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## Sulfur isotope and trace element systematics in arc magmas: seeing through the degassing via a melt inclusion study of Kyushu Island volcanoes, Japan

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1

2 **Sulfur isotope and trace element systematics in arc magmas: seeing**  
3 **through the degassing via a melt inclusion study of Kyushu Island**  
4 **volcanoes, Japan**

5

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25

26 **Abstract**

27 Sulfur is a minor element in magmas but one of the major volatile elements released in  
28 volcanic systems, from the magma to the fluid phase upon ascent. Not only are sulfur  
29 gases potentially toxic for humans and plants, they are also involved in causing drastic  
30 climate changes after major volcanic eruptions. Therefore, studies are carried out by the  
31 geoscience community to assess the magmatic sulfur flux by looking at the sulfur content  
32 and isotopes in erupted products, with the ultimate aim of improving understanding of  
33 the sulfur cycle in subduction zones. Kyushu island in Japan hosts 25 volcanoes among  
34 which 11 are active and represent a natural hazard for the local population. It is perhaps  
35 the most suitable site for the study of the sulfur cycle for its availability of recent volcanic  
36 deposits and its many, highly monitored volcanoes. We investigated sulfur and sulfur  
37 isotope compositions of the magma source of Kyushu Island arc using olivine-hosted melt  
38 inclusions in mafic tephra and lavas, from 8 volcanoes (9 Holocene samples) going from  
39 Northern Kyushu with Oninomi, Yufu, Kujū, and Aso, to Southern Kyushu volcanoes  
40 such as Kirishima-Ohachidake, Kirishima-Shinmoedake, Sumiyoshi-ike, and  
41 Kaimondake, and one back arc volcano, Fukue-Onidake. We measured major, trace and  
42 volatile elements and S isotopes ( $\delta^{34}\text{S}$ ) in melt inclusions. Magma composition recorded  
43 in the inclusions ranges from basalt to andesite ( $\text{SiO}_2$  ranging from 40.3 to 60.7 wt. %).  
44 For each edifice, we identified the least degassed and least differentiated compositions  
45 based on volatile and trace element systematics, and selected the melt inclusions closest  
46 to their primitive melts. Comparing these primitive magmas, Sr/Y underlines a  
47 compositional dichotomy between volcanoes from northern ( $\text{Sr}/\text{Y} > 20$ ) and southern  
48 Kyushu ( $\text{Sr}/\text{Y} < 20$ ), separated by a non-volcanic area corresponding to the subduction of  
49 the Kyushu-Palau ridge. The  $\delta^{34}\text{S}$  in melt inclusions range from  $-0.32 \pm 0.79$  ‰ to  
50  $+9.43 \pm 0.47$  ‰ ( $2\sigma$ ) and trace the source of the magma from the different volcanoes, rather  
51 than degassing or crustal fractionation processes.  $\delta^{34}\text{S}$  is not fractionated by the nature of  
52 the fluid (aqueous or melt) metasomatizing the mantle wedge, therefore it is not the first  
53 order factor controlling the sulfur isotope variations. Instead, this study illustrates the  
54 need for a heavy  $\delta^{34}\text{S}$  component, likely sulfate from seawater, contained in the agent that  
55 metasomatized the mantle beneath the arc. If such an observation is confirmed in other  
56 subduction zones, sulfur isotopes in melt inclusions may be an effective way to trace

57 seawater input into the mantle beneath arcs.

58

59 **Keywords**

60 volatile elements, trace elements, slab input, seawater, subduction.

61

## 62 **Introduction**

63       The behavior of sulfur (and of its isotopes) in magma is complex because of its  
64 multiple valency and its partitioning into melt, gas, fluids and S-bearing minerals.  
65 Because sulfur can become saturated in the magmatic system and crystallize as sulfides,  
66 its geochemical behavior is expected to differ significantly from lithophile trace elements,  
67 such as lanthanide series rare earth elements (REE), or high field strength elements (Nb,  
68 Ta, Hf: HFSE). Yet, assessments of the sulfur budget and its evolution in magma are  
69 important to volcanology and mantle geochemistry, especially since SO<sub>2</sub> in volcanic gas  
70 is considered as an indicator of magmatic flux beneath a volcano (e.g. Symonds et al.  
71 1994; Métrich and Mandeville, 2010). Furthermore, the sulfur isotope composition of arc  
72 magma sources is challenging to determine precisely, partly because significant sulfur  
73 isotope fractionation is overprinted in erupted products (e.g. Mandeville et al., 2009). It  
74 appears nearly impossible to determine the source sulfur isotope composition through  
75 subaerial eruption products. Instead, the systematic study of sulfur isotope composition  
76 in olivine-hosted magmatic inclusions is much more revealing, as the olivine can prevent  
77 diffusive sulfur-loss through the host crystal (e.g. Bucholz et al., 2013). There have also  
78 been successful attempts recently to find primitive sulfur isotopic compositions in melt  
79 inclusions from Lassen volcano and the 2018 Kilauea flow (Muth and Wallace 2021;  
80 Lerner et al. 2021). However, other olivine inclusions have not been as successful in  
81 sampling unmodified magmas; the melt inclusion studies of the Lesser Antilles, Canary  
82 Islands and Kamchatka have recorded indications of degassing or sulfide fractionation  
83 (Bouvier et al., 2008; Beaudry et al., 2018; Gurenko et al., 2018; 2021, respectively), with  
84  $\delta^{34}\text{S}$  values ranging from -9.0 to +7.0 ‰, -5.9 to +3.2 ‰ and -1.6 to +12.3 ‰,  
85 respectively. Clearly, further efforts to interpret sulfur systematics in different arc systems  
86 are needed to help improve our understanding of the sulfur cycle in subduction zones.

87       Kyushu Island is one of the four main islands of Japan and is known for its high  
88 concentration of active volcanoes; 12 volcanoes are considered active along the 240 km  
89 segment of Kyushu. This makes it an ideal location to sample Holocene eruptions along  
90 an arc section. Despite this ideal setting, there are no systematic studies reporting melt  
91 inclusion compositions along this arc or characterization of mantle variations below these  
92 arc volcanoes. Furthermore, sulfur fluxes of the volcanoes are well-monitored, and for

93 example, Aso volcano is known for its persistent sulfur emissions (approximately 600  
94 tons per day; Carn et al. 2017; Japan Meteorological Agency, 2020). It is therefore an  
95 ideal arc to carry out a systematic investigation of sulfur along the arc segment. This study  
96 reports the variation in sulfur isotopes recorded in this volcanic segment, and its relation  
97 to the geodynamic setting of this arc.

98

## 99 **Geological setting**

### 100 *Tectonic setting*

101 The modern volcanic activity of Kyushu Island is related to the subduction of the  
102 Philippine Sea plate (Nakada and Kamata, 1991; Miyoshi et al., 2008ab, 2010; Shibata et  
103 al., 2014). This is composed of two segments of different ages, divided by the Kyushu-  
104 Palau ridge (Fig. 1a): a young, hot northern segment (Shikoku Basin, 15 – 26 Ma; Okino  
105 et al., 1994), and an old, cold southern segment (West Philippine Basin, 30 – 54 Ma;  
106 Shibata et al., 1977; Deschamps and Lallemand, 2002; Ishizuka et al., 2018). The  
107 Kyushu-Palau ridge is a remnant of Eocene–Oligocene arc that split away from the Izu-  
108 Bonin-Mariana arc, due to spreading in the Shikoku Basin (e.g., Seno and Maruyama,  
109 1984). Recent seismic and geomagnetic studies indicate the presence of the subducting  
110 Kyushu-Palau ridge beneath central Kyushu, precisely beneath the non-volcanic region  
111 between Aso and Kirishima volcanoes (e.g. Park et al., 2009; Fig. 1b). The slab has been  
112 subducting aseismically, down to a depth of 430 km beneath Kyushu Island (Zhao et al.,  
113 2012; Fig. 1b).

114 P- and S-wave seismic tomography images identify a low-velocity zone along the  
115 subducting slab down to a depth of 300 – 400 km, but with no contrast between the two  
116 segments (Zhao et al., 2012; Liu and Zhao, 2016). On the other hand, the receiver function  
117 tomography shows dehydration at much shallower depths, with hydrated oceanic crust  
118 present to a depth of 70 km beneath Southern Kyushu, but only reaching 50 km beneath  
119 Northern Kyushu (Abe et al., 2013). This suggests extensive dehydration beneath the  
120 fore-arc region in the north. Lastly, numerical simulations place the location of the low-  
121 velocity zone beneath the volcanic front to fore-arc region in Northern Kyushu and  
122 beneath the volcanic front to the back-arc side in the case of central to Southern Kyushu.  
123 This north-south dichotomy is due to the contrasting thermal state of the slab because of

124 its age variation (Iwamori, 2007).

125 Low-velocity and high-Poisson's ratio anomalies are identified in the upper mantle  
126 beneath the back-arc region (Huang et al., 2013; Asamori and Zhao, 2015). This  
127 anomalous region extends from the Tsushima Strait in the north (between the Korean  
128 peninsula and Kyushu Island) to the southern edge of Kyushu Island. This anomaly  
129 extends beneath the back-arc to a depth of more than 200 km, reaching below the slab. It  
130 potentially indicates the existence of mantle upwelling, induced by a slab window. This  
131 mantle upwelling, originating beneath the Philippine Sea plate, is possibly caused by a  
132 deep dehydration reaction of the Pacific slab located below (at depths of 600-670 km),  
133 and suggests the presence of convective circulation above the stagnant Pacific slab, with  
134 the mantle flowing across the Philippine Sea plate via a window or around its leading  
135 edge. Such "big mantle wedge" convection is thought to be the cause of the continental  
136 rift systems and intraplate volcanism in East Asia (Tatsumi et al., 1990; Zhao et al., 2009;  
137 Zhao and Tian, 2013).

138

### 139 ***Volcanism***

140 The modern volcanism in Kyushu Island is grouped into three main volcanic  
141 provinces based on geographical distribution and geochemical signature: Northern  
142 Kyushu, Southern Kyushu, and the back-arc region. The volcanic activity in the frontal-  
143 arc of Northern Kyushu is characterized by hot-subduction zone magmatism, such as the  
144 eruption of adakitic-magmas from Yufu-Tsurumi volcano (YF in Fig. 1b; Sugimoto et  
145 al., 2006; Shibata et al., 2014) and the calc-alkaline high-magnesium andesites (6 – 3 Ma;  
146 Nakada and Kamata, 1991; Kakubuchi et al., 1995). Also in the north, high-alumina  
147 island-arc basalts are found in Kuju (KJ) and Aso (AS) volcanoes from 2 – 0 Ma (Nakada  
148 and Kamata, 1991; Mahony et al., 2011). The temporal evolution of the source mantle,  
149 showing progressive increase of the slab component, potentially explains the variations  
150 in major and trace element signatures of the area (Nakada and Kamata, 1991). Oninomi  
151 (ON) basalt is an exception with an adakite and OIB-like affinity (i.e. high Sr/Y no HFSE  
152 depletion), and an eruption age of 16 ka (Ohta et al., 1992; Shitaoka et al., 2014).

153 In Southern Kyushu, the modern activity is characterized by calc-alkaline magma  
154 for both the back-arc (7.6 – 0.4 Ma; Nagao et al., 1999) and the volcanic front, where

155 Kirishima (KR) Sumiyoshi-ike (SM), Sakurajima (SK) and Kaimondake (KM) volcanoes  
156 are located (Mahony et al., 2011). Alkali basalts erupted at ~1 Ma were reported in the  
157 back-arc side (Uto and Uchiumi, 1997; Kita et al., 2012) and at 0.08 Ma (Aojiki cone  
158 adjacent to Sumiyoshi-ike volcano, Kita et al., 2012). In the back-arc region, alkali to  
159 tholeiitic basalts have erupted episodically from ~15 Ma (Mahony et al., 2011). Onidake  
160 volcano on Fukue Island (FK, Fig. 1b; referred to as Fukue hereafter) is a typical active  
161 back-arc volcano in this region. Both alkali basalts and calc-alkaline andesites erupted at  
162 similar ages, in a zone between the frontal-arc and back-arc regions, such as Unzen  
163 volcano (UZ, Fig. 1b) and the adjacent area, (Sugimoto et al., 2005; Kita et al., 2001).

164 Magmatic H<sub>2</sub>O concentrations are estimated to be 3.6 – 6.0 wt. % for Northern  
165 Kyushu volcanoes, calculated from the observed high-An plagioclase and whole rock  
166 compositions (Zellmer et al., 2012). This indicates that the primary basaltic magma was  
167 potentially water-rich, although source lithology and slab-derived fluid/melt  
168 characteristics are poorly constrained. Miyoshi et al. (2008ab; 2010) determined boron  
169 concentrations in basaltic rocks and identified an increase in the B/Nb ratio from Yufu  
170 toward Kirishima, that they interpreted as an increase in slab-derived fluid contribution.  
171 Similarly, partial melting of the subducted young Philippine Sea plate has been suggested  
172 based on the systematics of its adakitic signature and Sr-Nd-Pb isotope ratios of rocks in  
173 Northern Kyushu (Sugimoto et al., 2006; Shibata et al., 2014).

174

## 175 **Samples and Methods**

### 176 *Sample description*

177 Nine olivine-bearing rocks were selected from 49 samples, covering major active  
178 volcanoes of Kyushu Island (Fig. 1ab and Table 1). We focused on lapilli-sized samples  
179 to have a higher chance of finding quenched volcanic products (thus avoiding daughter  
180 mineral crystallization within melt inclusions) and to minimize the effect of the diffusive  
181 volatile-loss (Danyushevsky et al., 2002; Lloyd et al., 2013). An exception was made for  
182 Yufu volcano, where we collected a block of lava from a pyroclastic flow deposit (YF1L)  
183 since no olivine-bearing tephra was found. The inclusions of Kujū (KJ5S), Aso (KSS),  
184 Kirishima-Ohachidake (KROHTh), Kirishima-Shinmoedake (KRSM11), Sumiyoshi-ike  
185 (SM1S), Kaimondake (KM1S12a) and Fukue (FKONON) were glassy while those of

186 YF1L were completely crystallized. In the case of Oninomi (ON2S), the inclusions hosted  
187 in the core of the olivines were mostly rounded and crystallized, while glass found at the  
188 rim of skeletal olivines tended to be clear, tubular and often open to the outside of the  
189 host crystal. Although the olivines of FKONON were also skeletal, the melt inclusions  
190 were fully enclosed. Complete descriptions of the appearance and size of the samples are  
191 given in Supplementary material Table S1 and document S2, following the recommended  
192 practices for melt inclusion studies (Rose-Koga et al., 2021).

