# The Role of Magma Storage Conditions and Excess Fluids in the Effusive to Explosive Eruption Styles

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#### Abstract

We investigate the role of pre-eruptive magma conditions in controlling eruption styles at basaltic andesite volcanoes, and their relationship to monitoring data. We focus on a cycle of explosive (1990, sub-Plinian) => effusive (2007 dome) => explosive (2014, sub-Plinian) eruptions at Kelud volcano, Indonesia. We conducted petrological analyses of eruption products and phase equilibria experiments using a pumice from the 1990 eruption across a wide range of conditions (temperature, pressure, oxygen fugacity, and volatile contents). We are able to reproduce the main mineral assemblage of all three magmas (plagioclase + pyroxenes + amphibole + olivine + magnetite) at T = 975 ± 39°C, P = 175 ± 25 MPa, NNO, and 4-6 wt. % water in the melt, which is also consistent with the phenocryst contents for explosive eruptions (30-50 wt. %). The high phenocryst content of the 2007 dome (~ 70 wt. %) implies a lower volatile content (~1 wt. % water in the melt), which likely resulted from slow magma ascent towards the surface alongside progressive equilibration. Mass balance calculations on the sulfur budget of the 1990 and 2014 magmas, show that they contained an excess fluid phase that was stored with the magma, and we propose that this led to their more explosive eruption style compared to the 2007 dome. Our hypothesis of accumulation of excess fluids could perhaps be recorded as changes in gravity data over time, and may open doors to better anticipate eruption styles.

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18	Key Points:
19 20	• We use phase equilibria experiments to evaluate the role of magmatic storage conditions on eruption style of Kelud volcano, Indonesia.
21 22	• Explosive eruptions retain an excess fluid phase, whereas the lava dome lose this phase gradually; both share similar storage conditions.
23 24	• The accumulation of fluids could lead to decreasing magma density over time, which could be monitored using gravity measurements.
25 26	

### 27 Abstract

We investigate the role of pre-eruptive magma conditions in controlling eruption styles at 28 29 basaltic andesite volcanoes, and their relationship to monitoring data. We focus on a cycle of 30 explosive (1990, sub-Plinian)  $\rightarrow$  effusive (2007 dome)  $\rightarrow$  explosive (2014, sub-Plinian) eruptions 31 at Kelud volcano, Indonesia. We conducted petrological analyses of eruption products and phase 32 equilibria experiments using a pumice from the 1990 eruption across a wide range of conditions 33 (temperature, pressure, oxygen fugacity, and volatile contents). We are able to reproduce the 34 main mineral assemblage of all three magmas (plagioclase + pyroxenes + amphibole + olivine + 35 magnetite) at T = 975  $\pm$  39°C, P = 175  $\pm$  25 MPa,  $fO_2$  = NNO, and 4-6 wt. % water in the melt, 36 which is also consistent with the phenocryst contents for explosive eruptions (30-50 wt. %). The 37 high phenocryst content of the 2007 dome ( $\sim$  70 wt. %) implies a lower volatile content ( $\sim$ 1 wt. 38 % water in the melt), which likely resulted from slow magma ascent towards the surface 39 alongside progressive equilibration. Mass balance calculations on the sulfur budget of the 1990 40 and 2014 magmas, show that they contained an excess fluid phase that was stored with the 41 magma, and we propose that this led to their more explosive eruption style compared to the 2007 42 dome. Our hypothesis of accumulation of excess fluids could perhaps be recorded as changes in 43 gravity data over time, and may open doors to better anticipate eruption styles.

44

#### 45 Plain Language Summary

46 Many volcanoes can erupt explosively or grow lava domes, and it is still difficult to know how 47 and when the volcano may erupt in the future. We use experiments to investigate whether the 48 temperature, pressure, and dissolved gas content of eruptible molten rock underneath a volcano 49 can control its eruption style, and then relate them to monitoring data signals typically used to 50 anticipate eruptions. We also study rocks from the 1990-2014 eruptions of Kelud volcano in 51 Indonesia, which is notorious for its explosive and short eruptions, but can also shifts to lava 52 dome growth. We find that the depth, temperature and gases dissolved in the melt are essentially 53 the same for the 2007 lava dome, and the 1990 and 2014 explosive eruptions. However, 54 explosive eruptions also accumulate a separate surplus gas phase before eruption (e.g. "pressure 55 cooker" effect), whereas before lava dome growth this gas phase is lost over time to the surface.

