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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 (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 ± 0.79 ‰ to
50 $+9.43 \pm 0.47$ ‰ (2σ) 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 SO₂ 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 H₂O 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 Kuju (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 μm) 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 μm and 1 μm 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 μm alumina paste was performed to remove
237 scratches on the exposed surface.

238 ***Analytical methods***

239 Volatile element concentrations (H_2O , 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 Cs^+ primary ion beam of 300 – 500 pA defocused to be 10 – 15 μm 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 μm on the sample
247 surface. Mass resolving power was set at ~ 6000 to separate mass interferences (for
248 example, to separate $^{34}\text{S}^1\text{H}$ interference from ^{35}Cl requires 5120 MRP; Burdo and

249 Morrison 1971). Negative secondary ions of ^{12}C , ^{16}OH , ^{19}F , ^{30}Si , ^{31}P , ^{32}S , and ^{35}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 ~ 6 min. Typical relative standard deviations (1σ) 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 H_2O , 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 Cs^+ ion beam of ~ 0.5 nA
263 defocused to be ~ 10 μm in diameter. A 10 keV electron beam of ~ 100 μm 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 15×15
266 μm^2 on the sample surface. Negative secondary ions of ^{32}S and ^{34}S were measured in
267 multi-detection mode with an FC and axial EM, respectively. Mass resolving power was
268 set at ~ 2200 and ~ 5000 for FC and EM detectors, respectively, to separate mass
269 interferences of $^{31}\text{P}^1\text{H}$ from ^{32}S and of $^{33}\text{S}^1\text{H}$ from ^{34}S . Each measurement consisted of 20
270 s pre-sputtering, 120 s auto-centering of ^{32}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 ~ 7
272 minutes. At CRPG, the set-up was similar except we used a stronger primary beam of 3
273 nA defocused to ~ 15 μm in diameter. Mass resolving power was set to 4500. Negative
274 secondary ions of ^{32}S and ^{34}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 ~ 5 minutes for each spot. Before
277 each measurement, we adjusted the center for ^{32}S . In both cases, the standard basaltic
278 glass, EPR-G3 ($\delta^{32}\text{S}_{\text{VCDT}} = -0.09 \pm 0.55\text{‰}$, 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}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 (2σ) 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 (2σ) 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 μm
297 depending on the size of the inclusion, and focused for olivine measurements. Relative
298 analytical uncertainties (1σ) 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 SiO_2 , 1% for Al_2O_3 , FeO and CaO, 3.5% for TiO_2 and Na_2O ,
301 10% for MnO, 1.5% for MgO, 4.5% for K_2O , 17% for P_2O_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 (1σ) 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 S^{2-} and
311 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 (1σ) was approximately

313 $\pm 10\%$ S^{6+}/S^{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 μm depending on the MI size, and a fluence of 2.8 mJ/cm^2 . Twenty-six masses were
321 collected: ^7Li , ^{45}Sc , ^{51}V , ^{65}Cu , ^{85}Rb , ^{88}Sr , ^{89}Y , ^{90}Zr , ^{93}Nb , ^{137}Ba , ^{139}La , ^{140}Ce , ^{141}Pr , ^{146}Nd ,
322 ^{147}Sm , ^{153}Eu , ^{157}Gd , ^{163}Dy , ^{166}Er , ^{172}Yb , ^{175}Lu , ^{178}Hf , ^{181}Ta , ^{208}Pb , ^{232}Th , ^{238}U . The internal
323 reference mass was ^{44}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 (1σ error of mean; σ/\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 H_2O 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 CO₂ 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 CO₂ 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 CO₂^{Melt} is the dissolved CO₂ concentration in the melt, ϕ is volume fraction of the
359 bubble in the melt inclusion, ρ_v is the volumetric CO₂ vapor density, and ρ_m is the melt
360 density, calculated from the glass composition. The ϕ 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 ρ_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 CO₂-H₂O solubility at the glass transition
365 temperature, T_{glass}, and an equation of state (EOS) for the fluid (Duan and Zhang, 2006).
366 The T_{glass} value was calculated from the temperature dependence of the viscosity at the
367 given hydrous glass composition and a constant viscosity of 10¹² Pa·s (Giordano et al.,
368 2005, 2008). The total CO₂ 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 ρ_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_{glass}, the bubble would expand without C exsolution, resulting in an

374 overestimation of ρ_v and of the total CO₂ concentration (e.g. Pichavant et al., 2013; Bali
375 et al., 2018). If this non-equilibrium expansion has occurred, the estimated total CO₂
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 “CO₂ bubble correction”. This is because the calculation of
384 total CO₂ 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 CO₂.

388 The major source of error of the total CO₂ 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 (1σ) of 37% to 48% (Tucker et al., 2019). We conservatively assigned a 50 % (1σ) relative
393 uncertainty to inclusion volumes. The uncertainties (1σ) on the total CO₂ 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 SiO₂ 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 SiO₂ 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 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 H_2O and 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. H_2O concentrations varied between 0.01 and 6.06 wt. % (Table S2), and there was
430 no systematic difference between Northern and Southern Kyushu. H_2O concentrations
431 within the same tephra sample varied less than 1 or 2 wt. % (Fig. 4). 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 SiO₂ 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 SiO₂ 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. % SiO₂, Ohta et al., 1992). While 10 out of the 11 inclusions have less than 48.5
520 wt. % SiO₂, one melt inclusion has an andesitic composition (U-I, 58.6 wt. % SiO₂, 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 ⁸⁷Sr/⁸⁶Sr 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 SiO₂-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 SiO₂ concentration. The lack of correlation with K₂O/TiO₂ or CaO/Al₂O₃ 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 SiO₂ 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 H₂O and CO₂, 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 H₂O and CO₂ content directly from
587 geological materials. In fact, because of the high-diffusivity of H₂O in host-olivines,
588 measured H₂O 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 H₂O 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 CO₂, 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 CO₂ degassing
595 starts before entrapment of the melt in the host olivine, the primitive CO₂ composition
596 cannot be trapped in a melt inclusion.

597 In this study, several methods were combined to estimate the minimum H₂O 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 H₂O values from 2.8
600 to 6.1 wt. % (Table S2). These would commonly qualify as “high-H₂O” values for the arc
601 magma (e.g. see compilation in Cooper et al., 2012). Alternatively, the H₂O 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. % H₂O (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 H₂O estimate for the primitive magma. For example,
608 for the case of Aso, the plagioclase hygrometer gives an H₂O concentration of 4.7 wt.%,
609 while measured H₂O 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 H₂O content.

612 Another method for estimating H₂O content in magma is to adopt the maximum
613 H₂O/K₂O 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 H₂O loss, if a magmatic suite shares the same parental
616 magma. In the case of mixed magmas, H₂O/K₂O of the mafic endmember likely defines
617 the least degassed ratio. This K₂O normalization also eliminates the effect of post-
618 entrapment crystallization. Specifically, we looked for the highest H₂O/K₂O values of the
619 melt inclusions in this study and the whole rock H₂O/K₂O from previous studies, in which
620 H₂O content from the plagioclase hygrometer is combined with measured K₂O
621 concentration (Table 3; Zellmer et al., 2012; 2014; Kawaguchi et al., 2021). The H₂O
622 concentration is then calculated by multiplying the highest H₂O/K₂O ratio by the K₂O
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 H₂O in

625 the inclusions was re-equilibrated to low values. It should be noted that the highest
626 H₂O/K₂O 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 H₂O/K₂O is not that of
630 the selected inclusions representing primitive magma compositions. For Kuju, the
631 difference in H₂O/K₂O 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 H₂O concentrations; H₂O/K₂O of the magma cannot be assumed to be constant
636 throughout mixing. In addition, judging from the low H₂O concentrations, these melt
637 inclusions lost a significant amount of their initial water. While we used the same
638 approach to present the highest H₂O concentrations, those of Yufu and Sumiyoshi-ike
639 remain truly minimum estimates.

640 We used the maximum CO₂ concentration of each sample as the minimum estimate
641 for the primitive value, using a similar reasoning as for H₂O. We considered the bubbles
642 present in melt inclusions as an exsolved fluid phase, and “added the CO₂ back” to the
643 melt using the equation of state method (e.g., Tucker et al., 2019). The CO₂ 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 CO₂ 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 H₂O and CO₂ 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 ± 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 ± 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 S⁶⁺ in the
724 magmas. However, the measured S speciation gives negligible S⁶⁺ 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 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 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 2σ 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 ~ 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 K_2O , (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 (~ 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 ± 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 ± 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 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. 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.% SiO₂ 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.** H₂O and CO₂ concentrations in melt inclusions. The concentrations have been corrected
1253 for PEC and for gas bubbles. Gas bubble-uncorrected CO₂ 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.% SiO₂ 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) K₂O vs. SiO₂ 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. SiO₂ 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.% SiO₂ on hydrous basis).

1270

1271 **Fig. 6.** K₂O-normalized volatile element abundances in the melt inclusions. (a) H₂O 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 H₂O-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/K₂O 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 H₂O concentration and the
1282 trapping temperature (T_{trap} of Table S7) of melt inclusions at 0.1 GPa. For primitive melt
1283 inclusions, we used minimum H₂O concentrations estimated from the highest H₂O/K₂O ratio
1284 (Table 3). We took the average measured H₂O concentration of the melt inclusions from each
1285 edifice and assigned this value to the melt inclusions of the same edifice for which H₂O 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/K₂O 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

1360 **References**

1361

- 1362 Abe, Y., Ohkura, T., Hirahara, K. & Shibutani, T. (2013). Along-arc variation in water
1363 distribution in the uppermost mantle beneath Kyushu, Japan, as derived from receiver
1364 function analyses. *Journal of Geophysical Research: Solid Earth* **118**, 3540–3556.
- 1365 Albers, E., Kahl, W.-A., Beyer, L. & Bach, W. (2020). Variant across-forearc compositions of
1366 slab-fluids recorded by serpentinites: implications on the mobilization of FMEs from an
1367 active subduction zone (Mariana forearc). *Lithos* **364–365**, 105525.
- 1368 Alt, J.C. and Burdett, J.W. (1992) Sulfur in Pacific deep-sea sediments (Leg 129) and
1369 implications for cycling of sediment in subduction zones, Proc. ODP Sci. Res.
1370 **129**, 283-294.
- 1371 Alt, J. C. III., Schwarzenbach, E. M., Früh-Green, G. L., Shanks, W. C., Bernasconi, S. M.,
1372 Garrido, C. J., Crispini, L., Gaggero, L., Padrón-Navarta, J. A. & Marchesi, C. (2013).
1373 The role of serpentinites in cycling of carbon and sulfur: seafloor serpentinization and
1374 subduction metamorphism. *Lithos* **178**, 40–54.
- 1375 Alt, J. C. & Shanks, W. C. (2011). Microbial sulfate reduction and the sulfur budget for a

1376 complete section of altered oceanic basalts, IODP Hole 1256D (eastern Pacific). *Earth*
1377 *and Planetary Science Letters* **310**, 73–83.