193 Whole rock sample descriptions are from the literature and brief descriptions are  
194 summarized here: YF1L is a mafic enclave found in a Nonokusa pyroclastic-flow deposit  
195 on the southern slope of the edifice (Ohta et al., 1990; Okuno et al., 1999), which is  
196 interpreted as being equivalent to the mafic endmember of the Yufu-Tsurumi volcanic  
197 group (Ohta and Aoki, 1991). The olivine-bearing mafic blocks are typically vesiculated  
198 augite-hornblende-plagioclase basalt to basaltic andesite. ON2S is a coarse black scoria  
199 forming the edifice of Oninomi scoria cone. Its composition ranges from basalt to basaltic  
200 andesite containing plagioclase, olivine, augite, occasional orthopyroxene, destabilized  
201 hornblende, and Ti-Fe oxides. It also contains abundant granitic xenoliths (Ohta et al.,  
202 1992). KJ5S is the Hiijidake scoria, a brown scoria fall deposit, erupted from the Hiijidake  
203 cone of Kuju volcano. It is a basaltic andesite containing plagioclase, clinopyroxene,  
204 orthopyroxene, olivine (Kamata, 1997; Nagaoka and Okuno, 2014; Kawanabe et al.,  
205 2015). KSS is the Kishimadake scoria, a coarse black scoria fall deposit, erupted from the  
206 Kishimadake cone of Aso volcano (Miyabuchi and Watanabe, 1997), which is the same  
207 sample as in Kawaguchi et al. (2021) and identified as the mafic end member of Holocene  
208 Aso eruptions. KROHTH is the Takaharu scoria, a black vesiculated basaltic andesite  
209 scoria fall deposit, erupted from Ohachidake cone, from the Kirishima volcanic group,  
210 which corresponds to the fall unit ThT of Tsutsui et al. (2007). The related lava unit  
211 contains plagioclase, clinopyroxene, orthopyroxene and olivine. KRSM11 is a mixed  
212 andesitic pumice erupted in 2011 from Shinmoedake cone, from the Kirishima volcanic  
213 group, and corresponds to Unit 2 of Miyabuchi et al. (2013), whose bulk composition is  
214 andesitic, containing phenocrysts of plagioclase, clinopyroxene, orthopyroxene, olivine,  
215 magnetite and ilmenite. SM1S is a fine to coarse black scoria produced by a  
216 phreatomagmatic eruption of Sumiyoshi-ike volcano (Moriwaki et al., 1986). It is a  
217 basaltic scoria with olivine, plagioclase and clinopyroxene phenocrysts and glassy

218 groundmass with microlites. KM1S12a is a lithic-rich dense to vesiculated scoria  
219 containing plagioclase, clinopyroxene and olivine phenocrysts, and corresponds to the  
220 fall unit Km12a of Fujino and Kobayashi (1997), erupted from Kaimondake volcano  
221 (Kawanabe and Sakaguchi, 2005); FKONON is a vesiculated coarse scoria, and  
222 corresponds to the Onidake scoria fall unit of Nagaoka and Furuyama (2004), erupted  
223 from the Onidake cone, from Fukue volcanic group.

224

### 225 ***Melt inclusion preparation***

226 Olivine-hosted melt inclusions of these nine samples were prepared for chemical  
227 analysis. Loose olivine crystals (250 to 630  $\mu\text{m}$ ) were hand-picked from crushed and  
228 sieved samples. The dimensions of the inclusions and vapor bubbles were measured under  
229 a microscope (Table S1). Crystallized inclusions of YF1L and ON2S were homogenized  
230 using a Vernadsky-type microscope heating stage following the method described by Le  
231 Voyer et al. (2010). A detailed description of the heating procedure is given in  
232 Supplementary material document S1. Fully enclosed inclusions were exposed using a  
233 silicon carbide mat for coarse polishing, and corundum mats (3  $\mu\text{m}$  and 1  $\mu\text{m}$  mats) were  
234 used to complete the polishing. To avoid possible carbon contamination, diamond-based  
235 paste was avoided. Polished olivine crystals were subsequently mounted in indium for  
236 analysis, and a final polish with a 0.25  $\mu\text{m}$  alumina paste was performed to remove  
237 scratches on the exposed surface.

### 238 ***Analytical methods***

239 Volatile element concentrations ( $\text{H}_2\text{O}$ ,  $\text{CO}_2$ , S, Cl, and F) in the inclusions were  
240 measured by SIMS (Cameca IMS-1280HR of Kochi Institute of Core Sample Research,  
241 JAMSTEC, Japan) following the procedure described by Shimizu *et al.* (2017),  
242 comparable to that of Rose-Koga et al. (2020) for F, S and Cl. In brief, we used a 20 keV  
243  $\text{Cs}^+$  primary ion beam of 300 – 500 pA defocused to be 10 – 15  $\mu\text{m}$  in diameter.  
244 Secondary ions were accelerated at 10 kV. Then a 10 keV electron beam with a diameter  
245 of  $\sim 100 \mu\text{m}$  was applied for electrostatic charge compensation over the area of the Cs  
246 beam spot. The field aperture was set to the size corresponding to 5 x 5  $\mu\text{m}$  on the sample  
247 surface. Mass resolving power was set at  $\sim 6000$  to separate mass interferences (for  
248 example, to separate  $^{34}\text{S}^1\text{H}$  interference from  $^{35}\text{Cl}$  requires 5120 MRP; Burdo and

249 Morrison 1971). Negative secondary ions of  $^{12}\text{C}$ ,  $^{16}\text{OH}$ ,  $^{19}\text{F}$ ,  $^{30}\text{Si}$ ,  $^{31}\text{P}$ ,  $^{32}\text{S}$ , and  $^{35}\text{Cl}$  and  
250 the mass position of 11.9 amu were measured by an axial electron multiplier using the  
251 peak switching method. Each analysis consisted of 10 cycles, and the total measurement  
252 time was  $\sim 6$  min. Typical relative standard deviations ( $1\sigma$ ) obtained from repeated  
253 analysis of a secondary basaltic glass standard from the East Pacific Rise, EPR-G3  
254 (Shimizu et al., 2017; Supp. Table S9), were 1.4, 3.2, 0.9, 2.5 and 1.5% for  $\text{H}_2\text{O}$ ,  $\text{CO}_2$ , S,  
255 Cl and F, respectively, and the measured values of EPR-G3 usually agreed with the  
256 known value. In the event that the secondary standard value deviated significantly, the  
257 calibration curves were revised.

258 Sulfur isotopes in the inclusions were measured by SIMS at 1) Kochi Institute of  
259 Core Sample Research, JAMSTEC, Japan (Cameca IMS-1280HR) and 2) Centre de  
260 Recherches Pétrographiques et Géochimiques (CRPG)-CNRS-Nancy, France (Cameca  
261 IMS-1270). At Kochi, analyses were conducted following the procedure described by  
262 Shimizu et al. (2019). To summarize, we used a 20 keV  $\text{Cs}^+$  ion beam of  $\sim 0.5$  nA  
263 defocused to be  $\sim 10$   $\mu\text{m}$  in diameter. A 10 keV electron beam of  $\sim 100$   $\mu\text{m}$  diameter was  
264 used to flood the sample surface for electrostatic charge compensation. Secondary ions  
265 were accelerated to 10 kV. The field aperture was set at a size corresponding to  $15 \times 15$   
266  $\mu\text{m}^2$  on the sample surface. Negative secondary ions of  $^{32}\text{S}$  and  $^{34}\text{S}$  were measured in  
267 multi-detection mode with an FC and axial EM, respectively. Mass resolving power was  
268 set at  $\sim 2200$  and  $\sim 5000$  for FC and EM detectors, respectively, to separate mass  
269 interferences of  $^{31}\text{P}^1\text{H}$  from  $^{32}\text{S}$  and of  $^{33}\text{S}^1\text{H}$  from  $^{34}\text{S}$ . Each measurement consisted of 20  
270 s pre-sputtering, 120 s auto-centering of  $^{32}\text{S}$  to the field and contrast apertures, and  $5 \text{ s} \times$   
271 50 cycles for measurements. The total measurement time for each analysis was  $\sim 7$   
272 minutes. At CRPG, the set-up was similar except we used a stronger primary beam of 3  
273 nA defocused to  $\sim 15$   $\mu\text{m}$  in diameter. Mass resolving power was set to 4500. Negative  
274 secondary ions of  $^{32}\text{S}$  and  $^{34}\text{S}$  were measured in the multi-detection mode with an FC and  
275 axial EM, respectively. Each measurement consisted of 60 s pre-sputtering and  $5 \text{ s} \times 40$   
276 cycles for measurements, giving a total analysis time of  $\sim 5$  minutes for each spot. Before  
277 each measurement, we adjusted the center for  $^{32}\text{S}$ . In both cases, the standard basaltic  
278 glass, EPR-G3 ( $\delta^{32}\text{S}_{\text{VCDT}} = -0.09 \pm 0.55\%$ , 2SD; Shimizu et al., 2019), was systematically  
279 measured during the analytical session to monitor EM drift and to define the instrumental  
280 mass fractionation factor (IMF) for  $^{34}\text{S}$ :  $\text{IMF} = (^{34}\text{S}/^{32}\text{S})_{\text{measured}} / (^{34}\text{S}/^{32}\text{S})_{\text{reference}}$ . At

281 CRPG, other standard glasses were analyzed with EPR-G3, and we did not find any  
282 matrix-dependent variation for IMF, unlike in the recent report of Taracsak et al. (2021)  
283 (further discussion in Supplementary document S1). Sulfur isotopes in the samples are  
284 expressed in  $\delta^{34}\text{S}$  relative to the Vienna Canyon Diablo Troilite (VCDT,  $^{34}\text{S}/^{32}\text{S} =$   
285  $1/22.6436$ ; Ding et al., 2001;  $\delta^{34}\text{S} = [(^{34}\text{S}/^{32}\text{S})_{\text{unknown}}/\text{IMF}/(^{34}\text{S}/^{32}\text{S})_{\text{VCDT}} - 1] \times 1000$ , in  
286 per mil). The analytical error ( $2\sigma$ ) obtained from repeated measurements of the EPR-G3  
287 standard during the analytical session was less than 1.2 ‰ for both SIMS laboratories,  
288 and the internal precision ( $2\sigma$ ) is also reported for individual analysis, with an average of  
289 0.8 ‰ (Supplementary material Table S4).

290 Major element compositions of melt inclusion and host-olivine phenocryst were  
291 determined using a CAMECA SX100 electron microprobe at the Laboratoire Magmas et  
292 Volcans, Clermont-Ferrand, France (LMV). The analytical settings for silicate glass were  
293 15 kV acceleration voltage, 4 nA beam current, with counting times at 30 s for Ti, at 20  
294 s for Si, Al, Fe, Mg, Na and K, and at 10 s for Mn, Ca, and P. For olivine, we used a 15  
295 nA beam current with counting time at 30 s for Ca, at 20 s for Si and Al, at 15 s for Ti  
296 and Cr, and at 10 s for Fe, Mn, Mg, and Ni. Beam diameter was set at 5 or 10  $\mu\text{m}$   
297 depending on the size of the inclusion, and focused for olivine measurements. Relative  
298 analytical uncertainties ( $1\sigma$ ) obtained from repeated measurements of the basaltic glass  
299 standard (VG-2, Juan de Fuca ridge basalt, Jarosewich et al., 1980; Dixon et al., 1991)  
300 were less than 0.6% for  $\text{SiO}_2$ , 1% for  $\text{Al}_2\text{O}_3$ , FeO and CaO, 3.5% for  $\text{TiO}_2$  and  $\text{Na}_2\text{O}$ ,  
301 10% for MnO, 1.5% for MgO, 4.5% for  $\text{K}_2\text{O}$ , 17% for  $\text{P}_2\text{O}_5$  (Supplementary Table S3).

302 In addition, the S, Cl and F concentrations of 12 melt inclusions were also  
303 determined, using a 40 nA beam current with counting times of 50 s for S and Cl, and  
304 200 s for F. For each analysis, an average of 5 measurements was taken. The standard  
305 deviations ( $1\sigma$ ) for S, Cl, and F were less than 143 ppm, 95 ppm, and 397 ppm,  
306 respectively. Further details of the analytical methods, accuracy and verification, are  
307 found in Rose-Koga et al. (2020).

308 Sulfur speciation of selected inclusions was determined by measuring a relative  
309 sulfur  $K\alpha$  peak position in a melt inclusion sample compared to the standard anhydrite  
310 mineral using the EPMA (Carroll and Rutherford, 1988), with an assumption that  $\text{S}^{2-}$  and  
311  $\text{S}^{6+}$  are the only two relevant species in silicate melt (e.g. Jugo et al., 2010). We took an  
312 average of three spots per melt inclusion, and the standard error ( $1\sigma$ ) was approximately

313  $\pm 10\%$   $S^{6+}/S^{\text{Total}}$  for samples with S concentration  $> 1000$  ppm, which is the case for 84%  
314 of the melt inclusions.

315 Trace element concentrations in melt inclusions were determined using a laser  
316 ablation system (193 nm Excimer Resonetics M-50E laser ablation system) coupled with  
317 an inductively coupled plasma mass spectrometer, Agilent 7500 cs (LA-ICP-MS) at LMV  
318 following the classical procedure described by Rose-Koga et al. (2012, 2017). Analyses  
319 were conducted with a laser pulse frequency of 1–3 Hz, a spot diameter between 15 and  
320 33  $\mu\text{m}$  depending on the MI size, and a fluence of 2.8  $\text{mJ}/\text{cm}^2$ . Twenty-six masses were  
321 collected:  $^7\text{Li}$ ,  $^{45}\text{Sc}$ ,  $^{51}\text{V}$ ,  $^{65}\text{Cu}$ ,  $^{85}\text{Rb}$ ,  $^{88}\text{Sr}$ ,  $^{89}\text{Y}$ ,  $^{90}\text{Zr}$ ,  $^{93}\text{Nb}$ ,  $^{137}\text{Ba}$ ,  $^{139}\text{La}$ ,  $^{140}\text{Ce}$ ,  $^{141}\text{Pr}$ ,  $^{146}\text{Nd}$ ,  
322  $^{147}\text{Sm}$ ,  $^{153}\text{Eu}$ ,  $^{157}\text{Gd}$ ,  $^{163}\text{Dy}$ ,  $^{166}\text{Er}$ ,  $^{172}\text{Yb}$ ,  $^{175}\text{Lu}$ ,  $^{178}\text{Hf}$ ,  $^{181}\text{Ta}$ ,  $^{208}\text{Pb}$ ,  $^{232}\text{Th}$ ,  $^{238}\text{U}$ . The internal  
323 reference mass was  $^{44}\text{Ca}$ , with the CaO concentration taken from the electron microprobe  
324 analysis. A typical signal acquisition started by collecting a background for 30s, which  
325 was followed by the laser firing for 70 s or less depending on the thickness of the  
326 inclusion. Data reduction was performed by the GLITTER software (GEMOC,  
327 Macquarie University, Australia). BCR-2G, NIST 610 and NIST 612 glasses (Gao et al.,  
328 2002; Gagnon et al. 2008) were also analyzed to monitor the reproducibility and accuracy  
329 of the analyses. Typical relative standard errors on samples ( $1\sigma$  error of mean;  $\sigma/\sqrt{n}$ ,  
330 where  $n$  is the number of cycles) were  $< 5\%$  for V, Rb, Sr, Y, Zr, Ba, La, and Ce,  $< 8\%$   
331 for Sc, Nb, Pr, and Nd,  $< 15\%$  for Cu, Sm, Eu, Gd, Dy, Er, Hf, Pb, Th, and U,  $< 25\%$  for  
332 Li, Yb, Lu, and Ta.