Thus it may be possible to anticipate eruption styles by tracking the gravity and density changesbelow the volcano.

58

### 59 **1 Introduction**

60 Volcanoes can display a range of eruption styles, spanning from small phreatic events, to 61 dome growth, or explosive events with tall eruptive columns. Such changes can occur between 62 consecutive eruptions of the same volcano, and even within a single eruption. Some well-known examples of volcanoes that display large variations in eruptive styles between eruptions, include 63 64 Merapi (Indonesia e.g. Surono et al., 2012), Soufrière Hills (Montserrat; e.g. Young et al., 1998), 65 and Mt. St Helens (USA; e.g. Pallister et al., 1992). Examples of how a single volcanic eruption 66 can also rapidly transition from effusive to explosive and vice versa were observed in the 1991 67 Pinatubo eruption (Bernard et al., 1996; Pallister et al., 1996), the 2009 eruption at Redoubt (Bull 68 & Buurman, 2013), or the 2011 eruption at Puyehue-Cordón Caulle (Elissondo et al., 2016). The 69 changes in eruptive style are typically abrupt and difficult to anticipate, which makes the 70 mitigation of associated hazards incredibly challenging.

71 Changes in eruptive styles are largely controlled by the physical and chemical properties 72 of the magmas at their storage zones, and how these change as the magma ascends towards the 73 surface and goes through the brittle-ductile transition (e.g. Cashman et al., 2013; Dingwell et al., 74 1993). Some parameters include the pre-eruptive magmatic storage pressure, temperature, 75 crystallinity, melt composition and magmatic volatile contents. The pre-eruptive volatile content 76 of the melt is especially important in determining key parameters, namely the viscosity, 77 buoyancy, and magma ascent rate (Cassidy et al., 2018; Giordano et al., 2008; Gonnermann & 78 Manga, 2013). In addition, the presence of an exsolved volatile phase at the pre-eruptive system 79 due to second boiling (Sisson & Bacon, 1999) or volatile flushing (Caricchi et al., 2018) can also 80 have a large impact on eruption dynamics. Moreover, such exsolved fluid phase has been 81 documented and quantified for many explosive eruptions (Scaillet et al., 1998). 82 Nothwithstanding our understanding of the parameters and processes leading to explosive 83 eruptions it is still very challenging to anticipate whether a given volcano will erupt explosively

84 or effusively (Cassidy et al., 2018; Shinohara, 2008).

87 In this study, we investigate the pre-eruptive conditions and processes that may have led 88 to a sequence of explosive-effusive-explosive eruption of Kelud volcano (Indonesia), as 89 exemplified by the 1990  $\rightarrow$  2007  $\rightarrow$  2014 eruptions. We also report new petrological observations 90 that we combine with various mineral-melt geothermobarometers, and new phase equilibria 91 experiments to constrain the pre-eruptive storage conditions of the three eruptions. The 92 combined petrological and experimental results suggest that the magma that fed the 1990, 2007, 93 and 2014 events were essentially stored at similar temperatures, pressures, and  $fO_2$  range. We 94 propose that the higher explosivity of the 1990 and 2014 eruptions was instead driven by the 95 presence of exsolved volatile phase that was lacking in the 2007 dome eruption, and this led to 96 slower ascent, enhanced degassing, and subsequent re-equilibration of the magma towards the 97 surface that ultimately produced a dome.

98

### 99 2 Geological Setting

### 100 2.1 Kelud volcano

101 Kelud (Kelut) volcano is an active basaltic-andesite semi-plugged composite strato-cone 102 complex situated in East Java, Indonesia (Whelley et al., 2015; Wirakusumah, 1991). The 103 volcano hosts a persistent crater lake fed by meteoric water (neutral pH) during repose, and 104 explosive eruptions typically generate syn-eruptive and post-eruptive cold lahars (Thouret et al., 105 1998). Kelud typically generates short-lived explosive eruptions with run-up times of mere hours 106 (Global Volcanism Program, 2013; Lesage & Surono, 1995; Maeno et al., 2019). Kelud has 107 erupted at least 32 times in the last 100 years (Global Volcanism Program, 2013), but here we 108 focus on the last three eruptions in 1990, 2007, and 2014.