1378 Alt, J. C., Shanks, W. C. III. & Jackson, M. C. (1993). Cycling of sulfur in subduction zones:
1379 the geochemistry of sulfur in the Mariana Island Arc and back-arc trough. *Earth and*
1380 *Planetary Science Letters* **119**, 477–494.

1381 Asamori, K. & Zhao, D. (2015). Teleseismic shear wave tomography of the Japan subduction
1382 zone. *Geophysical Journal International* **203**, 1752–1772.

1383 Baba, T., Tanioka, Y., Cummins, P. R. & Uhira, K. (2002). The slip distribution of the 1946
1384 Nankai earthquake estimated from tsunami inversion using a new plate model. *Physics of*
1385 *the Earth and Planetary Interiors* **132**, 59–73.

1386 Bali, E., Hartley, M. E., Halldórsson, S. A., Gudfinnsson, G. H. & Jakobsson, S. (2018). Melt
1387 inclusion constraints on volatile systematics and degassing history of the 2014–2015
1388 Holuhraun eruption, Iceland. *Contributions to Mineralogy and Petrology*. Springer Berlin
1389 Heidelberg **173**, 9.

1390 Barker, S. J., Rowe, M. C., Wilson, C. J. N., Gamble, J. A., Rooyakkers, S. M., Wysoczanski,
1391 R. J., Illsley-Kemp, F. & Kenworthy, C. C. (2020). What lies beneath? Reconstructing the
1392 primitive magmas fueling voluminous silicic volcanism using olivine-hosted melt
1393 inclusions. *Geology* **48**, 504–508.

1394 Beaudry, P., Longpré, M.A., Economos, R., Boswell, A.W., Thi Hao, B. & Stix, J. (2018)
1395 Degassing-induced fractionation of multiple sulphur isotopes unveils post-Archaean
1396 recycled oceanic crust signal in hotspot lava. *Nature Communications* **9**, 5093,
1397 <https://doi.org/10.1038/s41467-018-07527-w>

1398 Bénard, A., Klimm, K., Woodland, A.B., Arculus, R.J., Wilke, M., Botcharnikov, R.E.,
1399 Shimizu, N., Nebel, O., Rivard, C. and Ionov, D.A. (2018) Oxidising agents in sub-arc
1400 mantle melts link slab devolatilisation and arc magmas, *Nature Communications*, DOI:
1401 [10.1038/s41467-018-05804-2](https://doi.org/10.1038/s41467-018-05804-2)

1402 Bouvier, A.-S., Deloule, E. & Métrich, N. (2010a). Fluid inputs to magma sources of
1403 St. Vincent and Grenada (Lesser Antilles): new insights from trace elements in olivine-
1404 hosted melt inclusions. *Journal of Petrology* **51**, 1597–1615.

1405 Bouvier, A.-S., Métrich, N. & Deloule, E. (2008). Slab-derived fluids in the magma sources of
1406 St. Vincent (Lesser Antilles arc): volatile and light element imprints. *Journal of Petrology*
1407 **49**, 1427–1448.

1408 Bouvier, A.-S., Métrich, N. & Deloule, E. (2010b). Light elements, volatiles, and stable
1409 isotopes in basaltic melt inclusions from Grenada, Lesser Antilles: inferences for magma

1410 genesis. *Geochemistry, Geophysics, Geosystems* **11**, Q09004.

1411 Brown, J. R., Taylor, R. N. & Iguchi, M. (2020). Using high-resolution Pb isotopes to unravel
1412 the petrogenesis of Sakurajima volcano, Japan. *Bulletin of Volcanology* **82**, 36.

1413 Bucholz, C. E., Gaetani, G. A., Behn, M. D. & Shimizu, N. (2013). Post-entrapment
1414 modification of volatiles and oxygen fugacity in olivine-hosted melt inclusions. *Earth
1415 and Planetary Science Letters* **374**, 145–155.

1416 Burdo, R. A. & Morrison, H. G. (1971). *Table of atomic and molecular lines for spark source
1417 mass spectrometry of complex sample-graphite mixes. Report 1670*. Ithaca, New York:
1418 Materials Science Center, Cornell University.

1419 Canfield, D.E., Farquhar, J., (2009). Animal evolution, bioturbation, and the sulfate
1420 concentration of the oceans. *Proc. Natl. Acad. Sci. USA* **106**, 20, 8123–8127.

1421 Carn, S. A., Fioletov, V. E., McLinden, C. A., Li, C. & Krotkov, N. A. (2017). A decade of
1422 global volcanic SO₂ emissions measured from space. *Scientific Reports* **7**, 44095.

1423 Carroll, M. R. & Webster, J. D. (1994). Solubilities of sulfur, noble gases, nitrogen, chlorine
1424 and fluorine in magmas. *Reviews in Mineralogy and Geochemistry* **30**, 231–280.

1425 Chen, Y., Provost, A., Schiano, P. & Cluzel, N. (2011). The rate of water loss from olivine-
1426 hosted melt inclusions. *Contributions to Mineralogy and Petrology* **162**, 625–636.

1427 Churikova, T., Wörner, G., Mironov, N. & Kronz, A. (2007). Volatile (S, Cl and F) and fluid
1428 mobile trace element compositions in melt inclusions: implications for variable fluid
1429 sources across the Kamchatka arc. *Contributions to Mineralogy and Petrology* **154**, 217–
1430 239.

1431 Cooper, L. B., Ruscitto, D. M., Plank, T., Wallace, P. J., Syracuse, E. M. & Manning, C. E.
1432 (2012). Global variations in H₂O/Ce: 1. Slab surface temperatures beneath volcanic arcs.
1433 *Geochemistry, Geophysics, Geosystems* **13**, Q03024.

1434 Cottrell, E., Spiegelman, M. & Langmuir, C. H. (2002). Consequences of diffusive
1435 reequilibration for the interpretation of melt inclusions. *Geochemistry, Geophysics,
1436 Geosystems* **3**, 1–26.

1437 Cruse, A. M. & Lyons, T. W. (2004). Trace metal records of regional paleoenvironmental
1438 variability in Pennsylvanian (Upper Carboniferous) black shales. *Chemical Geology* **206**,
1439 319–345.

1440 Danyushevsky, L. V., Della-Pasqua, F. N. & Sokolov, S. (2000). Re-equilibration of melt
1441 inclusions trapped by magnesian olivine phenocrysts from subduction-related magmas:
1442 petrological implications. *Contributions to Mineralogy and Petrology* **138**, 68–83.

1443 Danyushevsky, L. V., McNeill, A. W. & Sobolev, A. V. (2002). Experimental and petrological

1444 studies of melt inclusions in phenocrysts from mantle-derived magmas: an overview of
1445 techniques, advantages and complications. *Chemical Geology* **183**, 5–24.

1446 Davidson, J., Turner, S., Handley, H., Macpherson, C. & Dosseto, A. (2007). Amphibole
1447 “sponge” in arc crust? *Geology* **35**, 787–790.

1448 Defant, M. J. & Drummond, M. S. (1990). Derivation of some modern arc magmas by melting
1449 of young subducted lithosphere. *Nature* **347**, 662–665.

1450 DePaolo, D. J. (1981). Trace element and isotopic effects of combined wallrock assimilation
1451 and fractional crystallization. *Earth and Planetary Science Letters* **53**, 189–202.

1452 Deschamps, A. & Lallemand, S. (2002). The West Philippine Basin: an Eocene to early
1453 Oligocene back arc basin opened between two opposed subduction zones. *Journal of*
1454 *Geophysical Research: Solid Earth* **107**, 2322.

1455 Deschamps, F., Guillot, S., Godard, M., Chauvel, C., Andreani, M. & Hattori, K. (2010). In
1456 situ characterization of serpentinites from forearc mantle wedges: timing of
1457 serpentinitization and behavior of fluid-mobile elements in subduction zones. *Chemical*
1458 *Geology* **269**, 262–277.

1459 Dixon, J. E., Clague, D. A. & Stolper, E. M. (1991). Degassing history of water, sulfur, and
1460 carbon in submarine lavas from Kilauea volcano, Hawaii. *Journal of Geology* **99**, 371–
1461 394.

1462 Duan, Z. & Zhang, Z. (2006). Equation of state of the H₂O, CO₂, and H₂O–CO₂ systems up to
1463 10 GPa and 2573.15 K: molecular dynamics simulations with ab initio potential surface.
1464 *Geochimica et Cosmochimica Acta* **70**, 2311–2324.

1465 Elburg, M. A., Kamenetsky, V. S., Foden, J. D. & Sobolev, A. (2007). The origin of medium-
1466 K ankaramitic arc magmas from Lombok (Sunda arc, Indonesia): mineral and melt
1467 inclusion evidence. *Chemical Geology* **240**, 260–279.

1468 Elliott, T. (2003). Tracers of the slab. In: Eiler, J. M. (ed.) *Inside the Subduction Factory*.
1469 American Geophysical Union, 23–45.

1470 Evans, K. A., Tomkins, A. G., Cliff, J. & Fiorentini, M. L. (2014). Insights into subduction
1471 zone sulfur recycling from isotopic analysis of eclogite-hosted sulfides. *Chemical*
1472 *Geology* **365**, 1–19.

1473 Ferriss, E., Plank, T., Newcombe, M., Walker, D. & Hauri, E. (2018). Rates of dehydration of
1474 olivines from San Carlos and Kilauea Iki. *Geochimica et Cosmochimica Acta* **242**, 165–
1475 190.

1476 Fiege, A., Holtz, F., Behrens, H., Mandeville, C. W., Shimizu, N., Crede, L. S. & Göttlicher, J.
1477 (2015). Experimental investigation of the S and S-isotope distribution between H₂O–S ±

1478 Cl fluids and basaltic melts during decompression. *Chemical Geology* **393–394**, 36–54.

1479 Foley, S. F., Barth, M. G. & Jenner, G. A. (2000). Rutile/melt partition coefficients for trace
1480 elements and an assessment of the influence of rutile on the trace element characteristics
1481 of subduction zone magmas. *Geochimica et Cosmochimica Acta* **64**, 933–938.