### 333 **Correction for post-entrapment modification**

#### 334 *Post-entrapment crystallization*

335 Major, volatile and trace element concentrations of melt inclusions were corrected  
336 for post-entrapment crystallization (PEC; e.g., Danyushevsky et al., 2000) by incremental  
337 addition of equilibrium olivine (0.1 wt. % step) into the melt, until it reached equilibrium  
338 with the host olivine (Toplis, 2005). We adopted a constant  $\text{Fe}^{2+}/\text{Fe}^{\text{Total}}$  ratio calculated  
339 from the empirical equation of Kilinc et al. (1983) from the averages of measured  $SK\alpha$   
340 peak shifts. For example, a single  $f\text{O}_2$  value was assumed to represent a rock sample (Jugo  
341 et al., 2010). Olivine-melt equilibrium temperatures were calculated using the olivine-  
342 saturated melt thermometer of Sugawara (2000), corrected for the effect of  $\text{H}_2\text{O}$  on the

343 olivine liquidus temperature (Médard and Grove, 2008). The correction for post-  
344 entrapment Fe-Mg exchange (also called “diffusive Fe-loss”; Danyushevsky et al., 2000)  
345 was not necessary (see Supplementary material document S1), because the FeO  
346 concentration in a melt inclusion plots along the compositional trend defined by the whole  
347 rock data. Concentration of volatile and trace elements was adjusted by assuming their  
348 perfect incompatibility in the host olivine (Table S2). The corrected concentrations are  
349 used throughout this paper (Table S8). Raw data are reported in Supplementary Tables  
350 S3, S4 and S5). It should be noted that PEC added less than 6 % of olivine to melt, for  
351 95% of the sample.

### 352 *Vapor-bubble formation*

353 In general, more than 40 – 90% of the initially dissolved CO<sub>2</sub> in the melt at the time  
354 of entrapment is lost to shrinkage bubbles, when vapor bubble growth occurs within an  
355 inclusion (e.g. Wallace et al., 2015). Therefore, the total CO<sub>2</sub> concentration in a melt  
356 inclusion was determined as (dissolved + exsolved),

$$357 \quad CO_2^{Total} = CO_2^{Melt} + \phi \frac{\rho_v}{\rho_m} \times 10^6$$

358 where CO<sub>2</sub><sup>Melt</sup> is the dissolved CO<sub>2</sub> concentration in the melt,  $\phi$  is volume fraction of the  
359 bubble in the melt inclusion,  $\rho_v$  is the volumetric CO<sub>2</sub> vapor density, and  $\rho_m$  is the melt  
360 density, calculated from the glass composition. The  $\phi$  was determined based on  
361 measurements of the two visible axes, a and b, under the microscope (Table S1) assuming  
362 the length of the third axis as  $c = (a + b) / 2$  (e.g. Tucker et al., 2019). The  $\rho_v$  value was  
363 estimated using RhyoliteMELTS ver. 1.2.0 (Ghiorso and Sack, 1995; Gualda et al., 2012;  
364 Ghiorso and Gualda, 2015), based on CO<sub>2</sub>-H<sub>2</sub>O solubility at the glass transition  
365 temperature, T<sub>glass</sub>, and an equation of state (EOS) for the fluid (Duan and Zhang, 2006).  
366 The T<sub>glass</sub> value was calculated from the temperature dependence of the viscosity at the  
367 given hydrous glass composition and a constant viscosity of 10<sup>12</sup> Pa·s (Giordano et al.,  
368 2005, 2008). The total CO<sub>2</sub> concentrations were first calculated using the measured glass  
369 composition and the equation above, and they were then corrected for PEC in the same  
370 way as for the other volatile elements. This EOS calculates  $\rho_v$  with an assumption that the  
371 bubble and melt are in equilibrium until the bubble ceases to expand. Since carbon (C)  
372 diffusion is temperature dependent, if the diffusive closure temperature is effectively  
373 higher than T<sub>glass</sub>, the bubble would expand without C exsolution, resulting in an

374 overestimation of  $\rho_v$  and of the total CO<sub>2</sub> concentration (e.g. Pichavant et al., 2013; Bali  
375 et al., 2018). If this non-equilibrium expansion has occurred, the estimated total CO<sub>2</sub>  
376 concentration represents a maximum value.

377 We conducted an additional test based on the method of Moore et al. (2015), which  
378 calculates the expected vapor bubble volume in a melt inclusion to assess the extent of  
379 the thermal contraction of the melt and host olivine due to post-entrapment cooling. This  
380 calculation provides a mechanical upper limit of the bubble size formed by post-  
381 entrapment cooling. For example, the test concludes that a melt inclusion contained a pre-  
382 entrapment bubble if a measured bubble volume fraction exceeds the expected value. In  
383 this case we did not apply the “CO<sub>2</sub> bubble correction”. This is because the calculation of  
384 total CO<sub>2</sub> is strongly dependent on the bubble volume fraction. Therefore, if a pre-  
385 entrapment bubble had been present, because the observed bubble volume would have  
386 been larger than the expected shrinkage bubble, it would result in an overestimation of  
387 the total CO<sub>2</sub>.

388 The major source of error of the total CO<sub>2</sub> in the inclusions is the uncertainty of the  
389 melt inclusion volume, as the volume is estimated assuming an ellipsoid with  
390 measurements of only two axes. A recent numerical simulation reports that the  
391 assumption taking an arithmetic mean for the third ellipsoid axis yields a relative error  
392 ( $1\sigma$ ) of 37% to 48% (Tucker et al., 2019). We conservatively assigned a 50 % ( $1\sigma$ ) relative  
393 uncertainty to inclusion volumes. The uncertainties ( $1\sigma$ ) on the total CO<sub>2</sub> concentration  
394 therefore ranged from 10 to 50% after accounting for the volume error (Table S6).

## 395 **Results**

### 396 *Major element compositions*

397 The melt inclusions in this study varied from basalt to andesite with SiO<sub>2</sub> ranging  
398 from 40.3 to 60.7 wt. % (Table S2, Fig. 2). Fo content [ $\text{Mg} / (\text{Fe} + \text{Mg}) \times 100$  in mole] of  
399 host olivine varied from 65.2 to 83.1 (Table S7). The composition of melt inclusions  
400 formed tight clusters for each tephra sample, except Aso and Oninomi. The melt  
401 inclusions of Fukue, Yufu and Sumiyoshi-ike were classified as alkali-basalt to  
402 microbasalt and basanite. Those of Kuju, Kirishima-Ohachidake, Kirishima-Shinmoedake  
403 and Kaimondake were sub-alkaline basaltic andesites. Melt inclusions of Aso exhibited  
404 a wide range of SiO<sub>2</sub> and were classified as transitional basalt and sub-alkaline basaltic

405 andesite to andesite (Kawaguchi et al., 2021). The melt inclusions of Oninomi varied  
406 from basanite to basalt, except for one andesite inclusion. Basanite magmas are rare  
407 around Kyushu Island especially at the volcanic front (Iwamori, 1991, 1992). Similar  
408 basanite rocks are found in the northern part of the Izu-Bonin volcanic arc (Oshika et al.,  
409 2014) and in primitive rocks from the central Chugoku district, SW Japan (Iwamori, 1991,  
410 1992). The subalkaline melt inclusions from Northern Kyushu were higher in total alkali  
411 ( $\text{Na}_2\text{O} + \text{K}_2\text{O}$ ) compared to those from the Southern Kyushu for the same  $\text{SiO}_2$   
412 concentration.

### 413 *Trace element compositions*

414 The melt inclusions in this study were enriched in large-ion lithophile elements  
415 (LILE) over high field strength elements (HFSE) resulting in high LILE/HFSE values  
416 (Fig. 3), characteristic of island arc basalts (IAB; e.g., Elliott, 2003). Positive K, Pb and  
417 Sr anomalies were present. Positive Sr anomalies in samples from the Northern Kyushu  
418 (Yufu, Kuju, Aso) were stronger than those from Southern Kyushu (Kirishima,  
419 Sumiyoshi-ike, Kaimondake). The trace element patterns of Fukue and Oninomi  
420 displayed no Nb-Ta negative anomalies and high Nb concentration ( $\text{Nb} \geq 23$  ppm), as  
421 found for the whole rock data (e.g., Ohta et al., 1992; Miyoshi et al., 2008). Melt  
422 inclusions from Oninomi had higher light rare earth element concentrations (LREE), and  
423 lower HREE compared to those from Fukue (Fig. 3). Two Oninomi melt inclusions were  
424 4 to 25 times richer in Ba than the other melt inclusions.

425

### 426 *Volatile element and sulfur isotope compositions*

427  $\text{H}_2\text{O}$  and  $\text{CO}_2$  concentrations in melt inclusions were corrected for PEC and post-  
428 entrapment vapor bubble growth, and the corrected values were used throughout this  
429 paper.  $\text{H}_2\text{O}$  concentrations varied between 0.01 and 6.06 wt. % (Table S2), and there was  
430 no systematic difference between Northern and Southern Kyushu.  $\text{H}_2\text{O}$  concentrations  
431 within the same tephra sample varied less than 1 or 2 wt. % (Fig. 4).  $\text{CO}_2$  concentrations  
432 ranged from a few ppm to 1.2 wt.% (Fig. 4). S, Cl and F concentrations varied from 142  
433 to 3800 ppm, from 89 to 1440 ppm, and from 55 to 1170 ppm, respectively (Table S2).  
434 The high volatile contents indicate successful sampling of less-degassed, near-primitive  
435 volatile in magmas.

436 Chlorine concentrations in the melt inclusions varied from 89 to 1440 ppm but with  
437 less than 7% of the melt inclusions with Cl <500ppm (8 out of 117; Table S2). Most Cl  
438 concentrations were higher than those of MORB (max ~500 ppm Cl; e.g. Le Voyer et al.,  
439 2015), indicating the addition of Cl to the sub-arc mantle from the subducting slab (e.g.,  
440 Straub and Layne, 2003). Similarly, F concentrations in melt inclusions (55 to 1170 ppm)  
441 were higher than those of MORB (71 to 576 ppm; e.g. Le Voyer et al., 2015), indicating  
442 a variable degree of F addition from the subducted slab. These elevated Cl and F values  
443 are characteristic of arc basalts (e.g., Rose-Koga et al., 2014; Van den Bleeken and Koga,  
444 2015).

445 Measured  $\delta^{34}\text{S}$  values ranged from  $-0.3 \pm 0.8 \text{ ‰}$  to  $+9.4 \pm 0.5 \text{ ‰}$  (Table S4), and  
446 were significantly higher than those of MORB ( $-0.91 \pm 0.50 \text{ ‰}$ , Kanehira et al., 1973;  
447 Sakai et al., 1984; Peters et al., 2010; Labidi et al., 2012, 2014), and within the range  
448 reported for bulk S isotope data of Japanese arc volcanic rocks (from  $-0.2$  to  $+18.3 \text{ ‰}$ ,  
449 Ueda and Sakai, 1984). Most samples have  $\delta^{34}\text{S} > +3.4 \text{ ‰}$  (Table S4), with Fukue and  
450 Oninomi showing relatively lower  $\delta^{34}\text{S}$  values ( $<+2.5 \text{ ‰}$  with one exception at  $+3.63 \text{ ‰}$ ;  
451 Table S4).

452

## 453 **Discussions**

454 The discussion is organized into six sections. The first three sections concern the  
455 determination of the primitive magmas of each volcano, for geochemical characteristics  
456 of major, trace, and volatile elements as well as sulfur isotopes: entitled “Major and trace  
457 element composition of the primitive magma”, “Volatile element concentration in the  
458 primitive magma”, and “S isotope composition of the primitive magma”. Then, the  
459 following two sections discuss the relationship between the geochemical characteristics  
460 of the primitive magmas and the geodynamic characteristics of the Kyushu Arc, which  
461 are entitled “The magma groups of Kyushu”, and “Geodynamic significance of the source  
462 regions”. Finally, the last section, “Sources of sulfur in the subduction zone”, outlines the  
463 contributions of sulfur isotope signatures to the arc.

464 Identification of the primitive magma makes it possible to characterize the magma  
465 source mantle, which is a mixture of mantle and slab-derived components in this case of  
466 arc volcanism. However, arc lavas are commonly modified through interaction with the

467 crust before eruption. An olivine-hosted melt inclusion is considered as being a droplet  
468 of magma trapped early in the growing crystal, and is generally assumed to have  
469 undergone negligible modification through crystal fractionation in the crust as olivine is  
470 typically the first liquidus phase in basalt. However, melt inclusions may not be  
471 completely free from modifications prior to entrapment. Therefore, it is necessary to  
472 exclude geochemical signals relating to any pre-entrapment crustal processes such as  
473 crystal fractionation, magma mixing, wall-rock assimilation, and pre-entrapment volatile  
474 element degassing, before interpreting the chemical variations of melt inclusion  
475 compositions. Once such an analysis has been done, the remaining variation represents  
476 the melting and metasomatic processes in the source region, and its relationship to the  
477 geodynamic setting.

#### 478 ***Major and trace element composition of the primitive magma***

479 Combinations of incompatible and compatible elements and their ratios are useful  
480 indicators for discriminating the effects of fractional crystallization, magma mixing and  
481 partial melting of source (e.g. Fourcade and Allegre, 1981; Schiano et al., 2010).  
482 Following the method of Schiano et al. (2010), we determine for each volcano, if the  
483 variation in melt inclusion composition is due to mixing, partial melting or crystal  
484 fractionation (Supplementary material document S1). In detail, some of the inclusions  
485 showed insignificant compositional variation within a sample (Fig. 5a), and those  
486 compositions were placed at the mafic end of whole rock variation (Fig. S4). In other  
487 cases, the inclusions showed variations of Ba/Nb ratio, which is a trace element ratio of  
488 two highly incompatible elements that does not vary during mantle partial melting and  
489 crystal fractionation in magma (Fig.5b). Overall, each sample appears to form either a  
490 distinct cluster (e.g. Kirishima, Kuju), or a trend (e.g. Kaimondake, Yufu) when plotted  
491 using different sets of major and trace elements (Fig. 5). From this initial analysis, we  
492 concluded that a distinct primitive magma (or magmas) exists for each sample, and we  
493 determined a primitive melt composition for each volcano (Table 2).