The 1990 explosive eruption began after ~3 months of unrest followed by quiescence for ~3 weeks (Bourdier et al., 1997). Escalation towards eruption occurred in hours, giving communities only one hour to evacuate their homes (Global Volcanism Program, 1990a). The explosive eruption of 2014 largely followed a similar pattern in terms of duration of heightened unrest and steep increase in seismicity hours to days before the eruption (Hidayati et al., 2018). These 1990 and 2014 events generated eruptive columns as high as 17 km (Bourdier et al., 1997; 115 Kristiansen et al., 2015; Nakashima et al., 2016) with a VEI of 4 and Dense Rock Equivalent 116 (DRE) volumes of  $6.3 \times 10^7 \text{ m}^3$  (1990) and  $2.1 \times 10^8 \text{ m}^3$  (2014), respectively (Maeno et al., 117 2019). Moreover, the eruptions were short-lived and volcanic activity and unrest returned to 118 background levels within days to weeks of the main eruptive phase (Global Volcanism Program, 119 2014; Pardyanto, 1990).

120

121 The lava dome extrusion of November 2007 and was preceded by unrest that started as 122 early as January 2007, according to Lubis (2014) who detected up to 11 cm inflation around the 123 summit region from remote sensing data. From July to August 2007 unrest involved intense 124 degassing on the lake floor and increase in seismicity (Hidayati et al., 2018) and lake 125 temperature rising up (from  $\sim 30^{\circ}$ C to  $\sim 75^{\circ}$ C), as well as heightened CO<sub>2</sub> emissions from the lake 126 (Caudron et al., 2012; Global Volcanism Program, 2007; Hidayati et al., 2009; Kunrat, 2009). 127 Continuous seismic unrest started around September 2007, involving multiple swarms and low 128 frequency earthquakes (Hidayati et al., 2009). The seismic unrest was further accompanied by inflation from 9<sup>th</sup> October until 3<sup>rd</sup> November, when the signal changed to deflation and gave 129 130 way to slow dome extrusion (Hidayati et al., 2018). The final dome was 200 m high and 400 m wide, with a volume of  $3.5 \times 10^7 \text{ m}^3$  which is about half the volume of the 1990 eruption, but 131 132 about one order of magnitude smaller than the 2014 event (Global Volcanism Program, 2008; 133 Maeno et al., 2019).

134

### 135 **3 Materials and Methods**

We performed petrological analyses of two 2007 dome fragments previously studied in Humaida (2013), and two pumices sampled following the 2014 eruption (sampled by H. Humaida), and also include the data from the six main-stage pumices from the 1990 eruption (already studied in Utami et al., in revision). We chose one basaltic andesite pumice from the 1990 eruption (KELUD1990-PL1 or PL1 from Utami et al. (in revision)) as the starting material for experiments, because it was the most mafic, and had the same mineral assemblage of the other samples except for the absence of olivine.

144 3.1 Selection of experimental conditions

145 We experimentally investigated a range of variables that includes previous estimates

146 from mineral-melt geothermobarometers (Utami et al., in revision). We varied temperature from

147 950 to 1060°C, pressure from 100 to 400 MPa,  $fO_2$  at the Ni-NiO buffer (NNO) and up to two

log units above (NNO + 2), and varied a volatile composition in the capsule from pure water to

- 149 pure  $CO_2$  (See below for more details).
- 150

### 151 3.2 Preparation of experimental charges

152 A starting glass was prepared by finely crushing the pumice using a ceramic mortar and 153 pestle, and homogenized by fusing the powder at 1400°C and at atmospheric pressure for around 154 6 hours in a standard Thermconcept furnace at 1750°C. EPMA analyses of the fused glass 155 confirmed that glass it was homogenous, and of the same composition as the whole rock (Table 156 1). This glass was crushed again to a fine dry powder with an agate mortar and pestle, and was 157 used as starting material for all experiments. About 0.03g of dry glass was loaded in Au capsules 158 (of size 10-15 mm x 2.5 x 0.2 mm wall thickness) for the 950°C and 1000°C experiments, 159 whereas Au-Pd capsules were used for the 1060°C experiments.