1482 Fortin, M.-A., Riddle, J., Desjardins-Langlais, Y. & Baker, D. R. (2015). The effect of water
1483 on the sulfur concentration at sulfide saturation (SCSS) in natural melts. *Geochimica et*
1484 *Cosmochimica Acta* **160**, 100–116.

1485 Fourcade, S. & Allegre, C. J. (1981). Trace element behavior in granite genesis: a case study
1486 The calc-alkaline plutonic association from the Querigut Complex (Pyrénées, France).
1487 *Contributions to Mineralogy and Petrology* **76**, 177–195.

1488 Fujino, N. & Kobayashi, T. (1997). Eruption history of Kaimondake volcano, Southern
1489 Kyushu, Japan. *Bulletin of the volcanological society of Japan* **42**, 195–211 (in Japanese
1490 with English abstract).

1491 Gaetani, G. A., O’Leary, J. A., Shimizu, N., Bucholz, C. E. & Newville, M. (2012). Rapid
1492 reequilibration of H₂O and oxygen fugacity in olivine-hosted melt inclusions. *Geology*
1493 **40**, 915–918.

1494 Gagnon, J. E., Fryer, B. J., Samson, I. M. & Williams-Jones, A. E. (2008). Quantitative
1495 analysis of silicate certified reference materials by LA-ICPMS with and without an
1496 internal standard. *Journal of Analytical Atomic Spectrometry* **23**, 1529–1537.

1497 Gao, S., Liu, X., Yuan, H., Hattendorf, B., Günther, D., Chen, L. & Hu, S. (2002).
1498 Determination of Forty Two Major and Trace Elements in USGS and NIST SRM Glasses
1499 by Laser Ablation-Inductively Coupled Plasma-Mass Spectrometry. *Geostandards and*
1500 *Geoanalytical Research* **26**, 181–196.

1501 Ghiorsso, M. S. & Gualda, G. A. R. (2015). An H₂O–CO₂ mixed fluid saturation model
1502 compatible with rhyolite-MELTS. *Contributions to Mineralogy and Petrology* **169**, 1–30.

1503 Ghiorsso, M. S. & Sack, R. O. (1995). Chemical mass transfer in magmatic processes IV. A
1504 revised and internally consistent thermodynamic model for the interpolation and
1505 extrapolation of liquid-solid equilibria in magmatic systems at elevated temperatures and
1506 pressures. *Contributions to Mineralogy and Petrology* **119**, 197–212.

1507 Giordano, D., Nichols, A. R. L. & Dingwell, D. B. (2005). Glass transition temperatures of
1508 natural hydrous melts: A relationship with shear viscosity and implications for the
1509 welding process. *Journal of Volcanology and Geothermal Research* **142**, 105–118.

1510 Giordano, D., Russell, J. K. & Dingwell, D. B. (2008). Viscosity of magmatic liquids: a model.
1511 *Earth and Planetary Science Letters* **271**, 123–134.

- 1512 Gualda, G. A. R., Ghiorso, M. S., Lemons, R. V. & Carley, T. L. (2012). Rhyolite-MELTS: a
1513 modified calibration of MELTS optimized for silica-rich, fluid-bearing magmatic
1514 systems. *Journal of Petrology* **53**, 875–890.
- 1515 Gurenko, A. A., Belousov, A. B., Kamenetsky, V. S. & Zelenski, M. E. (2018). Origin of
1516 volatiles emitted by Plinian mafic eruptions of the Chikurachki volcano, Kurile arc,
1517 Russia: trace element, boron and sulphur isotope constraints. *Chemical Geology* **478**,
1518 131–147.
- 1519 Gurenko, A. A. (2021). Origin of sulphur in relation to silicate-sulphide immiscibility in
1520 Tolbachik primitive arc magma (Kamchatka, Russia): insights from sulphur and boron
1521 isotopes. *Chemical Geology* **576**, 120244.
- 1522 Hart, S. R. & Gaetani, G. A. (2006). Mantle Pb paradoxes: the sulfide solution. *Contributions*
1523 *to Mineralogy and Petrology* **152**, 295–308.
- 1524 Hermann, J. (2002). Allanite: thorium and light rare earth element carrier in subducted crust.
1525 *Chemical Geology* **192**, 289–306.
- 1526 Hickey-Vargas, R., Yogodzinski, G. M., Ishizuka, O., McCarthy, A., Bizimis, M., Kusano, Y.,
1527 Savov, I. P. & Arculus, R. (2018). Origin of depleted basalts during subduction initiation
1528 and early development of the Izu-Bonin-Mariana island arc: evidence from IODP
1529 expedition 351 site U1438, Amami-Sankaku basin. *Geochimica et Cosmochimica Acta*
1530 **229**, 85–111.
- 1531 Hirata, Y., Ueta, K., Miyawaki, R., Iemura, K., Yokoyama, T. & Miyawaki, A. (2020). Tephra
1532 stratigraphy and an about 4000-year-old slope failure on Janoo volcano, NW Aso caldera.
1533 *Transactions, Japanese Geomorphological Union* **41**, 27-47 (in Japanese with English
1534 abstract).
- 1535 Hirose, F., Nakajima, J. & Hasegawa, A. (2008). Three-dimensional seismic velocity structure
1536 and configuration of the Philippine Sea slab in southwestern Japan estimated by double-
1537 difference tomography. *Journal of Geophysical Research: Solid Earth* **113**, B09315.
- 1538 Huang, Z., Zhao, D., Hasegawa, A., Umino, N., Park, J.-H. & Kang, I.-B. (2013). Aseismic
1539 deep subduction of the Philippine Sea plate and slab window. *Journal of Asian Earth*
1540 *Sciences* **75**, 82–94.
- 1541 Imura, R. & Kobayashi, T. (2001). *Geological map of Kirishima volcano, 1:50,000*.
1542 *Geological map of volcanoes 11*.
- 1543 Inoue, K. (1985). Lateral variations of K₂O, Rb in Kirishima volcano. *Programme and*
1544 *abstracts of the Volcanological Society of Japan 2017 fall meeting* 313 (in Japanese).
- 1545 Iriyama, Y. & Toramaru, A. (2015). Stratigraphic variations in grain-size characteristics of

1546 pyroclastic fall deposits during the 2011 subplinian eruption of Shinmoedake, Kirishima
1547 volcano, Japan. *Bulletin of the volcanological society of Japan* **60**, 399–410 (in Japanese
1548 with English abstract).

1549 Ishihara, S. & Chappell, B. W. (2010). Chemical compositions of the Miocene granitoids of the
1550 Okueyama, Hoei mine and Takakumayama plutons, Outer Zone of SW Japan. *Bulletin of*
1551 *the Geological Survey of Japan* **61**, 17–38.

1552 Ishizuka, O., Hickey-Vargas, R., Arculus, R. J., Yogodzinski, G. M., Savov, I. P., Kusano, Y.,
1553 McCarthy, A., Brandl, P. A. & Sudo, M. (2018). Age of Izu–Bonin–Mariana arc
1554 basement. *Earth and Planetary Science Letters*. Elsevier B.V. **481**, 80–90.

1555 Iwamori, H. (1991). Zonal structure of Cenozoic basalts related to mantle upwelling in
1556 southwest Japan. *Journal of Geophysical Research* **96**, 6157–6170.

1557 Iwamori, H. (1992). Degree of melting and source composition of Cenozoic basalts in
1558 southwest Japan: evidence for mantle upwelling by flux melting. *Journal of Geophysical*
1559 *Research: Solid Earth* **97**, 10983–10995.

1560 Iwamori, H. (2007). Transportation of H₂O beneath the Japan arcs and its implications for
1561 global water circulation. *Chemical Geology* **239**, 182–198.

1562 Japan Meteorological Agency (2020). *Results of volcanic gas observation at Aso volcano*.
1563 [http://www.data.jma.go.jp/svd/vois/data/fukuoka/rovdm/Asosan_rovdm/Asosan_rovdm.h](http://www.data.jma.go.jp/svd/vois/data/fukuoka/rovdm/Asosan_rovdm/Asosan_rovdm.html)
1564 [tml](http://www.data.jma.go.jp/svd/vois/data/fukuoka/rovdm/Asosan_rovdm.html) (accessed 20 April 2020).

1565 Jarosewich, E., Nelen, J. A. & Norberg, J. A. (1980). Reference samples for electron
1566 microprobe analysis. *Geostandards Newsletter* **4**, 43–47.

1567 Jugo, P. J., Luth, R. W. & Richards, J. P. (2005). An experimental study of the sulfur content
1568 in basaltic melts saturated with immiscible sulfide or sulfate liquids at 1300 °C and 1.0
1569 GPa. *Journal of Petrology* **46**, 783–798.

1570 Jugo, P. J., Wilke, M. & Botcharnikov, R. E. (2010). Sulfur K-edge XANES analysis of natural
1571 and synthetic basaltic glasses: implications for S speciation and S content as function of
1572 oxygen fugacity. *Geochimica et Cosmochimica Acta* **74**, 5926–5938.

1573 Kakubuchi, S., Kido, M. & Hikosan Collaborative Research Group (1995). High-magnesian
1574 andesites from the northwestern margin of the Pliocene Kuju-Beppu Tectonic Basin,
1575 Northern Kyushu, Japan. *Memoir of the Geological Society of Japan* **44**, 125–138 (in
1576 Japanese with English abstract).

1577 Kakubuchi, S. & Matsumoto, Y. (1990). Primitive tholeiitic basalt from the Yabakei district,
1578 Oita Prefecture, southwest Japan. *Journal of Mineralogy, Petrology and Economic*
1579 *Geology* **85**, 559–568.