494 All inclusion samples, except for those of Fukue, show significant variations in  
495 trace element ratios, such as Rb/Nd and Th/Nd. This indicates that magma mixing and/or  
496 partial melting is the dominant process rather than fractional crystallization (Fig.S3) since  
497 these incompatible element ratios do not fractionate with early crystallization of phases

498 such as olivine, plagioclase, and pyroxene (see inset, Fig.S3). Inspecting the geochemical  
499 variations for individual volcanoes (Fig. S4), we found that the magmas of Yufu, Aso,  
500 Sumiyoshi-ike and Kaimondake showed geochemical systematics indicating magma  
501 mixing and/or crustal assimilation. On the contrary, fractionation of elements during  
502 partial melting was identified for the magmas of Kuju, Kirishima-OH and Kirishima-SM.  
503 Lastly, the variations of the final group, Fukue and Oninomi, were best explained by  
504 crystal fractionation (Fig. S3). Because the major and trace element data of Fukue formed  
505 a tight cluster, we chose the least evolved melt based on trace elements (i.e. lowest  
506 concentrations), as being representative of the primitive melt of Fukue (A-I, Fig. S3).

507 These primitive melt compositions have the lowest SiO<sub>2</sub> contents of the magma  
508 series for each volcano, with high MgO, and low incompatible element concentration  
509 (Table 2). Trace element ratios are expected to reflect the composition of mantle derived  
510 magma, but the Fo contents of the host-olivines were more evolved (from 73 to 83) than  
511 the expected mantle value of 89. This suggests that our “primitive magmas” have  
512 potentially suffered from pre-entrapment fractional crystallization and/or pre-entrapment  
513 magma mixing. Alternatively, low Fo content can be acquired through Fe-Mg exchange  
514 with an evolved magma while maintaining the trace element abundance at the time of  
515 entrapment (e.g. Cottrell et al., 2002; Rose-Koga et al., 2021). In this case, primitive trace  
516 element and isotope compositions are conserved in the inclusion.

517 Melt inclusions of Oninomi show a wide range of SiO<sub>2</sub> concentration from 42.7 to  
518 58.6 wt. %, with the variation being greater than that reported for the bulk rocks (51.0 –  
519 54.5 wt. % SiO<sub>2</sub>, Ohta et al., 1992). While 10 out of the 11 inclusions have less than 48.5  
520 wt. % SiO<sub>2</sub>, one melt inclusion has an andesitic composition (U-I, 58.6 wt. % SiO<sub>2</sub>, Table  
521 S2). This andesitic inclusion is tubular and glassy, trapped in the rim of an olivine,  
522 indicating a late-stage entrapment during the crystallization of a cooler magma (Table S1  
523 and Supplementary document S2). We exclude this inclusion during the rest of the  
524 discussion, because of its texture, and the reported evidence of crustal assimilation in  
525 Oninomi basaltic lava, such as the presence of granitic xenoliths and disequilibrium  
526 quartz phenocrysts (Ohta et al., 1992), and a high <sup>87</sup>Sr/<sup>86</sup>Sr ratio (0.704821±0.000011,  
527 Kita et al., 2001; Shibata et al. 2014). Similarly, the anomalous enrichment of Ba, and  
528 LREE found in the two melt inclusions (L-I and DZ-I) is again inferred to be indicative  
529 of crustal assimilation (Fig.3). The remaining inclusions show nearly identical trace

530 element patterns (Fig. 3), suggesting a similar origin. We chose the least evolved melt in  
531 this group as the primitive melt composition to represent Oninomi volcano (Z-I).

532 The melt inclusions from Aso volcanoes (KSS), especially the ones from the  
533 eruptions of the last 3700 years, have been described previously, and showed  
534 compositional variations and phenocryst textures consistent with magma mixing  
535 (Kawaguchi et al., 2021). Typically, evolved melt inclusions were trapped in reversely  
536 zoned olivines, with lower Fo content in the core ( $< 72$ ) than the rim (68-78), while  
537 primitive melts were trapped in normally zoned olivine. The data in this study corroborate  
538 such a conclusion. As shown in Fig.S4, the melt inclusions and reported bulk rock data  
539 of Aso volcano indicate magma mixing (Fig. S4). For Aso (KSS), the least evolved melt  
540 of the mafic endmember is taken as being closest to the primitive composition (K-II).

541 There is no previous trace element data reported for Kaimondake volcano, and this  
542 limited our “test” to find definitive conclusions on the systematics of its magma  
543 composition. Kaimondake illustrates a weak concave curve on Rb versus Rb/V and Rb  
544 versus Rb/Sr plots, indicating magma mixing between a mafic magma and a more evolved  
545 magma (Fig.S4). Based on this, the composition of a melt inclusion from the mafic  
546 endmember was chosen as the primitive magma composition (J-I).

547 Melt inclusions of Sumiyoshi-ike (SM1S) show a variation requiring three  
548 component mixing (Supplementary material document S1). First, significant variations  
549 in  $K_2O$  (or  $K_2O/TiO_2$ ), Rb and Ba/Nb are seen for a given  $SiO_2$  (Fig. 5, and Fig. S5),  
550 indicating the presence of at least two distinct parental magmas since such incompatible  
551 element variations cannot be generated by fractional crystallization or partial melting of  
552 a single source. Furthermore, the plot of Ba/Nb against  $K_2O/TiO_2$  (or Rb/Sr) shows that  
553 a third component is required to explain the compositional variation of the inclusions  
554 (Fig. S5ab). One component appears to be granite, added via crustal assimilation, as  
555 already identified by Pb isotope systematics (Brown et al., 2020). The other two  
556 endmembers are the low  $K_2O/TiO_2$  with the lowest Ba/Nb endmember (S-II), and the low  
557  $K_2O/TiO_2$  with the highest Ba/Nb (AA-I). These are considered to be the two  
558 endmembers of the primitive magmas.

559 For Yufu volcano (YF1L), a negative linear correlation between  $SiO_2$  and  
560 incompatible element concentrations, such as  $K_2O$  and Rb, indicates the mixing of two  
561 magmas. This correlation is independent of the mixing reported from the bulk rock

562 chemical variations between a basaltic and a dacitic magma (Fig. 5 and Fig. S4, Ohta et  
563 al., 1990; Ohta and Aoki, 1991). Furthermore, this negative correlation cannot be  
564 generated by crustal assimilation, since the partial melt of a crust is SiO<sub>2</sub>-rich melt (e.g.  
565 granite) with high incompatible element concentrations (e.g., DePaolo, 1981). Melt  
566 inclusions of Yufu show a significant variation in Ba/Nb, which correlates inversely with  
567 SiO<sub>2</sub> concentration. The lack of correlation with K<sub>2</sub>O/TiO<sub>2</sub> or CaO/Al<sub>2</sub>O<sub>3</sub> ratios (not  
568 shown) indicates the Ba/Nb variations are not due to the assimilation of plagioclase or of  
569 crustal rocks. Therefore, the geochemical systematics of Yufu melt inclusions indicate  
570 mixing, and the two endmembers have two distinct primitive melt compositions: high and  
571 low Ba/Nb (we selected BQ-III and BD-I, respectively).

572 Magmas of Kirishima (KROHTH, KRSM11) and Kuju (KJ5S) show geochemical  
573 systematics consistent with a variation of degrees of partial melting from a single source  
574 (Fig.S4). For Kirishima volcano, this interpretation is consistent with whole rock studies  
575 showing the existence of two parental magmas generated by different degrees of partial  
576 melting of a similar source (Inoue, 1985; Miyamoto, 1997). The SiO<sub>2</sub> variations are so  
577 restricted that we decided to rely on trace elements for the choice of the most primitive  
578 compositions. We chose the melt inclusions with the consistently lowest trace element  
579 concentrations (i.e. highest degree of partial melting) representing the primitive magma  
580 compositions for Kuju (AD-V) and Kirishima (Ohachidake, G-I and Shinmoedake, E-II).  
581

### 582 *Volatile element concentrations in the primitive magma*

583 Volatile concentrations, especially the concentration of H<sub>2</sub>O and CO<sub>2</sub>, in primitive  
584 arc magmas are critical geochemical parameters that can constrain models of magma  
585 genesis. While it has been studied extensively, the current consensus is that it would be  
586 highly improbable to be able to measure the primitive H<sub>2</sub>O and CO<sub>2</sub> content directly from  
587 geological materials. In fact, because of the high-diffusivity of H<sub>2</sub>O in host-olivines,  
588 measured H<sub>2</sub>O concentrations of melt inclusions likely equilibrated at crustal magma  
589 chamber depths (Portnyagin et al., 2008; Chen et al. 2011; Gaetani et al., 2012; Ferriss et  
590 al., 2018). Nevertheless, the H<sub>2</sub>O concentrations trapped in melt inclusions are higher  
591 than those of erupted lava, and it is thus considered to provide a minimum estimate. On  
592 the contrary, the other volatile elements, such as CO<sub>2</sub>, S, Cl and F, do not diffuse out

593 through the host-olivine within the lifetime of a magma (e.g. Bucholz et al., 2013) and  
594 they retain the values at the time of entrapment. It should be noted that if CO<sub>2</sub> degassing  
595 starts before entrapment of the melt in the host olivine, the primitive CO<sub>2</sub> composition  
596 cannot be trapped in a melt inclusion.

597 In this study, several methods were combined to estimate the minimum H<sub>2</sub>O content  
598 in the primitive magmas of each edifice and the values are reported in Table 2. First, the  
599 melt inclusions from Kuju, Kirishima, and Kaimondake displayed H<sub>2</sub>O values from 2.8  
600 to 6.1 wt. % (Table S2). These would commonly qualify as “high-H<sub>2</sub>O” values for the arc  
601 magma (e.g. see compilation in Cooper et al., 2012). Alternatively, the H<sub>2</sub>O content in  
602 magma can be estimated using a plagioclase-hygrometer (Lange et al., 2009; Waters and  
603 Lange, 2015) in combination with an olivine-liquid thermometer (Sugawara, 2000;  
604 Médard et al., 2008) with a typical uncertainty of 23 °C and 0.6 wt. % H<sub>2</sub>O (Sakuyama et  
605 al., 2014). It should be noted that the hygrometer records the water content in the magma  
606 at the time of plagioclase crystallization, which may happen after melt entrapment in  
607 olivine. Again, this is a minimum H<sub>2</sub>O estimate for the primitive magma. For example,  
608 for the case of Aso, the plagioclase hygrometer gives an H<sub>2</sub>O concentration of 4.7 wt.%,  
609 while measured H<sub>2</sub>O concentration in melt inclusions is approximately 3.0 wt. % (Table  
610 S2, and Kawaguchi et al., 2021). In this case, 4.7 wt. % is taken as the minimum primitive  
611 H<sub>2</sub>O content.

612 Another method for estimating H<sub>2</sub>O content in magma is to adopt the maximum  
613 H<sub>2</sub>O/K<sub>2</sub>O ratio of the melt inclusions (Fig. 6a). As these elements are incompatible to  
614 silicate minerals in basalt, the ratio does not vary with crystallization. The highest ratio  
615 should indicate the minimum H<sub>2</sub>O loss, if a magmatic suite shares the same parental  
616 magma. In the case of mixed magmas, H<sub>2</sub>O/K<sub>2</sub>O of the mafic endmember likely defines  
617 the least degassed ratio. This K<sub>2</sub>O normalization also eliminates the effect of post-  
618 entrapment crystallization. Specifically, we looked for the highest H<sub>2</sub>O/K<sub>2</sub>O values of the  
619 melt inclusions in this study and the whole rock H<sub>2</sub>O/K<sub>2</sub>O from previous studies, in which  
620 H<sub>2</sub>O content from the plagioclase hygrometer is combined with measured K<sub>2</sub>O  
621 concentration (Table 3; Zellmer et al., 2012; 2014; Kawaguchi et al., 2021). The H<sub>2</sub>O  
622 concentration is then calculated by multiplying the highest H<sub>2</sub>O/K<sub>2</sub>O ratio by the K<sub>2</sub>O  
623 concentration of the selected primitive melt (Table 2). This approach is especially  
624 effective for the samples from Oninomi, Yufu, Aso, and Kirishima samples, as H<sub>2</sub>O in

625 the inclusions was re-equilibrated to low values. It should be noted that the highest  
626 H<sub>2</sub>O/K<sub>2</sub>O ratios among the melt inclusions for each edifice correspond to those of the  
627 selected primitive ones identified in the previous section (Table S2), confirming the least  
628 degassed characteristic of our chosen primitive magma compositions. The exceptions  
629 were for Kuju, Yufu, and Sumiyoshi-ike, for which the highest H<sub>2</sub>O/K<sub>2</sub>O is not that of  
630 the selected inclusions representing primitive magma compositions. For Kuju, the  
631 difference in H<sub>2</sub>O/K<sub>2</sub>O between the highest melt inclusion and the selected primitive  
632 endmember was within measurement uncertainty, so we chose the value of the  
633 endmember (KJ5S-AD-V). Yufu and Sumiyoshi-ike melt inclusions both have two  
634 parental magmas (see previous section) possibly indicating two sources with different  
635 H<sub>2</sub>O concentrations; H<sub>2</sub>O/K<sub>2</sub>O of the magma cannot be assumed to be constant  
636 throughout mixing. In addition, judging from the low H<sub>2</sub>O concentrations, these melt  
637 inclusions lost a significant amount of their initial water. While we used the same  
638 approach to present the highest H<sub>2</sub>O concentrations, those of Yufu and Sumiyoshi-ike  
639 remain truly minimum estimates.

640 We used the maximum CO<sub>2</sub> concentration of each sample as the minimum estimate  
641 for the primitive value, using a similar reasoning as for H<sub>2</sub>O. We considered the bubbles  
642 present in melt inclusions as an exsolved fluid phase, and “added the CO<sub>2</sub> back” to the  
643 melt using the equation of state method (e.g., Tucker et al., 2019). The CO<sub>2</sub> content in the  
644 melt inclusions is reported in Table S2, and the comparison before and after the correction  
645 is displayed on Figure 4. The highest CO<sub>2</sub> content is found in the alkali basalt of Fukue,  
646 reaching 1.2 wt. %. Taking these corrected values, nearly all melt inclusions were formed  
647 at depths exceeding 10 km (Figure 4). The highest entrapment pressure determined using  
648 the volatile content of an inclusion is 12 kb (Fukue), and 7.4 kb (Aso) for the inclusions  
649 from the volcanic front.

650 Cl and F concentrations in primitive magmas are likely representative of primary  
651 concentrations, since these elements exsolve at shallow pressure and depth (~100 MPa  
652 for Cl and ~10 MPa for F; Spilliaert et al., 2006). In addition, the expected minimum  
653 entrapment depth inferred from H<sub>2</sub>O and CO<sub>2</sub> concentrations in magma is mostly greater  
654 than 100 MPa, so that Cl and F could not have exsolved before entrapment. Therefore,  
655 the Cl and F concentrations of the selected melt inclusions (Table 2) are considered as  
656 being those of the primitive melt compositions. The primitive F and Cl concentrations

657 range from 250 to 1160 ppm and 580 to 1270 ppm, respectively. These values yield F/Nd  
658 ratios that are systematically higher by 27 to 45 than a typical MORB mantle value of  
659  $21 \pm 5$  (Workman et al., 2006) or 20 (NMORB Shimizu et al. 2016). Similarly, Cl/K varies  
660 from 0.074 to 0.43, higher than the canonical mantle value of  $0.0075 \pm 0.0025$  (Saal et al.  
661 2002). Such high ratios are consistent with all other arc magmas and attest to these  
662 magmas incorporating a volatile component from the slab (e.g. Van den Bleeken and  
663 Koga, 2015).