160

161 We varied the fluid composition (referred to as  $XH_2O_{in} = initial H_2O / [H_2O + CO_2]$  in 162 moles) from 0 to 1 by adding various amounts of deionized water and CO<sub>2</sub> (in the form of silver 163 oxalate) to the dry glass powder according to the methodology of Scaillet et al. (1995). All the 164 successful runs were fluid saturated. The capsules were arc-welded and weighed before being 165 stored in a 60°C oven to homogenize the fluid content for 6-12 hours. After each experiments, 166 the capsule was re-weighed again to check for fluid loss during storage and subsequent 167 experiments. To determine whether the charges were properly sealed, their weight had to be 168 within 0.0005 g of the original value, which was the precision of the analytical balance. We 169 observed that capsules from successfully quenched experiments were shrunk and flattened but 170 had the same weight as before the experiment. Upon opening the charges ejected gas with an 171 audible pop. In contrast, failed charges appeared bloated, and commonly showed a difference in 172 mass between before and after the experiments.

174 **Table 1.** *Representative compositions for the starting glass, whole rock, clinopyroxene-hosted* 

175 *melt inclusions, and interstitial glass from the 1990, 2007, and 2014 eruptions.* 

	Whole rock	Whole rock	Whole rock	Starting dry glass	Measured matrix glass <sup>a</sup>	Calculated matrix glass	Measured matrix glass	Cpx glass inclusions <sup>a</sup>	Cpx glass inclusions
Year	1990	2007	2014	1990	1990	2007	2014	1990	2014
n	1	1	1	10	27	1	5	15	5
Normalize	ed wt. %								
SiO <sub>2</sub>	54.3	54.9	55.3	55.0 (0.3)	68.4 (1.4)	69.3	67.1 (0.2)	69.1 (1.5)	68.5 (0.8)
TiO <sub>2</sub>	0.7	0.6	0.6	0.6 (0.1)	0.53 (0.07)	0.9	0.6 (0.1)	0.5 (0.1)	0.7 (0.1)
$AI_2O_3$	19.0	18.6	18.5	19.0 (0.1)	15.4 (0.5)	14.0	15.5 (0.2)	16.0 (0.5)	15.7 (0.6)
FeO*	9.3	9.2	8.9	8.9 (0.5)	4.1 (0.3)	3.7	4.6 (0.2)	3.3 (0.9)	4.0 (0.3)
MnO	0.2	0.2	0.2	0.19 (0.04)	0.15 (0.02)	0.1	0.17 (0.05)	0.14 (0.04)	0.16 (0.05)
MgO	3.7	3.7	3.6	3.8 (0.1)	1.1 (0.1)	0.6	1.4 (0.1)	0.7 (0.4)	1.0 (0.1)
CaO	9.1	9.0	9.0	8.74 (0.09)	3.9 (0.4)	5.3	4.5 (0.1)	3.3 (0.6)	3.5 (0.3)
Na <sub>2</sub> O	2.9	2.9	3.0	2.9 (0.1)	4.5 (0.2)	3.6	4.3 (0.2)	4.8 (0.5)	4.5 (0.3)
K <sub>2</sub> O	0.7	0.7	0.7	0.68 (0.03)	1.6 (0.1)	2.1	1.6 (0.1)	1.8 (0.2)	1.63 (0.06)
$P_2O_5$	0.2	0.1	0.1	0.13 (0.02)	0.21 (0.03)	0.4	0.25 (0.03)	0.22 (0.04)	0.26 (0.04)
Original total	99.7	101.1	101.0	100.4 (0.9)	98.9 (1.0)	100.0	99.0 (0.3)	94.8 (1.3)	93.7 (0.7)
Na₂O + K₂O	3.6	3.6	3.7	3.6 (0.1)	6.0 (0.2)	5.7	5.9 (0.2)	6.5 (0.5)	5.7 (0.3)
Range of SO <sub>2</sub> (ppm)	n.a.	n.a.	n.a.	n.a.	57-138	n.a.	b.d.	395-1130	348-706
Cl (wt. %)	, n.a.	n.a.	n.a.	0.030 (0.003)	0.12 (0.02)	n.a.	0.13 (0.01)	0.19 (0.01)	0.16 (0.02)