- 1580 Kamata, H. & Kobayashi, T. (1997). The eruptive rate and history of Kuju volcano in Japan
1581 during the past 15,000 years. *Journal of Volcanology and Geothermal Research* **76**, 163–
1582 171.
- 1583 Kamata, H. & Kodama, K. (1994). Tectonics of an arc-arc junction: an example from Kyushu
1584 Island at the junction of the southwest Japan arc and the Ryukyu arc. *Tectonophysics* **233**,
1585 69–81.
- 1586 Kamenetsky, V. S., Sobolev, A. V., Eggins, S. M., Crawford, A. J. & Arculus, R. J. (2002).
1587 Olivine-enriched melt inclusions in chromites from low-Ca boninites, Cape Vogel, Papua
1588 New Guinea: evidence for ultramafic primary magma, refractory mantle source and
1589 enriched components. *Chemical Geology* **183**, 287–303.
- 1590 Kanehira, K., Yui, S., Sakai, H. & Sasaki, A. (1973). Sulphide globules and sulphur isotope
1591 ratios in the abyssal tholeiite from the Mid-Atlantic Ridge near 30°N latitude.
1592 *Geochemical Journal* **7**, 89–96.
- 1593 Kawaguchi, M., Hasenaka, T., Koga, K. T., Rose-Koga, E. F., Yasuda, A., Hokanishi, N.,
1594 Mori, Y., Shimizu, K. & Ushikubo, T. (2021). Persistent gas emission originating from a
1595 deep basaltic magma reservoir of an active volcano: the case of Aso volcano, Japan.
1596 *Contributions to Mineralogy and Petrology* **176**, 6.
- 1597 Kawamoto, T., Yoshikawa, M., Kumagai, Y., Mirabueno, M. H. T., Okuno, M. & Kobayashi,
1598 T. (2013). Mantle wedge infiltrated with saline fluids from dehydration and
1599 decarbonation of subducting slab. *Proceedings of the National Academy of Sciences* **110**,
1600 9663–9668.
- 1601 Kawanabe, Y., Hoshizumi, H., Itoh, J. & Yamasaki, S. (2015). *Geological map of Kuju*
1602 *volcano. Geological map of volcanoes 19.*
- 1603 Kawanabe, Y. & Sakaguchi, K. (2005). *Geology of the Kaimon Dake district. Quadrangle*
1604 *Series, 1:50,000.* Geological Survey of Japan, AIST.
- 1605 Kelley, K. A. & Cottrell, E. (2009). Water and the oxidation state of subduction zone magmas.
1606 *Science* **325**, 605–607.
- 1607 Kessel, R., Schmidt, M. W., Ulmer, P. & Pettke, T. (2005). Trace element signature of
1608 subduction-zone fluids, melts and supercritical liquids at 120-180 km depth. *Nature* **437**,
1609 724–727.
- 1610 Kilinc, A., Carmichael, I. S. E., Rivers, M. L. & Sack, R. O. (1983). The ferric-ferrous ratio of
1611 natural silicate liquids equilibrated in air. *Contributions to Mineralogy and Petrology* **83**,
1612 136–140.
- 1613 Kiminami, K., Kishita, S. & Imaoka, T. (2009). Marked change in sandstone composition

1614 during the Middle Jurassic in Jurassic accretionary complexes of SW Japan, and geologic
1615 significance. *The Journal of the Geological Society of Japan* **115**, 578-596 (in Japanese
1616 with English abstract).

1617 Kimura, J.-I. (2017). Modeling chemical geodynamics of subduction zones using the Arc
1618 Basalt Simulator version 5. *Geosphere***13**, 992–1025.

1619 Kita, I., Asakawa, Y., Yuri, T., Yasui, M., Shimoike, Y., Yamamoto, M., Hasegawa, H.,
1620 Taguchi, S. & Sumino, H. (2012). Rifting of Kyushu, Japan, based on the fault-controlled
1621 concurrent eruption of oceanic island basalt-type and island arc-type lavas. *Bulletin of*
1622 *Volcanology* **74**, 1121–1139.

1623 Kita, I., Yamamoto, M., Asakawa, Y., Nakagawa, M., Taguchi, S. & Hasegawa, H. (2001).
1624 Contemporaneous ascent of within-plate type and island-arc type magmas in the Beppu-
1625 Shimabara graben system, Kyushu island, Japan. *Journal of Volcanology and Geothermal*
1626 *Research* **111**, 99–109.

1627 Klemme, S., Prowatke, S., Hametner, K. & Günther, D. (2005). Partitioning of trace elements
1628 between rutile and silicate melts: implications for subduction zones. *Geochimica et*
1629 *Cosmochimica Acta* **69**, 2361–2371.

1630 Klimm, K., Blundy, J. D. & Green, T. H. (2008). Trace element partitioning and accessory
1631 phase saturation during H₂O-saturated melting of basalt with implications for subduction
1632 zone chemical fluxes. *Journal of Petrology* **49**, 523–553.

1633 Kuritani, T., Sakuyama, T., Kamada, N., Yokoyama, T. & Nakagawa, M. (2017). Fluid-fluxed
1634 melting of mantle versus decompression melting of hydrous mantle plume as the cause of
1635 intraplate magmatism over a stagnant slab: implications from Fukue volcano group, SW
1636 Japan. *Lithos* **282–283**, 98–110.

1637 Labidi, J., Cartigny, P., Birck, J. L., Assayag, N. & Bourrand, J. J. (2012). Determination of
1638 multiple sulfur isotopes in glasses: a reappraisal of the MORB $\delta^{34}\text{S}$. *Chemical Geology*
1639 **334**, 189–198.

1640 Labidi, J., Cartigny, P., Hamelin, C., Moreira, M. & Dosso, L. (2014). Sulfur isotope budget
1641 (^{32}S , ^{33}S , ^{34}S and ^{36}S) in Pacific-Antarctic ridge basalts: a record of mantle source
1642 heterogeneity and hydrothermal sulfide assimilation. *Geochimica et Cosmochimica Acta*
1643 **133**, 47–67.

1644 Lange, R. A., Frey, H. M. & Hector, J. (2009). A thermodynamic model for the plagioclase-
1645 liquid hygrometer/thermometer. *American Mineralogist* **94**, 494–506.

1646 Lara, M. & Dasgupta, R. (2020). Partial melting of a depleted peridotite metasomatized by a
1647 MORB-derived hydrous silicate melt – Implications for subduction zone magmatism.

- 1648 *Geochimica et Cosmochimica Acta* **290**, 137–161.
- 1649 Leeman, W.P., Smith, D.R., Hildreth, W., Palcz, Z., Rogers, N. (1990) Compositional diversity
1650 of late Cenozoic basalts in a transect across the Southern Washington Cascades:
1651 implications for subduction zone magmatism, *Journal of Geophysical Research*, **95**,
1652 19,561-19,582.
- 1653 Le Maitre, R. W., Streckeisen, A., Zanettin, B., Le Bas, M. J., Bonin, B. & Bateman, P. (2002).
1654 *Igneous rocks: A classification and glossary of terms: recommendations of the*
1655 *International Union of Geological Sciences subcommission on the systematics of igneous*
1656 *Rocks (2nd ed.)*. Cambridge: Cambridge University Press.
- 1657 Lerner, A.H., Wallace, P.J., Shea, T., Mourey, A.J., · Kelly, P.J., · Nadeau, P.A., Elias, T., ·
1658 Kern, C., Clor, L.E., · Gansecki, C., · Lee, R.L., · Moore, L.R., · Werner C.A. (2021) The
1659 petrologic and degassing behavior of sulfur and other magmatic volatiles from the 2018
1660 eruption of Kīlauea, Hawai'i: melt concentrations, magma storage depths, and magma
1661 recycling. *Bulletin of Volcanology* **83**, 43, <https://doi.org/10.1007/s00445-021-01459-y>
- 1662 Le Voyer, M., Cottrell, E., Kelley, K. A., Brounce, M. & Hauri, E. H. (2015). The effect of
1663 primary versus secondary processes on the volatile content of MORB glasses: an example
1664 from the equatorial Mid-Atlantic Ridge (5°N–3°S). *Journal of Geophysical Research:*
1665 *Solid Earth* **120**, 125–144.
- 1666 Le Voyer, M., Hauri, E. H., Cottrell, E., Kelley, K. A., Salters, V. J. M., Langmuir, C. H.,
1667 Hilton, D. R., Barry, P. H. & Füre, E. (2019). Carbon fluxes and primary magma CO₂
1668 contents along the global mid-ocean ridge system. *Geochemistry, Geophysics,*
1669 *Geosystems* **20**, 1387–1424.
- 1670 Le Voyer, M., Kelley, K. A., Cottrell, E. & Hauri, E. H. (2017). Heterogeneity in mantle
1671 carbon content from CO₂-undersaturated basalts. *Nature Communications*. Nature
1672 Publishing Group **8**, 14062.
- 1673 Le Voyer, M., Rose-Koga, E. F., Shimizu, N., Grove, T. L. & Schiano, P. (2010). Two
1674 contrasting H₂O-rich components in primary melt inclusions from Mount Shasta. *Journal*
1675 *of Petrology* **51**, 1571–1595.
- 1676 Lee, C.-T. A., Erdman, M., Yang, W., Ingram, L., Chin, E. J. & DePaolo, D. J. (2018). Sulfur
1677 isotopic compositions of deep arc cumulates. *Earth and Planetary Science Letters* **500**,
1678 76–85.
- 1679 Leeman, W. P., Lewis, J. F., Evarts, R. C., Conrey, R. M. & Streck, M. J. (2005). Petrologic
1680 constraints on the thermal structure of the Cascades arc. *Journal of Volcanology and*
1681 *Geothermal Research* **140**, 67–105.
- 1682 Lesne, P., Kohn, S. C., Blundy, J., Witham, F., Botcharnikov, R. E. & Behrens, H. (2011).