664 S concentrations in primitive magmas are assessed from melt inclusion  
665 compositions, taking into account potential modifications by degassing and sulfide  
666 fractionation. S is a volatile element that is incorporated into the gas phase, and is also a  
667 major element constituting the sulfides that can crystallize in magmas. In silicate magmas,  
668 sulfur solubility depends on redox conditions and therefore on the relative proportions of  
669 sulfide and sulfate ions in the melt (e.g. Carrol and Webster, 1994; Jugo et al., 2005).  
670 Observed high dissolved S concentrations are characteristic of oxidized- and hydrous-  
671 basalts found in subduction zones (Roggensack, 2001; Wallace, 2005; Webster et al.,  
672 2010, Narvaez et al., 2018). The melt inclusions from our samples (except Kirishima-  
673 Ohachidake, Fukue and Oninomi) plot above the S concentration at the sulfide saturation  
674 (SCSS) curve (Fig. 7) and thus appear to be supersaturated in sulfide. The SCSS is  
675 calculated based on the saturation of FeS in which  $S^{2-}$  is the dominant S species. However,  
676 no sulfide was found in these samples, suggesting that the majority of S is in the form of  
677  $S^{6+}$ , preventing the formation of FeS. The remaining S is in the form of  $S^{2-}$ , and the  
678 concentration must be lower than the SCSS. This explains why these samples are in fact  
679 undersaturated. There are a few exceptions (Y-I and BC-I of Sumiyoshi-ike) in which  
680 sulfide globules were present, suggesting that the SCSS is reached for some  
681 compositions. Lastly, S concentrations of all our melt inclusions are under-saturated with  
682 respect to anhydrite saturation (SCAS), indicating sulfate fractionation had not occurred  
683 (Li and Ripley, 2009; calculated SCAS between 5270 to 8560 ppm for our samples).  
684 Considering that high S concentrations were found in the inclusions, and that inclusions  
685 were trapped deep in the crust, we conclude that S depletion due to degassing is minimal,  
686 except for Yufu and Sumiyoshi-ike.

687 Fukue and Oninomi are different, and occur at the line of sulfide saturation (Fig.  
688 7). This is consistent with the observations of sulfide globules commonly found in the

689 whole rock samples. For these samples, clearly it is not possible to determine the primitive  
690 S concentrations. The magmas are sulfide ( $S^{2-}$ ) saturated at the surface (Fig. 7) and as  
691 solubility of S decreases with pressure (e.g. Mavrogenes and O'Neill, 1999; Hart and  
692 Gaetani, 2006), then the magmas are also sulfide saturated at the depth of melting,  
693 assuming that the redox state of the sub-arc mantle remained reduced. Alternatively,  
694 because the sub-arc mantle is oxidized (e.g. Parkinson and Arculus, 1999; Kelley and  
695 Cottrell, 2009), then the magma remains undersaturated and the melt inclusions  
696 potentially record the primitive S compositions. In the latter case, the magmas of Fukue  
697 and Oninomi became reduced at a late stage, just prior to eruption, to explain the presence  
698 of sulfides. Kirishima-Ohachidake plots on the SCSS, but no sulfides were observed in  
699 the melt inclusions (Fig. 7). The sample has an elevated proportion of  $S^{6+}$  (0.5-0.9, Table  
700 S2), explaining the absence of sulfide (based on visual inspection of hand samples).  
701 Because the sample has the lowest S of all the Kyushu melt inclusions, and the  
702 concentration of  $H_2O$ ,  $CO_2$ , and S are lower than those of the sample from the near-by  
703 Shinmoedake edifice, the melt inclusions of Kirishima-Ohachidake have potentially  
704 experienced substantial degassing prior to entrapment. We report the sulfur concentration  
705 of the previously selected G-I melt inclusions (800 ppm) as the probable primitive value,  
706 with a footnote (Table 2).

707 Sulfur may leave the magma through partitioning into the fluid during degassing  
708 (i.e. degassing of  $H_2O$  and  $CO_2$ ). This S-loss via degassing results in a positive correlation  
709 of  $H_2O$  with S as reported in natural and experimental samples (e.g. Spilliaert et al. 2006;  
710 Lesne et al. 2011). Such correlations were found among the inclusions from Kirishima  
711 SM, Kaimondake, Kuju, but not for Sumiyoshi-ike, Kirishima-OH, and Yufu, whose  
712 samples showed sulfide saturation (Fig. 6a). The correlation of  $H_2O$  and S could  
713 alternatively derive from mixing, which is confirmed by correlations between the non-  
714 degassing elements F and S (Fig. 6b). It is possible to conclude that S variation is in fact  
715 due to processes other than degassing, and that the S concentration of the mafic  
716 endmember could therefore represent the primitive composition (e.g. Kaimondake). The  
717 compositional variations of Kuju and Kirishima are considered to represent a melting  
718 process. In this case, the highest degree melt represents the least fractionation from the  
719 source, and the lowest trace element concentrations (Table 2). Such primitive magmas  
720 globally coincide with the highest concentrations of S and  $H_2O$  in the melt inclusions

721 from a sample.

722 For Yufu, the S concentrations are paradoxical. The Yufu samples plot above the  
723 SCSS line (Fig. 7; for example, up to 3793), suggesting a high proportion of S<sup>6+</sup> in the  
724 magmas. However, the measured S speciation gives negligible S<sup>6+</sup> abundances in the melt  
725 inclusions, implying reduced  $fO_2$  conditions (Table S3). Furthermore, if the melt  
726 inclusion is really reduced, sulfide should precipitate and the S concentration should be  
727 at the SCSS, but no sulfides were found in Yufu melt inclusions. We argue that this  
728 paradox is a consequence of the heating procedure specific to the sample. As Yufu melt  
729 inclusions were partially crystallized, they were re-homogenized with the heating stage  
730 (e.g. Le Voyer et al., 2010). During this process, a purified He flux was flushed in and  
731 the oxygen fugacity was kept between  $10^{-10}$  and  $10^{-9}$  atm, to avoid oxidation of the host  
732 mineral and to ensure an efficient quench. These extremely reduced conditions affected  
733 the S speciation of the melt inclusion, even if the sample were to have originally been  
734 oxidized. In fact, independent analyses from amphibole compositions of basaltic mafic  
735 enclaves found in Yufu indicate oxidized conditions ( $\Delta NNO+1 \pm 0.4$ ; Ohta et al., 1990;  
736 Ridolfi et al., 2010). Thus, we conclude that the Yufu magma was originally oxidized and  
737 the high-S concentrations of its melt inclusions reflect the value at the time of entrapment.

738 Mineral phases such as carbonates, sulfides, sulfates and native sulfur were reported  
739 inside the bubbles upon cooling (Kamenetsky et al., 2002, Moore et al., 2015; Robidoux  
740 et al., 2018). Here we did not study the inside of the inclusion bubbles. If such S-bearing  
741 phases were present on the wall of the vapor bubble, primary S concentrations would be  
742 significantly underestimated. Although it is impossible to examine the exposed vapor  
743 bubble *a posteriori* after polishing, the bubbles in the melt inclusions of this study did not  
744 contain visible solid precipitates (inspection under an optical microscope, Supplementary  
745 material document S2). Without this quantification, we assumed our oxidized melt  
746 inclusions to be undersaturated, and to not crystallize either anhydrites or sulfides.

747

#### 748 ***S isotope composition of the primitive magma***

749 Identifying the source of magmatic volatiles through S-isotope characterization  
750 requires careful evaluation of all potential modifications to the initial isotopic signature.  
751 In general, modification and variation of sulfur isotope compositions is caused by two  
752 processes: mixing of sulfur derived from isotopically distinct sources, and isotopic

753 fractionation due to changes in sulfur speciation. For the latter case, a significant  
754 equilibrium sulfur isotope fractionation factor ( $\alpha_{\text{gas-melt}}$ ) was calculated for the degassing  
755 of a magma ( $\alpha_{\text{gas-melt}} = 0.996$  to  $0.998$  at the degassing conditions of the Mazama magma,  
756 e.g. Mandeville et al., 2009), leading to  $\delta^{34}\text{S}_{\text{melt}}$  being heavier than equilibrium  $\delta^{34}\text{S}_{\text{gas}}$  at  
757 oxidized conditions (typically higher than at  $\sim\Delta\text{FMQ}+1$ ; Marini, 2011; Fiege et al., 2015).  
758 The implication was that open-system degassing potentially reaches an even higher  
759  $\delta^{34}\text{S}_{\text{melt}}$  (Marini et al., 1994; example of Krakatau 1883 eruption, Mandeville et al., 1998).  
760 For the other case, where modification of S-isotope composition is due to mixing,  
761 previous studies have interpreted variations in S isotope composition of arc lavas as  
762 resulting from the addition of a slab-derived seawater sulfur component to the sub-arc  
763 mantle (Sasaki and Ishihara, 1979; Alt et al., 1993, 2012; Evans et al., 2014). However,  
764 these studies pointed out that the fractionation of S isotopes during the transformation  
765 mechanisms of seawater  $\text{SO}_4^{2-}$  to dissolved S in primitive arc magma is poorly  
766 constrained. The variation observed in the present sulfur isotope data (Fig. 8) is caused  
767 by mixing and is unlikely to have been influenced by degassing, because the melt  
768 inclusions of this study trapped magmas before degassing, as inferred from the high  $\text{CO}_2$   
769 contents (note, the bubble corrected values), with the exception of the Kirishima-OH  
770 sample.

771 The sulfur fractionation model showed that the variation in  $\delta^{34}\text{S}$  is maximal during  
772 open-system degassing (Marini et al., 2011; Supplementary document S1). In detail, the  
773 model predicts that even if there was extensive degassing and all the melt inclusions were  
774 affected by open-system degassing of up to 50% of the initial S, the  $\delta^{34}\text{S}$  of the melt  
775 inclusions would be changed by a maximum of 2 ‰, which is the  $2\sigma$  error of our  $\delta^{34}\text{S}$   
776 measurements. The  $\delta^{34}\text{S}$  measurement is insensitive to less than 50% degassing due to its  
777 uncertainty in this study. In the case of extremely degassed whole rocks, a study has  
778 reported  $\delta^{34}\text{S}$  of +18.0 ‰ for S contents down to 9 ppm (Aso Nakadake; Ueda and Saki,  
779 1984). Therefore, systematic degassing of 50% is unlikely among the inclusion data here,  
780 since S concentrations of Kyushu melt inclusions are in the range of those reported for  
781 other primitive arc basalts in the world (e.g., as high as  $\sim 3000$  ppm in Cascades, Wallace,  
782 2005). Furthermore, there is no along-arc correlation between  $\delta^{34}\text{S}$  and Cu (sulfide  
783 fractionation indicator, Beaudry et al. 2018), while local correlations within 3 ‰ are  
784 present, we conclude that the observed isotopic variation is not controlled by sulfide

785 fractionation. The  $\delta^{34}\text{S}$  values of our primitive melt inclusions therefore reflect the values  
786 of the metasomatized mantle below the volcanoes (Table 2).

787 A correlation between  $\delta^{34}\text{S}$  and S concentrations would be expected if magma  
788 degassing were to be significant. There is no such correlation among melt inclusions from  
789 each individual Kyushu volcano (Fig. 8a). The inclusions Yufu are the only samples  
790 showing a weak negative correlation (Fig. 8a) and the values of other volcanoes are  
791 clustered around a single value. There is an overall weak negative correlation for samples  
792 from several of the volcanic edifices. However, this correlation is less likely to be due to  
793 degassing as it would require a common S concentration in the parental magmas and  
794 exactly the same fractionation mechanism during degassing of individual edifices spread  
795 across the island. Sulfur depletion through degassing can be illustrated by normalizing  
796 the concentration with a non-degassing, incompatible element such as  $\text{K}_2\text{O}$ , (Fig. 8b).  
797 The negative correlations with  $\delta^{34}\text{S}$  are slightly more apparent for Yufu, and Sumiyoshi-  
798 ike. Given that trace element concentrations of these samples were controlled by mixing  
799 rather than degassing (Supplementary material document S1), we would argue that the  
800 negative trend is also due to mixing. Therefore, the  $\delta^{34}\text{S}$  values of the mixing endmembers  
801 potentially represent the magma source values, considering their mafic character and high  
802 S concentration. Both endmembers are reported as the potential primitive magmas of  
803 Yufu and Sumiyoshi-ike (Table 2). For Oninomi and Fukue,  $\delta^{34}\text{S}$  values are  
804 systematically lower than those of other oxidized magmas. In fact, the S concentration of  
805 these samples was controlled by sulfide saturation, so the low  $\delta^{34}\text{S}$  values are potentially  
806 explained by fractionation during sulfide precipitation, which leaves the remaining melt  
807 slightly lighter than the oxidized magmas (e.g. Marini et al., 2011).

808

### 809 *The three magma groups of Kyushu*

810 Magma sources are identified using trace element ratios of highly incompatible  
811 elements. Firstly, an inspection of the trace element spidergrams shows that Fukue and  
812 Oninomi magmas are different to the others due to their absence of Nb depletion,  
813 otherwise common in arc magmas (Fig. 3). While the differences between Northern and  
814 Southern Kyushu may not be obvious from the spidergram, they can be distinguished on  
815 ratio-ratio plots. The plot of Ba/Zr against Nb/Zr illustrates the contrast between the arc

816 magmas and the mantle array derived from MORB samples (Fig. 9a, Ba/Nb ~ 7, note also  
817 model mantle compositions are indicated). Because the contribution of slab-derived  
818 material to the magma source increases the Ba/Zr ratio (Fig. 9a; e.g. Leeman et al. 2005),  
819 arc magmas show elevated Ba compared to Nb. Fukue and Oninomi magmas plot close,  
820 and almost parallel, to the MORB mantle array, while other Kyushu magmas plot above  
821 it and reflect the addition of slab-derived material, rich in Ba. Furthermore, Northern  
822 Kyushu magmas (open symbols in Fig. 9a) show, in general, higher Ba/Zr (and Ba/Nb),  
823 than Southern Kyushu, with the exception of one endmember of Yufu.

