observed gas compositions  
1280 are those during quiescent periods (fumarole and lake gases) and during eruptive periods  
1281 (Shinohara *et al.* 2018). Some data points were reported as poor quality observations due  
1282 to atmospheric H<sub>2</sub>O, and they are plotted as smaller symbols. Solid curve is a modeled  
1283 gas composition of closed-system ascent of primitive magma with 800 ppm initial CO<sub>2</sub>  
1284 concentration, which is the same as that used in Fig. 8. The gas compositions on the curve  
1285 indicate the different depths of gas segregation, and thus, the variation among  
1286 observations can be attributed to a variation in segregation depths. Dashed curve shows a  
1287 mixing curve between a CO<sub>2</sub>-rich gas in equilibrium with the deep primitive magma at  
1288 2.6 kbar (crossed circle) and a gas discharged from NKD14 magma at shallow depth  
1289 (asterisk) representing the gas stored at the shallow reservoir. The shallow gas component  
1290 was calculated from the difference in volatile concentrations between pre-eruptive melt  
1291 and groundmass glass.

1292

1293 **Fig. 10** Schematic summary of the magma and gas evolution in the Aso plumbing system.  
1294 Magma passage is indicated by the color ranging red-orange-yellow. The gas is white,



1295 and the grey zone corresponds to the conductivity anomaly (Hata *et al.* 2016). C1, C2,  
1296 and LVL (black thick bars) are the zones of magma stagnations determined by  
1297 geophysical methods.





