- 1683 Experimental simulation of closed-system degassing in the system basalt–H₂O–CO₂–S–
1684 Cl. *Journal of Petrology* **52**, 1737–1762.
- 1685 Lew M., (1981) The distribution of some major and trace elements in sediments of the
1686 Atlantic Ocean (DSDP samples) 1. The distribution of sulphur isotopes, and Mn,
1687 Fe, Zn and Cu, *Chem. Geol.* **33**, 205-224.
- 1688 Li, C. & Ripley, E. M. (2009). Sulfur contents at sulfide-liquid or anhydrite saturation in
1689 silicate melts: empirical equations and example applications. *Economic Geology* **104**,
1690 405–412.
- 1691 Li, Y.-H. & Schoonmaker, J. E. (2003). Chemical composition and mineralogy of marine
1692 sediments. *Treatise on Geochemistry* **7**, 1–35.
- 1693 Li, J-L., Schwarzenbach, E.M., John, T., Ague, J.J., Haung, F., Gao, J., Klemd, R, Whitehouse,
1694 M.J. and Wang X-S. (2020) Uncovering and quantifying the subduction zone sulfur cycle
1695 from the slab perspective, Nature Communications, doi.org/10.1038/s41467-019-14110-4.
- 1696 Liu, X. & Zhao, D. (2016). P and S wave tomography of Japan subduction zone from joint
1697 inversions of local and teleseismic travel times and surface-wave data. *Physics of the*
1698 *Earth and Planetary Interiors* **252**, 1–22.
- 1699 Lloyd, A. S., Plank, T., Ruprecht, P., Hauri, E. H. & Rose, W. (2013). Volatile loss from melt
1700 inclusions in pyroclasts of differing sizes. *Contributions to Mineralogy and Petrology*
1701 **165**, 129–153.
- 1702 Lyubetskaya, T. & Korenaga, J. (2007). Chemical composition of Earth’s primitive mantle and
1703 its variance: 1. method and results. *Journal of Geophysical Research: Solid Earth* **112**,
1704 B03211.
- 1705 Mahony, S. H., Wallace, L. M., Miyoshi, M., Villamor, P., Sparks, R. S. J. & Hasenaka, T.
1706 (2011). Volcano-tectonic interactions during rapid plate-boundary evolution in the
1707 Kyushu region, SW Japan. *Bulletin of the Geological Society of America* **123**, 2201–
1708 2223.
- 1709 Mandeville, C. W., Sasaki, A., Saito, G., Faure, K., King, R. & Hauri, E. (1998). Open-system
1710 degassing of sulfur from Krakatau 1883 magma. *Earth and Planetary Science Letters*
1711 **160**, 709–722.
- 1712 Mandeville, C. W., Webster, J. D., Tappen, C., Taylor, B. E., Timbal, A., Sasaki, A., Hauri, E.
1713 & Bacon, C. R. (2009). Stable isotope and petrologic evidence for open-system degassing
1714 during the climactic and pre-climactic eruptions of Mt . Mazama , Crater Lake, Oregon.
1715 *Geochimica et Cosmochimica Acta* **73**, 2978–3012.
- 1716 Marini, L., Moretti, R. & Accornero, M. (2011). Sulfur isotopes in magmatic-hydrothermal

- 1717 systems, melts, and magmas. *Reviews in Mineralogy and Geochemistry* **73**, 423–492.
- 1718 Marini, L., Paiotti, A., Principe, C., Ferrara, G. & Cioni, R. (1994). Isotopic ratio and
1719 concentration of sulfur in the undersaturated alkaline magmas of Vulture Volcano (Italy).
1720 *Bulletin of Volcanology* **56**, 487–492.
- 1721 Martin, H., Smithies, R. H., Rapp, R., Moyen, J. F. & Champion, D. (2005). An overview of
1722 adakite, tonalite–trondhjemite–granodiorite (TTG), and sanukitoid: relationships and
1723 some implications for crustal evolution. *Lithos* **79**, 1–24.
- 1724 Mavrogenes, J. A. & O’Neill, H. S. C. (1999). The relative effects of pressure, temperature and
1725 oxygen fugacity on the solubility of sulfide in mafic magmas. *Geochimica et*
1726 *Cosmochimica Acta* **63**, 1173–1180.
- 1727 McDonough, W. F. & Sun, S. -s. (1995). The composition of the Earth. *Chemical Geology*
1728 **120**, 223–253.
- 1729 Mckenzie, D. & Bickle, M. J. (1988). The volume and composition of melt generated by
1730 extension of the lithosphere. *Journal of Petrology* **29**, 625–679.
- 1731 Médard, E. & Grove, T. L. (2008). The effect of H₂O on the olivine liquidus of basaltic melts:
1732 experiments and thermodynamic models. *Contributions to Mineralogy and Petrology*
1733 **155**, 417–432.
- 1734 Métrich, N. & Mandeville, C. W. (2010). Sulfur in Magmas. *Elements* **6**, 81–86.
- 1735 Miyabuchi, Y., Hanada, D., Niimi, H. & Kobayashi, T. (2013). Stratigraphy, grain-size and
1736 component characteristics of the 2011 Shinmoedake eruption deposits, Kirishima
1737 Volcano, Japan. *Journal of Volcanology and Geothermal Research* **258**, 31–46.
- 1738 Miyabuchi, Y. & Watanabe, K. (1997). Eruption ages of Holocene tephra from Aso volcano,
1739 southwestern Japan, inferred from ¹⁴C ages of buried andisols. *Bulletin of the*
1740 *Volcanological Society of Japan* **42**, 403–408 (in Japanese with English abstract).
- 1741 Miyamoto, T. (1997). The evolution of magmas in Kirishima volcanoes. *Bulletin of the*
1742 *Earthquake Research Institute, University of Tokyo* (in Japanese).
- 1743 Miyashiro, A. (1978). Nature of alkalic volcanic rock series. *Contributions to Mineralogy and*
1744 *Petrology* **66**, 91–104.
- 1745 Miyoshi, M., Fukuoka, T., Sano, T. & Hasenaka, T. (2008a). Subduction influence of
1746 Philippine Sea plate on the mantle beneath Northern Kyushu, SW Japan: an examination
1747 of boron contents in basaltic rocks. *Journal of Volcanology and Geothermal Research*
1748 **171**, 73–87.
- 1749 Miyoshi, M., Hasenaka, T. & Sano, T. (2005). Genetic relationship of the compositionally
1750 diverse magmas from Aso post-caldera volcanism. *Bulletin of the volcanological society*

- 1751 *of Japan* **50**, 269-283 (in Japanese with English abstract).
- 1752 Miyoshi, M., Shibata, T., Yoshikawa, M., Sano, T., Shinmura, T. & Hasenaka, T. (2011).
- 1753 Genetic relationship between post-caldera and caldera-forming magmas from Aso
- 1754 volcano, SW Japan: constraints from Sr isotope and trace element compositions. *Journal*
- 1755 *of Mineralogical and Petrological Sciences* **106**, 114–119.
- 1756 Miyoshi, M., Shimono, M., Hasenaka, T., Sano, T. & Fukuoka, T. (2008b). Determination of
- 1757 boron and other elements in volcanic rocks by prompt gamma-ray analysis: an application
- 1758 to magma genesis in Kyushu island, SW-Japan. *Journal of Radioanalytical and Nuclear*
- 1759 *Chemistry* **278**, 343–347.
- 1760 Miyoshi, M., Shimono, M., Hasenaka, T., Sano, T., Mori, Y. & Fukuoka, T. (2010). Boron
- 1761 systematics of Hisatsu and Kirishima basaltic rocks from Southern Kyushu, Japan.
- 1762 *Geochemical Journal* **44**, 359–369.
- 1763 Moore, L. R., Gazel, E., Tuohy, R., Lloyd, A. S., Esposito, R., Steele-MacInnis, M., Hauri, E.
- 1764 H., Wallace, P. J., Plank, T. & Bodnar, R. J. (2015). Bubbles matter: An assessment of
- 1765 the contribution of vapor bubbles to melt inclusion volatile budgets. *American*
- 1766 *Mineralogist* **100**, 806–823.
- 1767 Moriwaki, H., Machida, H., Hatsumi, Y. & Matsushima, Y. (1986). Phreatomagmatic
- 1768 eruptions affected by postglacial transgression in the northern coastal area of Kagoshima
- 1769 bay, Southern Kyushu, Japan. *Journal of Geography* **95**, 24-43 (in Japanese with English
- 1770 abstract).
- 1771 Moriwaki, H., Matsushima, Y., Iwai, M., Arai, F. & Fujiwara, O. (2002). Holocene
- 1772 geomorphic evolution around the Aira caldera, south Japan. *The Quaternary Research* **41**,
- 1773 253-268 (in Japanese with English abstract).
- 1774 Müller, R. D., Sdrolias, M., Gaina, C. & Roest, W. R. (2008). Age, spreading rates, and
- 1775 spreading asymmetry of the world's ocean crust. *Geochemistry, Geophysics, Geosystems*
- 1776 **9**, 1–19.
- 1777 Muth, M.J., Wallace, P.J. (2021) Slab-derived sulfate generates oxidized basaltic magmas in
- 1778 the southern Cascade arc (California, USA). *Geology* **49**, 1177-1181, doi:
- 1779 <https://doi.org/10.1130/G48759.1>
- 1780 Nagao, T., Hase, Y., Nagamine, S., Kakubuchi, S. & Sakaguchi, K. (1999). Late Miocene to
- 1781 middle Pleistocene Hisatsu volcanic rocks generated from heterogeneous magma sources:
- 1782 evidence form temporal-spatial variation of distribution and chemistry of rocks. *Journal*
- 1783 *of Mineralogy, Petrology and Economic Geology* **94**, 461-481 (in Japanese with English
- 1784 abstract).

- 1785 Nagao, T., Sawai, O., Itaya, T. & Kakubuchi, S. (1992). K-Ar ages and petrological
1786 characteristics of the tholeiitic basalts and high-magnesian andesite from Amakusa-
1787 Shimoshima, Kumamoto Prefecture, SW Japan. *Journal of Mineralogy, Petrology and*
1788 *Economic Geology* **87**, 283-290 (in Japanese with English abstract).
- 1789 Nagaoka, S. & Furuyama, K. (2004). Eruptive history of the Onidake volcano group on Fukue
1790 island, western Japan. *Journal of Geography (Chigaku Zasshi)* **113**, 349-382 (in Japanese
1791 with English abstract).
- 1792 Nagaoka, S. & Okuno, M. (2014). Tephra-stratigraphy of Kuju volcano in southwestern Japan.
1793 *Chikyu monthly* **36**, 281-296 (in Japanese).
- 1794 Nakada, S. (1986). Comparative study of chemistry of rocks from the Kirishima, and Daisen
1795 Volcanic Belts in Kyushu, southwest Japan. *Bulletin of the volcanological society of*
1796 *Japan* **31**, 95-110 (in Japanese with English abstract).
- 1797 Nakada, S. & Kamata, H. (1988). Petrogenetical relationship of basalts and andesites in
1798 southern part of Shimabara Peninsula, Kyushu, Japan. *Bulletin of the Volcanological*
1799 *Society of Japan* **33**, 273–289.
- 1800 Nakada, S. & Kamata, H. (1991). Temporal change in chemistry of magma source under
1801 Central Kyushu, Southwest Japan: progressive contamination of mantle wedge. *Bulletin*
1802 *of Volcanology* **53**, 182–194.
- 1803 Nakada, S., Nagai, M., Kaneko, T., Suzuki, Y. & Maeno, F. (2013). The outline of the 2011
1804 eruption at Shinmoe-dake (Kirishima), Japan. *Earth, Planets and Space* **65**, 475–488.
- 1805 Nakajima, J. & Hasegawa, A. (2007). Subduction of the Philippine Sea plate beneath
1806 southwestern Japan: slab geometry and its relationship to arc magmatism. *Journal of*
1807 *Geophysical Research: Solid Earth* **112**, B08306.
- 1808 Nakamura, M. (1971). Petrology of Kaimon-dake volcano. *Journal of Geological Society of*
1809 *Japan* **77**, 359-364 (in Japanese with English abstract).
- 1810 Nakamura, M. & Kaneshiro, S. (2000). Determination of subducted Philippine Sea Plate in the
1811 Nansei islands deduced from hypocenter data. *Bulletin of the College of Science,*
1812 *University of the Ryukyu* **70**, 73-82 (in Japanese with English abstract).
- 1813 Narvaez, D. F., Rose-Koga, E. F., Samaniego, P., Koga, K. T. & Hidalgo, S. (2018).
1814 Constraining magma sources using primitive olivine-hosted melt inclusions from
1815 Puñalica and Sangay volcanoes (Ecuador). *Contributions to Mineralogy and Petrology*
1816 **173**, 80.
- 1817 Newman, S. & Lowenstern, J. B. (2002). VolatileCalc: a silicate melt–H₂O–CO₂ solution
1818 model written in Visual Basic for excel. *Computers and Geosciences* **28**, 597–604.