824 The distinct groups are also clearly visible in Fig. 9b (Sr/Y against Th/Nb). The  
825 MORB melt inclusion compositions (small light blue triangles) cluster near the values of  
826 the model depleted MORB mantle, with Sr/Y = 2.3 and Th/Nb = 0.053 (Workman and  
827 Hart, 2005). For peridotite partial melting, Sr/Y in MORB melt inclusions show limited  
828 variation, whereas the Kyushu melt inclusions show a variation of more than a factor of  
829 5, with those of the North (open symbols) plotting at higher Sr/Y values (>30) than those  
830 of the South (<30) and MORB. There is also a variation in Th/Nb, a ratio of highly  
831 incompatible elements (Fig. 9b). This variation is caused by the input of a mobile agent  
832 from the slab to the arc magma source (i.e. metasomatism), and the resulting variation is  
833 characteristic of arc magmatism (see light pink solid circle representing arc olivine-hosted  
834 melt inclusions). The Th/Nb of the slab agent is potentially characterized both by its  
835 physical nature (aqueous flux, silicate melt, or supercritical fluid) and by the source  
836 composition (sediment, altered oceanic crust). For example, Nb is strongly partitioned  
837 into rutile,  $D_{\text{Nb}}^{\text{rutile/melt}}$  ranging 14 to 550 (Foley et al. 2000; Klemme et al., 2005), and  
838 the Th abundance can be controlled by allanite and apatite (Hermann, 2002). As these  
839 minerals are commonly found in the sedimentary and magmatic part of the slab, the slab-  
840 issued flux can be characterized by these phases (Klimm et al, 2008). Furthermore, the  
841 value of Th/Nb in the altered oceanic crust is expected to be similar to that of MORB,  
842 and variations in mantle fertility only causes a range in Th/Nb from 0.05 to 0.14 (Fig.  
843 9b). On the contrary, subducting sediments can vary significantly, from 0.10 to 1.5, with  
844 an average of 0.77 (Plank and Langmuir, 1998). As HFSE elements are poorly soluble in

845 aqueous fluid, it is generally inferred that only silicate melt, especially that deriving from  
846 subducting sediment, is the most effective slab agent that can change Th/Nb ratio.

847 The high Sr/Y values are often interpreted as being adakite signatures, as they are  
848 the result of partial melting of a slab containing residual garnet (e.g. Defant and  
849 Drummond, 1990; Martin et al. 2005). In this scenario, melting, instead of dehydration,  
850 is the process required to fractionate Sr and Y. High Sr/Y values in Northern Kyushu are  
851 comparable to those from other arc systems, for example the Southern Cascades (1 to  
852 ~10% slab melt addition into source mantle, Walowski et al., 2016). Alternatively, high  
853 Sr/Y ratios are potentially derived from magma interacting with garnet and/or amphibole  
854 in the lower arc crust during magma ascent (e.g. Davidson et al., 2007). Because the melt  
855 inclusions of this study have experienced little crystal fractionation and the primitive  
856 chemical components were carefully selected (Supplementary material document S1), the  
857 Sr/Y ratios of our inclusions are unlikely to reflect the extensive lower crust interaction  
858 processes required to change the Sr/Y ratios.

859 Putting together all of the characteristics of Fig. 9, Northern Kyushu magmas are  
860 higher in Sr/Y and Ba/Zr than Southern Kyushu ones, and the volcanic front magmas  
861 (Northern and Southern Kyushu) are higher in Th/Nb and lower in Nb/Zr than MORB.  
862 There are therefore three sources of the Holocene mafic magmas. Among those, Oninomi  
863 and Fukue are the closest to the mantle array (Fig. 9a), with the lowest Ba/Zr enrichments  
864 of about 1.5 and 1.8, respectively. This very modest slab contribution, particularly for  
865 Fukue, is explained by its back arc location, where the distance to the slab surface is over  
866 400 km (Fig. 1). A low Ba/Th enrichment is also seen in Akashima volcano from the  
867 same Fukue volcanic group (Kuritani et al., 2017). While Ba/Th and Ba/Nb are close to  
868 the mantle values, Sr/Y is significantly different from the depleted mantle value, both for  
869 Fukue and Oninomi. Because the DMM can yield a maximum Sr/Y of ~10 (determined  
870 using  $[\text{Sr}]_{\text{DMM}} / [\text{Y}]_{\text{DMM}} * D_{\text{Y}}^{\text{mantle/melt}} / D_{\text{Sr}}^{\text{mantle/melt}}$ ) during mantle partial melting  
871 (Workman and Hart, 2005), and the Sr/Y values of Fukue and Oninomi are >20, they  
872 must reflect the source composition.

873 The melt inclusions from the other edifices plot above the mantle array (Fig. 9a).

874 For example, fluid derived from the slab increased the Ba/Zr of melt inclusions (Fig. 9a)  
875 by a factor of 2.8 (Kaimon) to 9 (Aso). Th/Nb varies by up to a factor of 10 (1.0 for  
876 Kirishima Shinmoedake) compared to that of the MORB-cluster ( $\sim 0.1$ ). The slab  
877 contribution is the most significant for the two Kirishima samples, Aso and Yufu (Fig.  
878 9a), and is interpreted as being mainly due to silicate melt addition causing high Th/Nb  
879 and Sr/Y (Fig. 9ab). On the contrary, for Sumiyoshi-ike and Kaimondake the contribution  
880 is weaker and likely due to aqueous fluids.

### 881 *Geodynamic significance of the source regions*

882 The presence of these three magma groups indicates that there are at least three  
883 sources responsible for magma genesis below Kyushu Island. Compositional variation of  
884 arc primitive magmas is derived from variations in the degree of melting and the source  
885 composition of the metasomatized mantle, whose composition is controlled by the  
886 quantity and composition of flux added from the slab (e.g. Stolper and Newman, 1994;  
887 Tatsumi and Eggins, 1995). Such variation is observed on a scale of a whole arc (1000s  
888 of km) to within a single volcano (e.g. Le Voyer et al., 2010). The studied volcanic area  
889 covers a length of 250 km, and the subducting plate underneath has a discontinuity in the  
890 plate, separating a young northern plate from an older southern one (Shikoku Basin and  
891 West Philippine Basin, respectively, Fig. 1).

892 First, we examine the potential heterogeneity of the mantle below Kyushu  
893 volcanoes prior to metasomatism from the slab. This is particularly important for the  
894 Northern Kyushu volcanoes, for which basalts were previously described as “OIB-like”  
895 for their enriched trace element characteristics (e.g., Nakada and Kamata, 1991). In fact,  
896 Fukue and Oninomi magmas are alkali basalts, as commonly found in oceanic islands,  
897 with slightly elevated Th/Nb values compared to MORB-DM (Fig. 9b), and the Th/Nb  
898 ratio is closer to the value of the bulk silicate earth, which is the enriched mantle  
899 (McDonough and Sun, 1995; Palme and O’Neill, 2003; Lyubetskaya and Korenaga,  
900 2007). Similar systematics can be seen on a Ba/Zr - Nb/Zr plot (Fig. 9a). These diagrams  
901 also show that arc magmas in general have a significant addition of Ba and Th compared  
902 to Nb. The addition of a component rich in Ba and Th to a typical upper mantle (i.e.

903 depleted mantle) potentially creates similar characteristics to an “enriched” mantle and  
904 detecting the difference between the two is not easy from these diagrams. In detail, given  
905 the ranges of Th/Nb and Ba/Nb of enriched and depleted mantle are much smaller than  
906 the variation of these ratios observed in arcs, we conclude that the initial mantle  
907 heterogeneity does not significantly impact the trace element characteristics of arc  
908 magmas. Therefore, it does not matter if we consider the mantle as being initially enriched  
909 or depleted before the slab agent addition. For the following discussion, we consider  
910 depleted mantle (Workman and Hart, 2005; Salters and Stracke, 2004) as the reference  
911 mantle composition below the arc volcanoes prior to metasomatism.

912         The dichotomy of Sr/Y values separating North and South Kyushu is thus explained  
913 by slab contribution (Fig. 9b and Fig. 10). Fractionation of Sr/Y is due to the presence of  
914 garnet as a residual phase, and the residual garnet is potentially present in the mantle,  
915 metasomatized mantle, and slab (i.e. sediment and AOC). Following the consensus about  
916 the thermal structure of the arc mantle (e.g., Syracuse et al. 2010), the hottest part of the  
917 mantle below a volcano is located about midway between the bottom of the Moho and  
918 the top of the slab, which is approximately 65-75 km (Fig. 1). This is not a depth at which  
919 melt would form in garnet peridotite (e.g. Walter, 1998). Considering that there is no  
920 strong upward movement of the solid mantle below a volcano, the maximum melting is  
921 expected to be attained at the level of this hottest area of the mantle, and this is likely the  
922 condition of equilibration between mantle and melt. The garnet signature cannot be  
923 produced in the mantle beneath the volcanic front. The residual garnet signature must  
924 therefore come from the slab either from subducted sediments or AOC (Defant and  
925 Drummond, 1990; Martin et al. 2005), or the metasomatized mantle near the slab-mantle  
926 interface (e.g. Lara and Dasgupta, 2020). Furthermore, Sr/Y is more strongly fractionated  
927 by the garnet-melt pair, than by garnet-aqueous fluid (Martin et al., 2005). On this basis,  
928 the high Sr/Y magmas in Northern Kyushu are more consistent with the presence of a  
929 silicate melt near the slab - mantle interface with the presence of residual garnet in  
930 sediment and/or AOC.

931         There remains the question, does the difference between the north and south simply

932 represent the presence/absence of silicate melt or does it represent variable contributions  
933 from different lithologies of the slab (i.e. AOC and sediments)? The answer to this  
934 question is non-unique, as the exact compositions of the subducting material are  
935 undeterminable, as are the exact pressure-temperature conditions at which the  
936 dehydration/melting reactions take place. Nevertheless, there exists a wealth of databases  
937 which allow an educated guess to be made about the composition of the slab material  
938 (e.g. Plank and Langmuir, 1998; Plank, 2014), and geodynamic models of pressure-  
939 temperature paths along the subduction zones (Syracuse et al., 2010). Here, we used the  
940 model package, Arc Basalt Simulator 5 (ABS 5, Kimura 2017), to test the role of the  
941 fluxes from sediments and AOC, as well as the presence/absence of silicate melt  
942 (Supplementary document S1 for detail). ABS 5 conveniently includes sediment and  
943 AOC compositions from around Japan. Furthermore, it includes the models of pressure -  
944 temperature trajectory, pseudosection diagrams for related lithologies, a composite slab  
945 with six lithologies, and chromatographic fluid/melt modification adjustments. In Fig.  
946 11a,b, the endmember fluids/melts derived from a sediment (Nankai sediment, ABS 5)  
947 and AOC (Shikoku basin, ABS 5) are plotted, together with depleted mantle. As expected,  
948 the Th/Nb ratio of the fluxes derived from AOC is close to the value of the depleted  
949 mantle, and those derived from the sediments have significantly higher Th/Nb. Because  
950 the composition of the slab derived agent controls the ratios of highly incompatible  
951 elements, both fluid/melt phases from sediment and AOC must modify the source mantle  
952 to cover the range of variation observed (see mixing curve in Fig. 11b). However, because  
953 of the contrast in the proportion of water in the fluid (~90%) and melt (~10%), it requires  
954 a lower aqueous fluid flux to attain the equivalent extent of wet mantle melting. In other  
955 words, to reach the same amount of melted mantle, the proportion of slab melt needed  
956 will be greater than that of fluid. On Fig. 11, this tendency is displayed by the position of  
957 thick dashed lines, which are the result of the flux melting model with degrees of melting  
958 ranging from 5- 20%. For example, the arc magmas derived from slab melt (blue and  
959 orange lines) systematically plot at higher Sr/Y than those derived from fluid (Fig. 11b).  
960 This explains why slab derived melt modifies the mantle more than fluid at a given

961 melting percentage.

962 The compositions of the mantle fluxing agent illustrate the first-order influence on  
963 the trace element characteristics of arc magma, as has been known and discussed for many  
964 years (e.g., Tatsumi, 1989; Kessel et al., 2005). As shown in Fig. 11, one approach is to  
965 use a detailed forward model to predict the flux from the slab. Alternatively, ABS 5 also  
966 includes a function of the inverse fit, by iteratively searching for the best fit solution by  
967 changing conditions of fluid release along the slab. Given the multiple parameters, it was  
968 fairly easy to create an inverse model that yielded results showing a contrast of slab  
969 surface temperatures between North and South Kyushu. However, we concluded that the  
970 fit-results of the inverse calculation of ABS 5 do not strengthen the conclusion already  
971 shown with Fig. 11, so we did not discuss this further. Instead, we tested a mixing model  
972 using three components: the depleted mantle, a sediment flux, and an AOC flux (Table  
973 S8), and calculated the proportion of a mixture satisfying the observed incompatible ratios  
974 (Th/Nb, Rb/Ba, Ba/Nb, Sr/Y, Sr/Nd, Nb/Zr). The results of the calculation also confirm  
975 the conclusion drawn from Figs. 9, 10, and 11. Based on the proportion of the mixing  
976 model (Fig. 12), Northern Kyushu volcanoes show stronger slab input than the Southern  
977 volcanoes, suggesting a higher amount of flux due to higher temperatures. Sediment -  
978 AOC contributions are also variable but do not correlate with the geographical  
979 distribution of volcanoes. Lastly, we caution that this trend is just based on the Holocene  
980 mafic melt inclusions that we could find, and the existence of the north-south dichotomy  
981 needs to be tested by extending the sampling to other mafic eruptions of different ages.

982 The Northern Kyushu group, Yufu, Kuju, and Aso, are associated with silicate melt  
983 input from the slab, and the southern Kyushu group, Kirishima, Sumiyoshi-ike,  
984 Kaimondake, are associated with aqueous fluids (Fig. 11b). However, the amount of flux  
985 from sediment compared to the amount of flux from AOC creates different sub-groups.  
986 Aso, Kirishima and one endmember of Yufu (BQ-III) have higher Th/Nb values than  
987 Kuju, Kaimondake, Sumiyoshi-ike, and the other endmember of Yufu (BD-I). This  
988 difference is explained by a higher proportion of sediment flux than that of AOC (Fig.  
989 11ab). Aso, Kirishima, and Yufu (BQ-III) plot closer to the mixing curves of sediment-

990 depleted mantle (Fig. 11ab). Curiously, this variation of sediment-AOC contributions  
991 does not correlate with the geographical north-south separation, or with the depth to the  
992 slab surface, or the dip of subduction. It should be noted that Yufu volcano melt inclusion  
993 magmas are a mixture of both melts, that with more sediment-derived flux and that with  
994 less. This variability within a single volcano indicates that primitive magmas arriving at  
995 the base of the crust can be heterogeneous for different reasons. We also note that the  
996 degrees of dehydration of the slab in the forearc region along Kyushu are variable, with  
997 patchy regions of high and low water contents in the forearc mantle wedge, as identified  
998 by a seismic tomography study (Abe et al. 2013). This indicates that availability of  
999 “water” in the slab below the arc volcanic front may vary with respect to spatial and  
1000 perhaps temporal distributions, causing non-systematic contributions of sediment and  
1001 AOC fluxes.