176 Note. n.a. = not analyzed. b.d.l. = below detection limit. <sup>a</sup>Average values of compositions from

177 Chapter 1. Individual major element oxide compositions are normalized in wt. %, but original
178 total is presented.

179

### 180 3.3 Experimental equipment

All experiments were conducted at Institut de Sciences de la Terre Orléans (France) using internally heated pressure vessel (IHPV), following the procedure outlined in Scaillet and Evans (1999). Argon was used as a pressurizing medium mixed with different H<sub>2</sub> concentrations at room temperature to establish different  $fO_2$  conditions (ranging from NNO to NNO + 2), and using a Shaw membrane to establish and maintain the  $fH_2$  osmotic equilibrium (Scaillet et al., 1992). A factory calibrated pressure transducer was used to measure and monitor pressure (± 2)

187 MPa), whereas temperature was monitored with unsheathed type-S thermocouples, and the 188 temperature of the hotspot was maintained at  $< 10^{\circ}$ C and with an uncertainty of  $\pm 5^{\circ}$ C (Scaillet 189 et al., 1992). Experiments were raised up to run temperature directly from room temperature in 190 around an hour, where in each experimental run contained around 4-7 capsules, including one 191 sensor for  $fO_2$  (see below). Once the target run conditions were achieved, experiments were run 192 for 18-66 hours. Experiments were terminated with a drop quench device, allowing the sample 193 holder to fall in the cold part of the vessel at the bottom ( $< 50^{\circ}$ C) ensuring isobaric quenching 194 and cooling rates of 100°C/s. A fast quench device was set up as part of the experiments (Roux 195 & Lefèvre, 1992). Previous studies have shown that crystal-melt equilibrium is attained using the 196 same protocols. General observations of equilibrium include homogeneous glass composition 197 within a given charge, lack of chemical zoning in experimental crystals, and systematic changes 198 in petrologic variables (e.g. crystal content) with experimental variables (temperature and water 199 content). Equilibrium, rather than quench, textures are characteristic of the run products we 200 report and we believe they reflect attainment of chemical equilibrium at the desired run 201 conditions.

202

### 3.4 Calculation of $fO_2$ and dissolved water content in experimental charges

The oxygen fugacity ( $fO_2$ ) was calculated using a sensor capsule comprising two watersaturated (10mg added water), hand-pressed Ni-Pd-NiO powder pellets separated by inert ZrO powder (Taylor et al., 1992). We forgot to add a the sensor capsule for one experimental run (T = 1060°C, P = 200 MPa), and thus the final calculated  $fO_2$  for the fluid saturated charge (XH<sub>2</sub>O<sub>in</sub> = 1.0) as the result of the average between the two closest isobaric runs (T = 1060°C, P = 100 and 400 MPa). For undersaturated charges, we calculated the  $fO_2$  according to the following formula employed by Scaillet and Evans (1999):

$$fO_{2}(charge) = fO_{2}(sensor) + 2 \left[ log(H_{2}O)_{fluid} (charge)_{final} \right]$$
(1)

Where  $fO_2(\text{charge})$  is the calculated oxygen fugacity of the charge,  $fO_2(\text{sensor})$  is the sensor oxygen fugacity, and the H<sub>2</sub>O<sub>fluid</sub>(charge)<sub>final</sub> is the dissolved water content in the glasses. We could not calculate  $fO_2$  for dry charges (no water in the melt). Finally, the water content of the experimental glasses was obtained by calculating the fluid composition through mass-balance along with the solubility model for mixed  $H_2O-CO_2$  fluids from Shishkina et al. (2010), with a relative error of  $\pm 12$  %.