- 1819 Ohta, T. & Aoki, K. (1991). Origin of andesitic magma in Yufu-Tsurumi volcano group – a
1820 binary mixing model –. *Journal of Mineralogy, Petrology and Economic Geology* **86**, 1-
1821 15 (in Japanese with English abstract).
- 1822 Ohta, T., Hasenaka, T., Ban, M. & Sasaki, M. (1992). Characteristic geology and petrology of
1823 non-arc type volcanism at Oninomi monogenetic volcano, Yufu-Tsurumi graben. *Bulletin*
1824 *of the volcanological society of Japan* **37**, 119-131 (in Japanese with English abstract).
- 1825 Ohta, T., Hasenakai, T. & Fujimaki, H. (1990). Geology and petrography of Yufu-Tsurumi
1826 volcano group, Oita Prefecture. *Journal of Mineralogy, Petrology and Economic Geology*
1827 **85**, 113-129 (in Japanese with English abstract).
- 1828 Okino, K., Kasuga, S. & Ohara, Y. (1998). A new scenario of the Parece Vela Basin genesis.
1829 *Marine Geophysical Research* **20**, 21–40.
- 1830 Okino, K., Shimakawa, Y. & Nagaoka, S. (1994). Evolution of the Shikoku Basin. *Journal of*
1831 *geomagnetism and geoelectricity* **46**, 463–479.
- 1832 Okuno, M. (2002). Chronology of tephra layers in Southern Kyushu, SW Japan, for the last
1833 30,000 years. *The Quaternary Research* **41**, 225-236 (in Japanese with English abstract).
- 1834 Okuno, M., Fujisawa, Y., Kusanagi-Tajima, M., Ui, T., Nakamura, T. & Kobayashi, T. (1999).
1835 AMS radiocarbon dates of the Ikeshiro and Nonokusa pyroclastic-flow deposits from
1836 Yufu-dake volcano, northeastern Kyushu, Japan. *Summaries of Researchers using AMS*
1837 *at Nagoya University* **10**, 199-205 (in Japanese with English abstract).
- 1838 Okuno, M., Nakamura, T. & Kobayashi, T. (1998). AMS ¹⁴C dating of historic eruptions of the
1839 Kirishima, Sakurajima and Kaimondake volcanoes, Southern Kyushu, Japan.
1840 *Radiocarbon* **40**, 825–832.
- 1841 Palme, H. & O'Neill, H. S. C. (2003). Cosmochemical estimates of mantle composition.
1842 *Treatise on Geochemistry* **2**, 568.
- 1843 Park, J.-O., Hori, T. & Kaneda, Y. (2009). Seismotectonic implications of the Kyushu-Palau
1844 ridge subducting beneath the westernmost Nankai forearc. *Earth, Planets and Space* **61**,
1845 1013–1018.
- 1846 Parkinson, I. J. & Arculus, R. J. (1999). The redox state of subduction zones: insights from arc-
1847 peridotites. *Chemical Geology* **160**, 409–423.
- 1848 Pearce, J. A., Stern, R. J., Bloomer, S. H. & Fryer, P. (2005). Geochemical mapping of the
1849 Mariana arc-basin system: implications for the nature and distribution of subduction
1850 components. *Geochemistry, Geophysics, Geosystems* **6**.
- 1851 Peters, M., Strauss, H., Farquhar, J., Ockert, C., Eickmann, B. & Jost, C. L. (2010). Sulfur
1852 cycling at the mid-Atlantic ridge: a multiple sulfur isotope approach. *Chemical Geology*

- 1853 **269**, 180–196.
- 1854 Pichavant, M., Di Carlo, I., Rotolo, S. G., Scaillet, B., Burgisser, A., Le Gall, N. & Martel, C.
1855 (2013). Generation of CO₂-rich melts during basalt magma ascent and degassing.
1856 *Contributions to Mineralogy and Petrology* **166**, 545–561.
- 1857 Plank, T. (2014). The chemical composition of subducting sediments. *Treatise on*
1858 *Geochemistry: Second Edition* **4**, 607–629.
- 1859 Plank, T. & Langmuir, C. H. (1998). The chemical composition of subducting sediment and its
1860 consequences for the crust and mantle. *Chemical Geology* **145**, 325–394.
- 1861 Portnyagin, M., Almeev, R., Matveev, S. & Holtz, F. (2008). Experimental evidence for rapid
1862 water exchange between melt inclusions in olivine and host magma. *Earth and Planetary*
1863 *Science Letters* **272**, 541–552.
- 1864 Portnyagin, M., Hoernle, K., Plechov, P., Mironov, N. & Khubunaya, S. (2007). Constraints on
1865 mantle melting and composition and nature of slab components in volcanic arcs from
1866 volatiles (H₂O, S, Cl, F) and trace elements in melt inclusions from the Kamchatka Arc.
1867 *Earth and Planetary Science Letters* **255**, 53–69.
- 1868 Rees C. E., Jenkins, W. J. & Monster, J. (1978). The sulphur isotopic composition of ocean
1869 water sulphate. *Geochimica et Cosmochimica Acta* **42**, 377–381.
- 1870 Reimer, P. J., Austin, W. E. N., Bard, E., Bayliss, A., Blackwell, P. G., Bronk Ramsey, C.,
1871 Butzin, M., Cheng, H., Edwards, R. L., Friedrich, M., Grootes, P. M., Guilderson, T. P.,
1872 Hajdas, I., Heaton, T. J., Hogg, A. G., Hughen, K. A., Kromer, B., Manning, S. W.,
1873 Muscheler, R., Palmer, J. G., Pearson, C., van der Plicht, J., Reimer, R. W., Richards, D.
1874 A., Scott, E. M., Southon, J. R., Turney, C. S. M., Wacker, L., Adolphi, F., Büntgen, U.,
1875 Capano, M., Fahrni, S. M., Fogtmann-Schulz, A., Friedrich, R., Köhler, P., Kudsk, S.,
1876 Miyake, F., Olsen, J., Reinig, F., Sakamoto, M., Sookdeo, A. & Talamo, S. (2020). The
1877 IntCal20 northern hemisphere radiocarbon age calibration curve (0–55 cal kBP).
1878 *Radiocarbon* **62**, 725–757.
- 1879 Ridolfi, F., Renzulli, A. & Puerini, M. (2010). Stability and chemical equilibrium of amphibole
1880 in calc-alkaline magmas: an overview, new thermobarometric formulations and
1881 application to subduction-related volcanoes. *Contributions to Mineralogy and Petrology*
1882 **160**, 45–66.
- 1883 Robidoux, P., Frezzotti, M. L., Hauri, E. H. & Aiuppa, A. (2018). Shrinkage bubbles: the C-O-
1884 H-S magmatic fluid system at San Cristóbal volcano. *Journal of Petrology* **59**, 2093–
1885 2122.
- 1886 Roggensack, K. (2001). Unraveling the 1974 eruption of Fuego volcano (Guatemala) with

1887 small crystals and their young melt inclusions. *Geology* **29**, 911–914.

1888 Rose-Koga, E. F., Bouvier, A.-S., Gaetani, G. A., Wallace, P. J., Allison, C. M., Andrys, J. A.,
1889 Angeles de la Torre, C. A., Barth, A., Bodnar, R. J., Bracco Gartner, A. J. J., Butters, D.,
1890 Castillejo, A., Chilson-Parks, B., Choudhary, B. R., Cluzel, N., Cole, M., Cottrell, E.,
1891 Daly, A., Danyushevsky, L. V., DeVitre, C. L., Drignon, M. J., France, L., Gaborieau,
1892 M., Garcia, M. O., Gatti, E., Genske, F. S., Hartley, M. E., Hughes, E. C., Iveson, A. A.,
1893 Johnson, E. R., Jones, M., Kagoshima, T., Katzir, Y., Kawaguchi, M., Kawamoto, T.,
1894 Kelley, K. A., Koornneef, J. M., Kurz, M. D., Laubier, M., Layne, G. D., Lerner, A., Lin,
1895 K.-Y., Liu, P.-P., Lorenzo-Merino, A., Luciani, N., Magalhães, N., Marschall, H. R.,
1896 Michael, P. J., Monteleone, B. D., Moore, L. R., Moussallam, Y., Muth, M., Myers, M.
1897 L., Narváez, D. F., Navon, O., Newcombe, M. E., Nichols, A. R. L., Nielsen, R. L.,
1898 Pamukcu, A., Plank, T., Rasmussen, D. J., Roberge, J., Schiavi, F., Schwartz, D.,
1899 Shimizu, K., Shimizu, N., Thomas, J. B., Thompson, G. T., Tucker, J. M., Ustunisik, G.,
1900 Waelkens, C., Zhang, Y. & Zhou, T. (2021). Silicate melt inclusions in the new
1901 millennium: a review of recommended practices for preparation, analysis, and data
1902 presentation. *Chemical Geology* **570**, 120145.

1903 Rose-Koga, E. F., Koga, K. T., Devidal, J.-L., Shimizu, N., Le Voyer, M., Dalou, C. & Döbeli,
1904 M. (2020). In-situ measurements of magmatic volatile elements, F, S, and Cl, by electron
1905 microprobe, secondary ion mass spectrometry, and heavy ion elastic recoil detection
1906 analysis. *American Mineralogist* **105**, 616–626.