1002 Magma from Fukue and Oninomi are significantly different from those of the main  
1003 volcanic front. They appear to record significantly different mantle processes. Fukue  
1004 magma is slightly higher in Ba/Th and Th/Nb than the depleted mantle alignment (Fig.  
1005 9a). Fukue volcano is located in the back-arc region, where slab surface depth is  
1006 approximately 400 km. At such a depth, the slab is mostly dehydrated and unlikely to  
1007 contribute significant aqueous flux that could lead to melting. Instead, magma genesis at  
1008 Fukue is caused by mantle upwelling, potentially responding to the extension of the back-  
1009 arc region, because Fukue is located broadly at the northern extension of Okinawa trough.  
1010 If the degree of partial melting is low, it explains the LREE enriched pattern of the  
1011 spidergram (Fig. 3). Alternatively, the magma genesis could be related to a deep fluid  
1012 released from the older Pacific plate stagnating at the mantle transition zone, extending  
1013 over a much broader area than the young Philippine Sea plate (Kuritani et al., 2017). This  
1014 model is constrained by isotopic composition of Fukue magmas, which are similar to  
1015 deeply recycled sediment (EM2), and provides the mechanism for broader magmatic  
1016 activities extending to China. A recent seismic tomography study appears to indicate the  
1017 that Philippine Sea plate is present beneath Fukue (Zhao et al., 2012; Liu and Zhao, 2016),  
1018 and if the contribution of the Pacific plate is critical, such mantle upwellings must flow

1019 around the upper Philippine sea plate.

1020       Lastly, Oninomi volcano is unique in character, quite different from the near-by  
1021 volcanoes (e.g. Yufu, Kuju, and Tsurumi-dake, not studied). The spidergram of Oninomi  
1022 magma shows no arc-magma-like character (Fig. 3), and it is somewhat similar to that of  
1023 Fukue. The Ba/Nb and Th/Nb values are close to those of depleted mantle, suggesting it  
1024 records little input from the slab. It seems that a small degree of partial melting of a  
1025 slightly metasomatized mantle would be sufficient to explain the magma composition. As  
1026 the volcano is located in the middle of a 40 km wide graben, active for the last 6 Ma  
1027 (Kamata and Kodama, 1994), then if the extension is accompanied by lithospheric  
1028 thinning, corresponding upward decompressing in the mantle below could cause adiabatic  
1029 melting (McKenzie and Bickle, 1988). It should be noted that mantle melting in response  
1030 to a small change in crustal load has been observed and demonstrated in Iceland, where  
1031 deglaciation has caused magmatism (Slater et al., 1998). We consider that this upward  
1032 decompression melting in response to the extension is the most plausible mechanism for  
1033 magma genesis at Oninomi.

1034

### 1035 ***Sources of sulfur in the subduction zone***

1036       The sulfur isotope compositions of melt inclusions from the Kyushu volcanoes in  
1037 this study reflect the compositional variation of magma sources rather than degassing  
1038 process. Now we need to look at what could be the main contribution to sulfur isotope  
1039 variation in the subduction zone magmas? In our data set,  $\delta^{34}\text{S}$  weakly correlates with  
1040 Th/Nb, which is sensitive to slab input (Fig. 13a). As Th/Nb indicates the composition of  
1041 the metasomatized magma source, and the depleted mantle is at -0.91 ‰ for  $\delta^{34}\text{S}$ , and  
1042 0.06 for Th/Nb, the variation is reflecting the slab input. Comparing with other melt  
1043 inclusion measurements from Antilles and Kamchatka (Bouvier et al., 2008; Gurenko et  
1044 al., 2018; 2021), our data plot in a similar range, showing global systematics. Fig. 13b  
1045 illustrates a lack of systematics of  $\delta^{34}\text{S}$  and Cl/F, which is strongly influenced by the  
1046 presence of fluid and hydroxyl minerals (amphibole and mica). It is inferred that an  
1047 aqueous fluid flux from lithologies with residual hydroxyl minerals yields higher Cl/F  
1048 than a slab melt flux (e.g. Van den Bleeken and Koga, 2015). For example, Cl/F of a cold

1049 subduction volcano is  $3.0\pm 2.0$  (e.g. melt inclusions from Iwate volcano; Rose-Koga et  
1050 al., 2014), and melt inclusions from typical fresh MORB are expected to have values  
1051 around  $0.4\pm 0.4$  (Le Voyer et al., 2015, 2019; Wanless et al., 2014). Kyushu magmas  
1052 cluster around  $Cl/F = 2.1\pm 0.8$ . We conclude from this systematics that the nature of the  
1053 fluid (aqueous or melt) is not the main factor controlling the variation in sulfur isotopes.  
1054 We have also examined other trace element indicators that are considered sensitive to  
1055 fractionation processes rather than source composition. For example, V (either  
1056 concentration or ratios normalized to other trace elements) does not correlate with  $\delta^{34}S$ .  
1057 This suggests that variation in sulfur isotopes may not be due to changes in oxidation state  
1058 of the fluid carrying the sulfur. Similarly,  $\delta^{34}S$  does not correlate with Sr/Y (residual  
1059 garnet), F/Nd (residual hydroxyl minerals or breakdown), or La/Sm (degree of melting).  
1060 The variation in  $\delta^{34}S$  in Kyushu magma must be related to the compositional variation of  
1061 the slab component, which is transported into the mantle wedge by a flux of mobile  
1062 agents, likely a solute rich melt.

1063 Positive correlations between  $\delta^{34}S$  and trace element ratios are also found, for  
1064 example with Rb/Ba, and Pb/Ce, in addition to Th/Nb (Fig. 14). Such recurring  
1065 correlations underlines that the variation in  $\delta^{34}S$  of these Kyushu magmas is again related  
1066 to the slab flux. Careful inspection of these correlations identifies two distinct  
1067 contributions in addition to the depleted mantle. In Fig. 14a, the positive trend for the  
1068 samples from the main volcanic front (excluding Fukue and Oninomi) can be explained  
1069 by mixing between a first endmember with a low  $\delta^{34}S$  and a Rb/Ba lower than that of  
1070 depleted mantle (approximately  $\delta^{34}S < +3$  ‰, Rb/Ba < 0.03), and a second endmember  
1071 with a high  $\delta^{34}S$  and high Rb/Ba. Along this positive trend, the Rb/Ba variations of the  
1072 slab flux come from the different source composition, combined with Rb/Ba  
1073 fractionations due to the presence of specific minerals, for example (i) the presence of  
1074 residual phyllosilicates, which have an affinity for Rb and Ba, and can fractionate them  
1075 from the slab flux, (ii) Ba is preferentially incorporated into feldspar relative to a silica  
1076 rich melt. As for the sources from the slab, first, the AOC is expected to inherit the Rb/Ba  
1077 ratio of MORB, because only slight addition of Rb is expected during MORB alteration  
1078 (0.09; Staudigel, 2003), and this value is similar to that of the depleted mantle (0.089;  
1079 Workman and Hart, 2005 and 0.073; Salters and Stracke, 2004). During subduction of  
1080 this AOC there is no significant amount of phyllosilicates in the metamorphosed basalt

1081 and gabbro, therefore neither endmember can be derived from the AOC. We note that  
1082 altered basalts sampled at the ocean floor often have high Rb/Ba (e.g. Shu et al., 2017;  
1083 Hickey-Vargas et al., 2018) but such basalts are volumetrically insignificant compared to  
1084 the total mafic material subducted. Second, the sediment flux component used in Fig. 11  
1085 gives Rb/Ba as 0.007 and 0.009 for fluid and melt respectively. Even considering oceanic  
1086 manganese nodules and carbonates, they all have Rb/Ba <0.01 (e.g. Li and Schoomaker,  
1087 2005). So, based on the Rb/Ba ratio, the sediments would be the best candidate for the  
1088 endmember with low Rb/Ba and low  $\delta^{34}\text{S}$ . Subduction zone sediments are reported to  
1089 have a range of negative  $\delta^{34}\text{S}$  values (-40 to +10 ‰; e.g. Sasaki and Ishihara, 1979; Lew,  
1090 1981; Alt and Burdett, 1992; Canfield and Farquhar, 2009; Alt and Shanks, 2011; Peters  
1091 et al., 2010).

1092 For the high Rb/Ba endmember, we considered other specific slab components. For  
1093 example, the Rb/Ba ratio of shale is high (0.24) and seawater has a Rb/Ba of 8 (Li and  
1094 Schoomaker, 2005). The corresponding  $\delta^{34}\text{S}$  values range from -16 to -38 ‰ (Sasaki and  
1095 Ishihara, 1979; Cruse and Lyons, 2004) for shale, and +21 ‰ for seawater (Rees et al.,  
1096 1978). Seawater sulfur is sulfate, and thus seawater derived material rich in sulfate tends  
1097 to give strong positive sulfur isotope values. The systematics of  $\delta^{34}\text{S}$  and Rb/Ba suggest  
1098 a slab component of seawater itself (or rocks impregnated by seawater sulfate). Forearc  
1099 serpentinites also show variable but high Rb/Ba, because of varying interactions with  
1100 seawater (Savov et al. 2005; Deschamps et al., 2010; Albers et al. 2020). Similarly,  
1101 feldspar-bearing sandstone, as well as granite from south-west Japan, is also reported to  
1102 have high Rb/Ba (Shinjoe 1997; Kiminami et al. 2009). These rocks are magnetite-  
1103 bearing (and often sulfide-bearing) and sulfate is not expected to be stable. Only brown  
1104 clasts of serpentinites are reported to have an oxidized state due to extensive interaction  
1105 with seawater expected to have occurred near the seafloor (Albers et al. 2020). Because  
1106 of this, it is not clear if serpentinite is an adequate sink for high  $\delta^{34}\text{S}$ , commonly related  
1107 to sulfate. Alternatively, serpentinites in the subducting slab can potentially carry sulfides  
1108 with positive  $\delta^{34}\text{S}$ , due to extensive seawater interaction. Such serpentinites are expected  
1109 to release sulfate-bearing fluids with  $\delta^{34}\text{S} = 14$  ‰, at the time of serpentinite dehydration  
1110 to chlorite harzburgite (Alt et al. 2013). It is not clear if these ultramafic rocks are the  
1111 definitive carrier satisfying both the high  $\delta^{34}\text{S}$  and high Rb/Ba, but it appears that they are  
1112 certainly the potential carriers of a seawater sulfur isotope signature.

1113           Considering the discussion above and our current knowledge, we conclude that  
1114 seawater is a plausible vector responsible for causing the high  $\delta^{34}\text{S}$  and high Rb/Ba  
1115 geochemical signature in Kyushu arc magmas. Direct injection of seawater into the source  
1116 region of arc magmas is a possible scenario as there are reports indicating the presence  
1117 of seawater in the subduction zone eclogite (seawater fluid inclusions; e.g. Sumino et al.,  
1118 2010; Kawamoto et al., 2013). The model of the deep injection of seawater fluids  
1119 contradicts with observations of mantle-like sulfur isotopes in the most primitive deep  
1120 arc cumulates (Lee et al., 2018), where the  $\delta^{34}\text{S}$  in a suite from Sierra Nevada cumulates  
1121 correlates with an index of magma evolution indicating that high  $\delta^{34}\text{S}$  is a result of crustal  
1122 assimilation and crystal fractionation process (Lee et al., 2018). Consequently, this  
1123 suggests an efficient release of sulfate from the slab before the arc magmatic front,  
1124 leaving the reduced sulfide with lighter  $\delta^{34}\text{S}$  as the dominant sulfur species transported to  
1125 the deep mantle (Lee et al., 2018). Similar conclusion about the negative  $\delta^{34}\text{S}$  of the slab  
1126 component was presented by Li et al. 2020 based on the measurements of sulfides in  
1127 metamorphic veins. On the contrary, Walters et al. 2019 concluded that the slab  
1128 component had a positive  $\delta^{34}\text{S}$  based on their analysis of sulfides in subducted ophiolites.  
1129 Lastly, the analysis of subarc xenoliths indicates the presence of positive  $\delta^{34}\text{S}$  primitive  
1130 magma (Bénard et al. 2018). We think this disparity illustrates the complexity of sulfur  
1131 behavior in magma. It should be noted that sulfur in the Kyushu inclusions are dominantly  
1132 sulfate, while the cumulates in Lee et al.'s study (2018) and the slab component of Li et  
1133 al. (2020) suggest the presence of reduced magma and crystallization at reduced condition  
1134 dominated by sulfide. Such a contrast may mean that there is a range of redox states of  
1135 arc magmas controlling the sulfur isotopic signature.

1136           The ratio of Pb/Ce correlates with  $\delta^{34}\text{S}$ . This correlation is consistent with the  
1137 interpretation based on the systematics of Fig. 14a ( $\delta^{34}\text{S}$  vs. Rb/Ba). The flux from the  
1138 slab gives a Pb/Ce value ranging from 0.13 ~ 0.45 for an AOC-derived flux (ABS 5 model  
1139 in our study), of 1.1 to 4.0 for a sediment-derived flux, and 1.7 for seawater (Li and  
1140 Schoemaker 2005). Furthermore, no negative  $\delta^{34}\text{S}$  component is needed to describe the  
1141 systematics of the Kyushu magmas, so long as a strongly negative sulfur isotope  
1142 component is subducted. For example, sulfide in sediments and oceanic crust is as low as  
1143 - 40 ‰ (Peters et al., 2010; Alt and Shanks, 2011). The lack of a negative component  
1144 suggests that the dominant sulfur mobility from the slab is carried out by sulfate rather

1145 than sulfide, and the negative  $\delta^{34}\text{S}$  of sulfide is perhaps not incorporated into the slab flux.  
1146 While our findings may apply only to the local sulfur cycle in Kyushu volcanoes, it shows  
1147 potential isotopic filters operating at the subduction zone that are primarily controlled by  
1148 initial sulfur speciation. Lastly, some negative  $\delta^{34}\text{S}$  values are reported in melt inclusions  
1149 and embayments from the Lesser Antilles, where there are known to be large sediment  
1150 inputs into the mantle wedge (Bouvier et al., 2008), and a decreasing  $\delta^{34}\text{S}$  with decreasing  
1151 S concentration for melt inclusions from Kamchatka (Gurenko, 2021); they are  
1152 interpreted as the result of sulfur fractionation in the crust (i.e. sulfide precipitation and/or  
1153 degassing). The presence or absence of negative  $\delta^{34}\text{S}$  slab flux is thus critical in  
1154 understanding S cycle systematics in the subduction zone system.

1155

## 1156 **Conclusions**

- 1157 - Olivine-hosted magmatic melt inclusions along the Kyushu arc in this study are  
1158 from mafic tephra deposits. We identified the melt inclusion with the most  
1159 primitive magma composition for each volcano.
- 1160 - The highest water content was 6.1 wt. %, in agreement with the high values of  
1161 other arcs, while the  $\text{CO}_2$  in the inclusions was lost. The minimum entrapment  
1162 pressures determined by the volatile content of inclusions reaches the base of the  
1163 arc crust, indicating deep entrapment of the inclusion.
- 1164 - We found three primitive magma groups: North and South Kyushu, and a group  
1165 with little slab input (Oninomi and Fukue). The northern group systematically  
1166 shows higher Sr/Y values than the southern group. This can be explained by the  
1167 slab agent being hotter and recording a stronger slab melt signature with residual  
1168 garnet.
- 1169 - Magma at Oninomi volcano is likely produced by a small degree of mantle melt  
1170 with nearly no input from the slab. The crustal extension in the area may have  
1171 triggered such a small degree of melting.
- 1172 - Fukue is located in the back-arc and its magma with small slab input is consistent  
1173 with its geodynamic setting.
- 1174 - The sulfur isotope composition of the magma source was determined by carefully  
1175 examining the trace element systematics and checking that the melt inclusions

1176 were not influenced by any degassing processes. Careful analysis of such results  
1177 reveals that sulfur isotopes from the Kyushu arc magmas are characterized by a  
1178 mixture of the depleted mantle, slab sediment, and a third component, which could  
1179 potentially be seawater.