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218 3.5 Analytical techniques

219 Whole-rock analyses of samples, including the starting material pumice, were ground and 220 fused into a lithium tetraborate disk before being analyzed for whole rock major element oxide 221 compositions using X-Ray Fluorescence (XRF) at the Activation Laboratory in Ancaster, Canada 222 following the approach outlined in Norrish and Hutton (1969). Mineral proportions were 223 obtained by counting the phenocryst content using the JMicrovision software on plain polarized 224 images of the entire thin section (for the 1990 samples), and using a Pelcon mechanical point 225 counting stage (for the 2007 and 2014 samples). For each sample, we counted to 1400-1600 226 points to ensure statistically significant results with relative errors of < 10%. Results from point 227 counting and mineral analyses (see below) allowed us to calculate the matrix glass of the 2007 228 dome samples through mass balance. Such calculations are necessary to remove the effect of the 229 extensive microlite crystallization of the 2007 matrix, and we use such compositions for 230 geothermobarometric calculations.

231

We made general observations of experimental charges, and identified phases using the scanning electron microscope (SEM) at ISTO-Orléans, France and the Earth Observatory of Singapore. We determined the crystal content of the experimental charges by thresholding the greyscale of multiple back-scattered electron (BSE) images of a charge map using ImageJ, we estimated the volumetric area of phases in all charges. From these results, we calculated the mass proportions of the main minerals using standard mineral densities.

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Minerals and glass from natural samples and experimental charges were analyzed for
their chemical compositions on the JEOL JXA-8530F electron microprobe (EPMA) at the
Facility for the Analyses Characterization, Testing and Simulation (FACTS), Nanyang
Technological University, Singapore. Analyses of orthopyroxene, clinopyroxene, olivine and FeTi oxides were done on the EPMA in spot mode with 15 kV accelerating voltage, beam size of 1
µm, and 20nA beam current. Amphibole and plagioclase were analyzed in spot mode with 15 kV

245 accelerating voltage, beam size of 1 µm, and 10nA current. Glass was analyzed in spot mode 246 with 6 kV, and beam size of 10 µm and 5 nA current. The same conditions and procedures were 247 used to observe and analyze the glasses and minerals of the 2007 and 2014 rocks. For 248 clinopyroxene-hosted glass inclusions, we corrected for post-entrapment crystallization of the 249 melt inclusion walls (Bucholz et al., 2013). The relative errors for glasses are 0.4-0.8 % for SiO<sub>2</sub>, 250 Al<sub>2</sub>O<sub>3</sub>, and CaO; 1-3 % for MgO, FeO, Na<sub>2</sub>O, K<sub>2</sub>O; and 9-41% MnO, TiO<sub>2</sub>, P<sub>2</sub>O<sub>5</sub>, and Cl. For 251 pyroxenes, the relative errors are 0.2-1.0 % for SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, and CaO, MgO, FeO; 1-30% for 252 Na<sub>2</sub>O, K<sub>2</sub>O, and 2-55% MnO, TiO<sub>2</sub>, and Cr<sub>2</sub>O<sub>3</sub>. Relative errors for Fe-Ti oxide analyses are 0.3-253 0.6% for FeO, 0.5-7.5% for TiO<sub>2</sub>, 0.5-1.8% for MgO. For olivine analyses are 0.2-0.5% SiO<sub>2</sub>, 254 MgO, FeO; and 0.8-18% Al<sub>2</sub>O<sub>3</sub>, CaO, MnO. The relative error for the only amphibole analyses 255 are 0.3-0.8% for SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, CaO, FeO, MgO; 4.2-4.4% MnO, TiO<sub>2</sub>; and 11-30% for Na<sub>2</sub>O and 256 K<sub>2</sub>O.