1907 Rose-Koga, E. F., Koga, K. T., Hamada, M., H elouis, T., Whitehouse, M. J. & Shimizu, N.
1908 (2014). Volatile (F and Cl) concentrations in Iwate olivine-hosted melt inclusions
1909 indicating low-temperature subduction. *Earth, Planets and Space* **66**, 81.

1910 Rose-Koga, E. F., Koga, K. T., Moreira, M., Vlastelic, I., Jackson, M. G., Whitehouse, M. J.,
1911 Shimizu, N. & Habib, N. (2017). Geochemical systematics of Pb isotopes, fluorine, and
1912 sulfur in melt inclusions from S ao Miguel, Azores. *Chemical Geology* **458**, 22–37.

1913 Rose-Koga, E. F., Koga, K. T., Schiano, P., Le Voyer, M., Shimizu, N., Whitehouse, M. J. &
1914 Clocchiatti, R. (2012). Mantle source heterogeneity for South Tyrrhenian magmas
1915 revealed by Pb isotopes and halogen contents of olivine-hosted melt inclusions. *Chemical*
1916 *Geology* **334**, 266–279.

1917 Sadofsky, S. J., Portnyagin, M., Hoernle, K. & van den Bogaard, P. (2008). Subduction cycling
1918 of volatiles and trace elements through the Central American volcanic arc: evidence from
1919 melt inclusions. *Contributions to Mineralogy and Petrology* **155**, 433–456.

1920 Saito, E. (2017). GIS data of the depth of the oceanic plate surfaces beneath the Japanese

1921 archipelago. *GSJ Open-File Report, no. 647, Geological Survey of Japan, AIST* (in
1922 Japanese).

1923 Sakai, H., Marais, D. J. D., Ueda, A. & Moore, J. G. (1984). Concentrations and isotope ratios
1924 of carbon, nitrogen and sulfur in ocean-floor basalts. *Geochimica et Cosmochimica Acta*
1925 **48**, 2433–2441.

1926 Sakuyama, T., Nakai, S., Yoshikawa, M., Shibata, T. & Ozawa, K. (2014). Progressive
1927 interaction between dry and wet mantle during high-temperature diapiric upwelling:
1928 constraints from cenozoic Kita-Matsuura intraplate basalt province, Northwestern
1929 Kyushu, Japan. *Journal of Petrology* **55**, 1083–1128.

1930 Sakuyama, T., Ozawa, K., Sumino, H. & Nagao, K. (2009). Progressive melt extraction from
1931 upwelling mantle constrained by the Kita-Matsuura basalts in NW Kyushu, SW Japan.
1932 *Journal of Petrology* **50**, 725–779.

1933 Salters, V. J. M. & Stracke, A. (2004). Composition of the depleted mantle. *Geochemistry,*
1934 *Geophysics, Geosystems* **5**, Q05B07.

1935 Sasaki, A. & Ishihara, S. (1979). Sulfur isotopic composition of the magnetite-series and
1936 ilmenite-series granitoids in Japan. *Contributions to Mineralogy and Petrology* **68**, 107–
1937 115.

1938 Savov, I. P., Ryan, J. G., D’Antonio, M., Kelley, K. & Mattie, P. (2005). Geochemistry of
1939 serpentinized peridotites from the Mariana forearc Conical Seamount, ODP leg 125:
1940 implications for the elemental recycling at subduction zones. *Geochemistry, Geophysics,*
1941 *Geosystems* **6**, Q04J15.

1942 Sawamura, K. & Matsui, K. (1957). *Explanatory text of the geological map of Japan,*
1943 *“Kirishimayama”, Scale 1:50,000. Geological Survey of Japan.*

1944 Schiano, P., Monzier, M., Eissen, J.-P., Martin, H. & Koga, K. T. (2010). Simple mixing as the
1945 major control of the evolution of volcanic suites in the Ecuadorian Andes. *Contributions*
1946 *to Mineralogy and Petrology* **160**, 297–312.

1947 Seno, T. & Maruyama, S. (1984). Paleogeographic reconstruction and origin of the Philippine
1948 Sea. *Tectonophysics* **102**, 53–84.

1949 Shaw, A. M., Behn, M. D., Humphris, S. E., Sohn, R. A. & Gregg, P. M. (2010). Deep pooling
1950 of low degree melts and volatile fluxes at the 85°E segment of the Gakkel Ridge:
1951 evidence from olivine-hosted melt inclusions and glasses. *Earth and Planetary Science*
1952 *Letters* **289**, 311–322.

1953 Shibata, K., Mizuno, A., Yuasa, M., Uchiumi, S. & Nakagawa, T. (1977). Further K-Ar dating
1954 of tonalite dredged from the Komahashi-daini seamount. *Bulletin of the Geological*

- 1955 *Survey of Japan* **28**, 1–4.
- 1956 Shibata, T., Suzuki, J., Yoshikawa, M., Kobayashi, T., Miki, D. & Takemura, K. (2013).
- 1957 Geochemical and Sr-Nd-Pb isotopic constraints on the origin and magmatic evolution of
- 1958 Quaternary lavas of Sakurajima volcano, Southern Kyushu Island, Japan. *Bulletin of the*
- 1959 *volcanological society of Japan* **58**, 43–58.
- 1960 Shibata, T., Yoshikawa, M., Itoh, J., Ujike, O., Miyoshi, M. & Takemura, K. (2014). Along-arc
- 1961 geochemical variations in Quaternary magmas of Northern Kyushu Island, Japan.
- 1962 *Geological Society Special Publication* **385**, 15–29.
- 1963 Shimizu, K., Ito, M., Chang, Q., Miyazaki, T., Ueki, K., Toyama, C., Senda, R., Vaglarov, B.
- 1964 S., Ishikawa, T. & Kimura, J.-I. (2019a). Identifying volatile mantle trend with the water–
- 1965 fluorine–cerium systematics of basaltic glass. *Chemical Geology* **522**, 283–294.
- 1966 Shimizu, K., Saal, A. E., Hauri, E. H., Perfit, M. R. & Hékinian, R. (2019b). Evaluating the
- 1967 roles of melt-rock interaction and partial degassing on the CO₂/Ba ratios of MORB:
- 1968 implications for the CO₂ budget in the Earth’s depleted upper mantle. *Geochimica et*
- 1969 *Cosmochimica Acta* **260**, 29–48.
- 1970 Shimizu, K., Ushikubo, T., Hamada, M., Itoh, S., Higashi, Y., Takahashi, E. & Ito, M. (2017).
- 1971 H₂O, CO₂, F, S, Cl, and P₂O₅ analyses of silicate glasses using SIMS: report of volatile
- 1972 standard glasses. *Geochemical Journal* **51**, 299–313.
- 1973 Shimizu, K., Ushikubo, T., Murai, T., Matsu’ura, F. & Ueno, Y. (2019c). In situ analyses of
- 1974 hydrogen and sulfur isotope ratios in basaltic glass using SIMS. *Geochemical Journal* **53**,
- 1975 195–207.
- 1976 Shinjoe, H. (1997). Origin of the granodiorite in the forearc region of southwest Japan: melting
- 1977 of the Shimanto accretionary prism. *Chemical Geology* **134**, 237–255.
- 1978 Shinno, I. (1966). Petrology of the Kirishima volcano. *Journal of Mineralogy, Petrology and*
- 1979 *Economic Geology* **56**, 56-74 (in Japanese with English abstract).
- 1980 Shitaoka, Y., Miyoshi, M., Yamamoto, J., Shibata, T., Nagatomo, T. & Takemura, K. (2014).
- 1981 Thermoluminescence age of quartz xenocrysts in basaltic lava from oninomi monogenetic
- 1982 Volcano, Northern Kyushu, Japan. *Geochronometria* **41**, 30–35.
- 1983 Shu, Y., Nielsen, S. G., Zeng, Z., Shinjo, R., Blusztajn, J., Wang, X. & Chen, S. (2017).
- 1984 Tracing subducted sediment inputs to the Ryukyu arc-Okinawa Trough system: evidence
- 1985 from thallium isotopes. *Geochimica et Cosmochimica Acta* **217**, 462–491.
- 1986 Slater, L., Jull, M., McKenzie, D. & Gronvöld, K. (1998). Deglaciation effects on mantle
- 1987 melting under Iceland: results from the northern volcanic zone. *Earth and Planetary*
- 1988 *Science Letters* **164**, 151–164.

- 1989 Sorbadere, F., Schiano, P. & Métrich, N. (2013). Constraints on the origin of nepheline-
1990 normative primitive magmas in Island arcs inferred from olivine-hosted melt inclusion
1991 compositions. *Journal of Petrology* **54**, 215–233.
- 1992 Sorbadere, F., Schiano, P., Métrich, N. & Garaebiti, E. (2011). Insights into the origin of
1993 primitive silica-undersaturated arc magmas of Aoba volcano (Vanuatu arc). *Contributions*
1994 *to Mineralogy and Petrology* **162**, 995–1009.
- 1995 Spilliaert, N., Métrich, N. & Allard, P. (2006). S-Cl-F degassing pattern of water-rich alkali
1996 basalt: modelling and relationship with eruption styles on Mount Etna volcano. *Earth and*
1997 *Planetary Science Letters* **248**, 772–786.
- 1998 Staudigel, H. (2003). Hydrothermal alteration processes in the oceanic crust. *Treatise on*
1999 *Geochemistry* **3**, 511–535.
- 2000 Stolper, E. & Newman, S. (1994). The role of water in the petrogenesis of Mariana trough
2001 magmas. *Earth and Planetary Science Letters* **121**, 293–325.
- 2002 Straub, S. M. & Layne, G. D. (2003). The systematics of chlorine, fluorine, and water in Izu
2003 arc front volcanic rocks: implications for volatile recycling in subduction zones.
2004 *Geochimica et Cosmochimica Acta* **67**, 4179–4203.
- 2005 Stuiver, M. & Reimer, P. J. (1993). Extended ¹⁴C data base and revised CALIB 3.0 ¹⁴C age
2006 calibration program. *Radiocarbon* **35**, 215–230.
- 2007 Stuiver, M., Reimer, P. J. & W, R. R. (2020). *CALIB 8.2*. <http://calib.org> (accessed 26
2008 November 2020).
- 2009 Sugawara, T. (2000). Empirical relationships between temperature, pressure, and MgO content
2010 in olivine and pyroxene saturated liquid. *Journal of Geophysical Research: Solid Earth*
2011 **105**, 8457–8472.
- 2012 Sugimoto, T., Ishibashi, H., Wakamatsu, S. & Yanagi, T. (2005). Petrologic evolution of Pre-
2013 Unzen and Unzen magma chambers beneath the Shimabara Peninsula, Kyushu, Japan:
2014 Evidence from petrography and bulk rock chemistry. *Geochemical Journal* **39**, 241–256.
- 2015 Sugimoto, T., Shibata, T., Yoshikawa, M. & Takemura, K. (2006). Sr-Nd-Pb isotopic and
2016 major and trace element compositions of the Yufu-Tsurumi volcanic rocks: implications
2017 for the magma genesis of the Yufu-Tsurumi volcanoes, northeast Kyushu, Japan. *Journal*
2018 *of Mineralogical and Petrological Sciences* **101**, 270–275.
- 2019 Sumino, H., Burgess, R., Mizukami, T., Wallis, S. R., Holland, G. & Ballentine, C. J. (2010).
2020 Seawater-derived noble gases and halogens preserved in exhumed mantle wedge
2021 peridotite. *Earth and Planetary Science Letters*. Elsevier B.V. **294**, 163–172.
- 2022 Sun, S. -s. & McDonough, W. F. (1989). Chemical and isotopic systematics of oceanic basalts:

- 2023 implications for mantle composition and processes. *Geological Society, London, Special*
2024 *Publications* **42**, 313–345.
- 2025 Symonds, R. B., Rose, W. I., Bluth, G. J. S. & Gerlach, T. M. (1994). Volcanic-gas studies:
2026 methods, results, and applications. *Volatiles in Magmas*. Reviews in Mineralogy and
2027 geochemistry **30**, 1–66.
- 2028 Syracuse, E. M., van Keken, P. E. & Abers, G. A. (2010). The global range of subduction zone
2029 thermal models. *Physics of the Earth and Planetary Interiors* **183**, 73–90.
- 2030 Tajima, Y., Hayashi, S., Yasuda, A. & Itoh, H. (2013). Tephrostratigraphy and eruptive history
2031 of Shinmoedake volcano of the Kirishima volcanoes, Kyushu, Japan. *The Quaternary*
2032 *Research* **52**, 151-171 (in Japanese with English abstract).
- 2033 Tajima, Y., Matsuo, Y., Shoji, T. & Kobayashi, T. (2014). Eruptive history of Ebinokogen
2034 volcanic area of Kirishima volcanoes for the past 15,000 years in Kyushu, Japan. *Bulletin*
2035 *of the volcanological society of Japan* **59**, 55-75 (in Japanese with English abstract).
- 2036 Taracsák, Z., Neave, D.A., Beaudry, P. , Gunnarsson-Robin, J., Burgess, R., Edmonds, M.,
2037 Halldórsson, S.A., Longpré, M-A., Ono, S., Ranta, E., Stefánsson, A., Turchyn, A.V.,
2038 EIMF, Hartley, M.E. (2021) Instrumental mass fractionation during sulfur isotope
2039 analysis by secondary ion mass spectrometry in natural and synthetic glasses, *Chemical*
2040 *Geology*, **578**, 120318, <https://doi.org/10.1016/j.chemgeo.2021.120318>.
- 2041 Tatsumi, Y. (1989). Migration of fluid phases and genesis of basalt magmas in subduction
2042 zones. *Journal of Geophysical Research*, **94**, 4697-4707.
- 2043 Tatsumi, Y. & Eggins, S. (1995). *Subduction zone magmatism*. Boston, Blackwell Science.
- 2044 Toplis, M. J. (2005). The thermodynamics of iron and magnesium partitioning between olivine
2045 and liquid: criteria for assessing and predicting equilibrium in natural and experimental
2046 systems. *Contributions to Mineralogy and Petrology* **149**, 22–39.
- 2047 Tsutsui, M., Okuno, M. & Kobayashi, T. (2007). Eruptive history of Ohachi volcano,
2048 Kirishima volcano group, Southern Kyushu, Japan. *Bulletin of the Volcanological Society*
2049 *of Japan* **52**, 1-21 (in Japanese with English abstract).
- 2050 Tucker, J. M., Hauri, E. H., Pietruszka, A. J., Garcia, M. O., Marske, J. P. & Trusdell, F. A.
2051 (2019). A high carbon content of the Hawaiian mantle from olivine-hosted melt
2052 inclusions. *Geochimica et Cosmochimica Acta* **254**, 156–172.
- 2053 Ueda, A. & Sakai, H. (1984). Sulfur isotope study of Quaternary volcanic rocks from the
2054 Japanese islands arc. *Geochimica et Cosmochimica Acta* **48**, 1837–1848.
- 2055 Uto, K., Hoang, N. & Matsui, K. (2004). Cenozoic lithospheric extension induced magmatism
2056 in southwest Japan. *Tectonophysics* **393**, 281–299.

2057 Uto, K. & Uchiumi, S. (1997). K-Ar ages of Maruyama lava domes in Hiwaki town,
2058 Kagoshima Prefecture, southwest Japan: a Quaternary volcano constituting a second
2059 volcanic row of Ryukyu arc. *Bulletin of the Volcanological Society of Japan* **42**, 299-302
2060 (in Japanese).

2061 Van den Bleeken, G. & Koga, K. T. (2015). Experimentally determined distribution of fluorine
2062 and chlorine upon hydrous slab melting, and implications for F-Cl cycling through
2063 subduction zones. *Geochimica et Cosmochimica Acta* **171**, 353–373.

2064 Vigouroux, N., Wallace, P. J. & Kent, A. J. R. (2008). Volatiles in high-K magmas from the
2065 western trans-Mexican volcanic belt: evidence for fluid fluxing and extreme enrichment
2066 of the mantle wedge by subduction processes. *Journal of Petrology* **49**, 1589–1618.

2067 Wallace, P. J. (2005). Volatiles in subduction zone magmas: concentrations and fluxes based
2068 on melt inclusion and volcanic gas data. *Journal of Volcanology and Geothermal
2069 Research* **140**, 217–240.

2070 Wallace, P. J., Kamenetsky, V. S. & Cervantes, P. (2015). Melt inclusion CO₂ contents,
2071 pressures of olivine crystallization, and the problem of shrinkage bubbles. *American
2072 Mineralogist* **100**, 787–794.

2073 Walowski, K. J., Wallace, P. J., Clyne, M. A., Rasmussen, D. J. & Weis, D. (2016). Slab
2074 melting and magma formation beneath the southern Cascade arc. *Earth and Planetary
2075 Science Letters* **446**, 100–112.

2076 Walter, M. J. (1998). Melting of garnet peridotite and the origin of Komatiite and depleted
2077 lithosphere. *Journal of Petrology* **39**, 29–60.

2078 Walters, J. B., Cruz-Uribe A.M. and Marschall H. R. (2019) Isotopic Compositions of Sulfides
2079 in Exhumed High-Pressure Terranes: Implications for Sulfur Cycling in Subduction Zones,
2080 *Geochem. Geophys. Geosyst.*, doi:10.1029/2019GC008374.

2081 Wanless, V. D., Behn, M. D., Shaw, A. M. & Plank, T. (2014). Variations in melting dynamics
2082 and mantle compositions along the eastern volcanic zone of the Gakkel ridge: insights
2083 from olivine-hosted melt inclusions. *Contributions to Mineralogy and Petrology* **167**,
2084 1005.

2085 Wanless, V. D. & Shaw, A. M. (2012). Lower crustal crystallization and melt evolution at mid-
2086 ocean ridges. *Nature Geoscience* **5**, 651–655.

2087 Waters, L. E. & Lange, R. A. (2015). An updated calibration of the plagioclase-liquid
2088 hygrometer-thermometer applicable to basalts through rhyolites. *American Mineralogist*
2089 **100**, 2172–2184.

2090 Webster, J. D., Mandeville, C. W., Goldoff, B., Coombs, M. L. & Tappen, C. (2010).

2091 Augustine Volcano—The influence of volatile components in magmas erupted A.D. 2006
2092 to 2,100 years before present. In: Power, J. A., Coombs, M. L. & Freymueller, J. T. (eds)
2093 *The 2006 eruption of Augustine Volcano, Alaska: U.S. Geological Survey Professional*
2094 *Paper 1769.*

2095 Workman, R. K. & Hart, S. R. (2005). Major and trace element composition of the depleted
2096 MORB mantle (DMM). *Earth and Planetary Science Letters* **231**, 53–72.

2097 Wysoczanski, R. J., Wright, I. C., Gamble, J. A., Hauri, E. H., Luhr, J. F., Eggins, S. M. &
2098 Handler, M. R. (2006). Volatile contents of Kermadec arc-Havre trough pillow glasses:
2099 fingerprinting slab-derived aqueous fluids in the mantle sources of arc and back-arc lavas.
2100 *Journal of Volcanology and Geothermal Research* **152**, 51–73.

2101 Yamasaki, S., Hoshizumi, H. & Matsumoto, A. (2016). Growth history of western to central
2102 part of Kuju volcanic group, Kyushu, Japan. *Bulletin of the Volcanological Society of*
2103 *Japan* **61**, 519-531 (in Japanese with English abstract).

2104 Zellmer, G. F., Iizuka, Y., Miyoshi, M., Tamura, Y. & Tatsumi, Y. (2012). Lower crustal H₂O
2105 controls on the formation of adakitic melts. *Geology* **40**, 487–490.

2106 Zellmer, G. F., Sakamoto, N., Iizuka, Y., Miyoshi, M., Tamura, Y., Hsieh, H.-H. & Yurimoto,
2107 H. (2014). Crystal uptake into aphyric arc melts: insights from two-pyroxene pseudo-
2108 decompression path, plagioclase hygrometry, and measurement of hydrogen in olivines
2109 from mafic volcanics of SW Japan. *Geological Society, London, Special Publications*
2110 **385**, 161–184.

2111 Zhao, D. & Tian, Y. (2013). Changbai intraplate volcanism and deep earthquakes in east asia: a
2112 possible link? *Geophysical Journal International* **195**, 706–724.

2113 Zhao, D., Tian, Y., Lei, J., Liu, L. & Zheng, S. (2009). Seismic image and origin of the
2114 Changbai intraplate volcano in East Asia: role of big mantle wedge above the stagnant
2115 Pacific slab. *Physics of the Earth and Planetary Interiors* **173**, 197–206.

2116 Zhao, D., Yanada, T., Hasegawa, A., Umino, N. & Wei, W. (2012). Imaging the subducting
2117 slabs and mantle upwelling under the Japan Islands. *Geophysical Journal International*
2118 **190**, 816–828.

2119