1180

### 1181 **Credit authorship contribution statement**

1182 Masataka Kawaguchi field, measurements and calculation. Kenneth Koga  
1183 conceptualization, field, measurements, supervision, calculation and writing. Estelle  
1184 Rose-Koga conceptualization, field, supervision and writing. Kenji Shimizu S isotope  
1185 measurements, review and editing. Takayuki Ushikubo S isotope measurements, review  
1186 and editing. Akira Yoshiasa field, supervision.

1187

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1212

1213

## 1214 **Figure captions**

1215

1216 **Fig. 1** Map of the Japanese Island arc and Kyushu sample locations. a) Map shows the tectonic  
1217 setting of Japanese Islands (after Miyoshi et al., 2008 and Mahony et al., 2011). Gray solid lines  
1218 represent the volcanic front. Major ridge and plateau of the Philippine Sea plate are indicated by  
1219 gray dashed lines. The formation ages of the Philippine Sea plate are from Okino et al. (1994,  
1220 1998) and Deschamps and Lallemand (2002), and that of the Pacific plate is from Müller et al.  
1221 (2008). Location of the study area is indicated by a red square. b) Map of Kyushu Island showing  
1222 target volcanoes. Open and solid triangles indicate the locations of Quaternary volcanoes, with  
1223 active volcanoes shown as solid red triangles. Gray solid lines and dotted lines denote the iso-  
1224 depth contours to the upper boundary of the seismic (Nakamura and Kaneshiro, 2000; Baba et al.,  
1225 2002; Nakajima and Hasegawa, 2007; Hirose et al., 2008; Saito, 2017) and aseismic parts of the  
1226 subducting plate (Zhao et al., 2012), respectively. The blue field and red dotted line mark the  
1227 inferred subducting Kyushu-Palau ridge and slab fracture, respectively (Park et al., 2009). FK:  
1228 Fukue, UZ: Unzen, TS: Tsurumi, ON: Oninomi, YF: Yufu, KJ: Kuju, AS: Aso, KR: Kirishima,  
1229 SM: Sumiyoshi-ike, SK: Sakurajima, KM: Kaimondake.

1230

1231 **Fig. 2.** Total alkali ( $\text{Na}_2\text{O} + \text{K}_2\text{O}$ ) vs.  $\text{SiO}_2$  diagram for olivine-hosted melt inclusions. Dashed  
1232 lines represent boundaries of classification of volcanic rocks (Le Maitre et al., 2002). Black solid  
1233 line indicates the boundary between alkalic and sub-alkalic volcanic rocks (Miyashiro, 1978). All  
1234 oxide concentrations have been corrected for post-entrapment crystallization and normalized to  
1235 100 wt. % as volatile-free. Reported whole rock compositions of Kyushu Island volcanic rocks  
1236 are shown as small solid circles for comparison: pink, volcanic rocks along the volcanic front;  
1237 grey, other volcanic rocks of Kyushu. WR data: Sawamura and Matsui, 1957; Shinno, 1966;  
1238 Nakamura, 1971; Nakada, 1986; Nakada and Kamata, 1988; Kakubuchi and Matsumoto, 1990;  
1239 Ohta et al., 1990, 1992, Ohta and Aoki, 1991; Iwamori et al., 1991, 1992; Nagao et al., 1992;  
1240 Kakubuchi et al., 1995; Imura and Kobayashi, 2001; Kita et al., 2001, 2012; Uto et al., 2004;

1241 Kawanabe and Sakaguchi, 2005; Sugimoto et al., 2005, 2006; Miyoshi et al., 2005, 2008a, b,  
1242 2010, 2011; Sakuyama et al., 2009, 2014; Hoang et al., 2013; Tajima et al., 2013, 2014; Shibata  
1243 et al., 2013, 2014; Kawanabe et al., 2015; Yamasaki et al., 2016; Kuritani et al., 2017; Brown et  
1244 al., 2020; Nche et al., 2021.

1245

1246 **Fig. 3.** Primitive mantle-normalized trace element compositions of melt inclusions. The evolved  
1247 melt inclusions of KSS (higher than 52 wt.% SiO<sub>2</sub> on hydrous basis) and assimilated inclusions  
1248 of ON2S are shown in different colors, as they may be influenced by secondary processes.  
1249 Composition of the primitive mantle is taken from Sun and McDonough (1989). The order of  
1250 elements after Pearce et al. (2005).

1251

1252 **Fig. 4.** H<sub>2</sub>O and CO<sub>2</sub> concentrations in melt inclusions. The concentrations have been corrected  
1253 for PEC and for gas bubbles. Gas bubble-uncorrected CO<sub>2</sub> concentrations measured in melt  
1254 inclusion are also shown in light gray. Dashed contour lines represent solubility at 1, 3 and 5 kbars  
1255 (from inside to outside) at 1100 °C, calculated for a typical basaltic melt of 49 wt.% SiO<sub>2</sub> and  
1256 1100 °C using VolatileCalc (black lines, Newman and Lowenstern, 2002) and for our K-rich melt  
1257 inclusion (FKONON-O-I) using MagmaSat (red lines, Ghiorso and Gualda, 2015). Error bar is  
1258 standard error (1σ).

1259

1260 **Fig. 5** Variations of melt inclusion compositions. (a) K<sub>2</sub>O vs. SiO<sub>2</sub> shows an absence of overall  
1261 correlation of Kyushu magmas. For individual volcanoes, Aso, Kaimondake and Kirishinma-  
1262 Shinmoedake (SM) magmas show a positive correlation and Yufu shows a weak negative  
1263 correlation. (b) Ba/Nb vs. SiO<sub>2</sub> shows a scatter of melt inclusion compositions, while showing  
1264 tight clusters for each volcano. The scatter suggests the absence of a common geochemical  
1265 process along the arc. Some individual clusters show a small variation in Ba/Nb, a ratio of two  
1266 highly incompatible elements, indicating the absence of mixing, either of the source, magma, or  
1267 crust. On the contrary, Yufu and Sumiyoshi-ike magmas show a negative slope with significant  
1268 variation of Ba/Nb, indicating the presence of mixing. Note that the evolved melt inclusions of  
1269 Aso are excluded for clarity (higher than 52 wt.% SiO<sub>2</sub> on hydrous basis).

1270

1271 **Fig. 6.** K<sub>2</sub>O-normalized volatile element abundances in the melt inclusions. (a) H<sub>2</sub>O and S show  
1272 a positive correlation (except for Oninomi, Sumiyoshi-ike, and Yufu). Such a correlation is often  
1273 interpreted as a result of magmatic degassing. However, (b) F vs. S also shows a strong correlation  
1274 (i.e. Kaimon) indicating that the correlation is not caused by H<sub>2</sub>O-loss through degassing because

1275 F is only weakly soluble in magmatic fluids. Note, the horizontal axis is on a log-scale to illustrate  
1276 the wide range of S/K<sub>2</sub>O values.

1277

1278 **Fig. 7** Comparison of measured sulfur concentrations in primitive melt inclusions with the sulfur  
1279 content at sulfide saturation (SCSS). Concentrations for all the other melt inclusions are shown  
1280 as grey symbols. Symbols are the same as in Figure 2. Dashed black line indicates the 1:1 line.  
1281 SCSS was calculated according to Fortin et al. (2015) using measured H<sub>2</sub>O concentration and the  
1282 trapping temperature ( $T_{\text{trap}}$  of Table S7) of melt inclusions at 0.1 GPa. For primitive melt  
1283 inclusions, we used minimum H<sub>2</sub>O concentrations estimated from the highest H<sub>2</sub>O/K<sub>2</sub>O ratio  
1284 (Table 3). We took the average measured H<sub>2</sub>O concentration of the melt inclusions from each  
1285 edifice and assigned this value to the melt inclusions of the same edifice for which H<sub>2</sub>O data was  
1286 missing. Decrease of SCSS caused by pressure increments does not change our conclusions, since  
1287 it does not significantly decrease the S concentration. For example, an increase in pressure from  
1288 0.1 GPa to 0.5 GPa decreases the S concentration in the primitive melt inclusions by a maximum  
1289 of 300 ppm.

1290

1291 **Fig. 8.** Sulfur isotope composition plotted against sulfur abundance. (a)  $\delta^{34}\text{S}$  vs. S plot to illustrate  
1292 isotope fractionation due to degassing. Here, there is no obvious trend, except perhaps for Yufu.  
1293 (b)  $\delta^{34}\text{S}$  vs. S/K<sub>2</sub>O plot may illustrate the degassing process better by compensating concentration  
1294 increase due to magma differentiation. Even here, Yufu, Kaimondake and Sumiyoshi-ike show a  
1295 weak negative trend. Error bars represent 2 standard errors, corresponding to approximately 95  
1296 % confidence interval.

1297

1298 **Fig.9** Slab-derived material contribution to the magma source. a) Ba/Zr vs. Nb/Zr plot. Small blue  
1299 triangles are melt inclusion and glass MORB data (Shaw et al. 2010; Wanless et al., 2012; 2014;  
1300 Le Voyer et al. 2015; 2017; 2019; Shimizu et al. 2019), and they define a trend with a slope of  
1301 Ba/Nb of 7, shown with a blue line (also called the “MORB-OIB mantle array; Leeman et al.,  
1302 1990). Small pink circles are arc melt inclusions (Straub and Layne, 2003; Churikova et al., 2007;  
1303 Elburg et al., 2007; Portnyagin et al., 2007; Sadofsky et al., 2008; Vigouroux et al., 2008;  
1304 Wysoczanski et al., 2006; Bouvier et al., 2008, 2010a,b; Sorbadere et al., 2011, 2013; Rose-Koga  
1305 et al., 2014; Walowski et al., 2016; Narvaez et al., 2018; Barker et al., 2020). Blue circles with a  
1306 cross indicate the models of the depleted mantle (Workman and Hart 2005; Salters and Stracke  
1307 2004). Purple circles with a cross indicate the model bulk silicate Earth (McDonough and Sun  
1308 1995; Palme and O’Neill, 2003; Lyubetskaya and Korenaga, 2007). All the melt inclusions from

1309 Kyushu volcanoes plot above the blue line, in the arc magma field. b) Sr/Y vs. Th/Nb plot. Melt  
1310 inclusions from volcanoes of Northern Kyushu (open symbols) have Sr/Y values higher than  
1311 those from Southern Kyushu volcanoes (filled symbols), with a Sr/Y value of 20 separating the  
1312 two groups (see text for explanation).

1313

1314 **Fig.10** Sr/Y ratio and Y concentration of melt inclusions. As in Fig. 9, pink and blue filled  
1315 symbols indicate olivine hosted melt inclusion compositions for arc magmas, and MORB,  
1316 respectively. Symbols filled in white are samples from Northern Kyushu, and filled with color  
1317 indicates samples from Southern Kyushu. Sr/Y clearly separates the north from the south.

1318

1319 **Fig.11** Mixing and melting trajectories of subduction zone endmembers are overlaid on Fig. 9ab.  
1320 Blue cloud is MORB melt inclusion data, and pink cloud is arc basaltic melt inclusion data. Green  
1321 square indicates the depleted mantle (Workman and Hart, 2005), and filled circles are slab derived  
1322 fluxes: blue, silicate melt from AOC; light blue, aqueous fluid from AOC; orange, sediment melt;  
1323 brick-red, sediment fluid (exact compositions of these fluids are in Table S8). Black lines are  
1324 mixing curves between a flux and the mantle. Metasomatized mantle plots along this curve. In  
1325 the case of three component mixing between the mantle and other fluxes, the metasomatized  
1326 mantle plots in the region between two mixing curves. Thick colored lines indicate the trajectory  
1327 of a melting model (modified after the flux model of Van den Bleeken and Koga 2015). Tick  
1328 marks indicate 5% intervals of the degree of melting ranging from 5 to 20 %. The larger degree  
1329 of melting approaches the mixing curve. It should be noted that variation caused by melting does  
1330 not deviate significantly from the mixing lines. (a) the figure illustrates a clear mantle array, and  
1331 the input from the slab is the main cause of deviation from the array. The melt flux is more  
1332 efficient than the fluid flux at raising Ba/Zr. (b) the figure indicates high Sr/Y, a signature of  
1333 residual garnet. Again, the melt flux is more efficient at producing high Sr/Y magmas than the  
1334 fluid flux.

1335

1336 **Fig. 12.** Results of mixing calculations for the primitive end members. Note that trace element  
1337 characteristics are dominantly controlled by the depleted mantle. Small additions of fluxes from  
1338 the slab are sufficient to satisfy the model. It should be noted that this calculation is based on the  
1339 incompatible element ratios. Specifically, the optimal solution for mixing was confirmed by  
1340 element ratios rather than concentration match. Thus the mixing fraction is independent of the  
1341 element fractionation by partial melting. Northern Kyushu samples are more strongly influenced  
1342 by slab signature, i.e. a slightly smaller fraction of the deleted mantle.

1343

1344 **Fig. 13.** Isotopic composition of sulfur plotted against trace element ratios. (a) Th/Nb; the plot  
1345 distinguishes AOC and sediment fluxes as illustrated by Fig. 11a. (b) Cl/F; the plot potentially  
1346 separates fluid from melt. However, there is no systematic separation. Blue circles with a cross  
1347 indicate the depleted mantle. Filled sky blue triangles are MORB values (Labidi et al., 2012;  
1348 2014). Filled pink circles are the arc melt inclusion data (Bouvier et al. 2012; Gurenko et al.,  
1349 2018; 2021), shown for comparison. Blue circles with a cross indicate the depleted mantle (trace  
1350 elements: Workman and Hart 2005; Salters and Stracke 2004, S-isotope Labidi et al., 2012, 2014).

1351

1352 **Fig. 14.** Isotopic composition of sulfur plotted against trace element ratios. (a) Rb/Ba; the plot  
1353 shows the data distribution that requires three component mixing. (b) Pb/Ce; the plot shows Pb  
1354 input from the slab, correlating with  $\delta^{34}\text{S}$ . Blue circles with cross indicate the depleted mantle.  
1355 Filled sky blue triangles are the MORB values (Labidi et al., 2012; 2014). The arc melt inclusion  
1356 data are not shown as Rb and Pb data are not available. (trace elements: Workman and Hart 2005;  
1357 Salters and Stracke 2004, S-isotope Labidi et al., 2012, 2014). Note, the samples with little slab  
1358 input (Fukue and Oninomi) plots close to the depleted mantle and MORB.

1359

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