257

258 The mineral end-members were calculated using their molar compositions as in Utami et 259 al. (in revision) and according to Deer et al. (1992). Pyroxenes are characterized with the Mg #  $(Mg \# = 100 \times Mg / [Mg + Fe^*])$  in mol, where Fe<sup>\*</sup> is total iron as Fe<sup>2+</sup>). We further characterize 260 261 pyroxenes using their wollastonite (Wo =  $100 \times Ca / [Ca + Mg + Fe^*]$ ), ferrosilite (Fs =  $100 \times Fe$ / [Ca + Mg + Fe<sup>\*</sup>]), and enstatite (En =  $100 \times Mg / [Ca + Mg + Fe<sup>*</sup>]$ ) endmembers. Plagioclase 262 263 end member compositions were classified using their molar compositions for anorthite (An = 264  $100 \times \text{Ca} / [\text{Ca} + \text{Na} + \text{K}]$ , albite (Ab =  $100 \times \text{Na} / [\text{Ca} + \text{Na} + \text{K}]$ ), and orthoclase (Or =  $100 \times \text{Ca} / (\text{Ca} + \text{Na} + \text{K})$ ) 265 K / [Ca + Na + K]). Olivine is characterized using the forsterite content (Fo =  $100 \times Mg / [Mg +$ 266 Fe\*]), whereas amphibole is classified according to the compositional end-members outlined in 267 Leake et al. (1997), as determined by Ridolfi et al. (2010). Glass compositions were 268 characterized using their normalized to 100% anhydrous major element oxides and total alkali-269 silica (TAS) composition. It was difficult to obtain good quantitative analyses of plagioclase 270 crystals in experimental charges as they are typically small and may contain Fe-Ti oxide 271 inclusions. To be able to report approximate An contents of plagioclase, we have filtered for 272 analyses with FeO\* < 1 wt. % and TiO<sub>2</sub> < 0.1 wt. %, and subtracted the composition of 273 magnetite from the electron microprobe analyses to match these constraints. We report these 274 recalculated compositions to fit the structural formulae.

### 276 **4 Results**

4.1 Petrological observations and geothermobarometry of the 1990, 2007 and 2014 magmas

278 We made new petrological observations and geothermobarometric estimates from the 279 deposits of the 1990, 2007, and 2014 eruptions (Tables 1-4). Whole rock major elements vary 280 over a small range across eruptions and are classified as basaltic andesite (Table 1). The 281 calculated matrix compositions, the measured interstitial glasses, and clinopyroxene-hosted melt 282 inclusions are of a similar dacitic composition. The three eruptions have deposits with different 283 phenocryst (> 100  $\mu$ m) content: 31-47 wt. % in the 1990 pumices; 70-72 wt. % for the 2007 284 dome; and 29-35 wt. % in 2014 pumices (Table 2). The mineral assemblage for all three 285 eruptions includes phenocrysts of plagioclase (Plag), clinopyroxene (Cpx), orthopyroxene (Opx), 286 and magnetite (Mag). The 1990 and 2014 pumices also host partially reacted amphibole (Amph), 287 and we have also identified pseudomorphs of Amph megacrysts and symplectites after Ol 288 breakdown in the 2007 dome, meaning that both Amph and Ol may have also been stable in the 289 2007 magma. We also found partially reacted olivine (Ol) microphenocrysts ( $< 100 \mu m$ ) in the 290 2014 pumice.

291

Table 2: Mineral assemblage and phenocryst content of samples from 1990, 2007, and 2014
eruptions.

Sample ID	90-23	PL1	2007A	2007B	14-PUMI-1	14-PUMI-2
Eruption	1990	1990	2007	2007	2014	2014
Phase <sup>b</sup>	wt. %	wt. %				
Glm*	51.0	64.5	27.8*	30.2*	70.5	65.1
Plag	36.1	13.1	48.2	47.7	21.2	19.9
Срх	4.5	6.0	4.6	3.9	1.2	2.1
Орх	4.1	7.9	10.6	12.0	3.0	7.5
Mag	4.3	8.4	8.5	6.2	3.6	4.7
Amph		0.1	0.3		0.5	
OI						0.7
Total	100.0	100.0	100.0	100.0	100.0	100.0
Phenocryst content	49	35	72	70	29	34

295 Notes. <sup>*a*</sup>For these samples Glm means matrix and include glass and microlites. <sup>*b*</sup>Opx =

296 *orthopyroxene;* Cpx = clinopyroxene; Plag = plagioclase; Mag = magnetite; Ol = olivine;

- 297 Amph = amphibole.
- 298

299 Accessory minerals include ubiquitous apatite (Apa), which is found in all three 300 eruptions; quartz (Qz) rich enclaves occur in the 1990 late-stage pumice; and cristobalite (Crs) is 301 found as vesicle infill in the 2007 dome samples (See Supplementary Material). Ilmenite (Ilm) is 302 present as rims around Mag in the 1990 late-stage pumice, and as exsolution lamellae in Mag in 303 the 2007 deposits (See Supplementary Material). The compositions of the main minerals of the 304 three eruptions are also broadly similar, with the histograms of compositional mineral indicators 305 (e.g. Mg #) that typically overlap for the three eruptions (Fig. 1a-c; Table 3), and are also similar 306 to those of previous studies from the same eruptions (Cassidy et al., 2016, e.g. 2019; Jeffery et 307 al., 2013).

308

**Table 3**: Summary of representative mineral compositions from the 1990-2014 eruption deposits.

Mineral	Срх	Срх	Срх	Орх	Орх	Орх	Mineral	Amph <sup>a</sup>	Amph
Eruption year	1990	2007	2014	1990	2007	2014	Eruption year	1990	2014
rock type/stage	main-stage pumice	dome	pumice	main-stage pumice	dome	pumice	rock type/stage		
Texture	cores, glom	cores, glom	cores, glom	cores, glom	glom	cores, glom	Texture		
n	8	16	17	11	7	9	n	18	4
wt. %									
SiO <sub>2</sub>	51.6 (0.7)	51.8 (0.4)	51.1 (0.6)	52.8 (0.2)	52.9 (0.1)	52.6 (0.3)	SiO <sub>2</sub>	41.6 (0.6)	43.0 (2.7)
TiO <sub>2</sub>	0.5 (0.3)	0.33 (0.04)	0.4 (0.1)	0.19 (0.03)	0.16 (0.01)	0.19 (0.02)	TiO <sub>2</sub>	1.5 (0.1)	1.7 (0.7)
Al <sub>2</sub> O <sub>3</sub>	1.8 (0.6)	1.7 (0.1)	2.2 (0.8)	1.0 (0.2)	0.9 (0.2)	1.0 (0.2)	$AI_2O_3$	14.9 (0.7)	11.9 (3.4)
FeO*	9.2 (0.6)	9.5 (0.1)	9.5 (0.4)	19.5 (0.3)	19.2 (0.2)	19.4 (0.3)	Cr <sub>2</sub> O <sub>3</sub>	0.05 (0.03)	
MnO	0.5 (0.1)	0.51 (0.03)	0.5 (0.1)	1.0 (0.2)	0.90 (0.04)	0.91 (0.04)	FeO*	11.3	10.5 (1.8)
MgO	14.5 (0.1)	14.3 (0.2)	14.5 (0.1)	23.1 (0.2)	22.84 (0.07)	23.1 (0.2)	MnO	0.15	0.13 (0.07)
CaO	21.1 (0.4)	20.8 (0.3)	20.6 (0.4)	1.4 (0.2)	1.44 (0.08)	1.45 (0.03)	MgO	13.9	14.7 (0.1)
Na <sub>2</sub> O	0.27 (0.02)	0.29 (0.02)	0.3 (0.1)	0.01 (0.01)	0.02 (0.01)	0.015 (0.003)	CaO	12.0 (0.2)	12.0 (0.6)
K <sub>2</sub> O							Na₂O	2.24	2.1 (0.4)
Total	99.4 (0.2)	99.3 (0.4)	99.0 (0.3)	99.0 (0.3)	98.4 (0.1)	98.7 (0.4)	K₂O	0.31 (0.02)	0.4 (0.02)
							CI	0.016 (0.005)	
							F	0.13 (0.07)	
							Total	97.9 <sup>´</sup> (0.4)	96.6 (0.2)
Pyroxene end-me	embers <sup>b</sup>							( )	
Wo	43.6 (0.8)	43.2 (0.6)	42.0 (0.6)	2.9 (0.4)	3.0 (0.2)	3.1 (0.1)	Amph end- member <sup>d</sup>	Mg-Hst	Ts-Prg
En	41.6 (0.3)	41.3 (0.5)	43.2 (0.8)	65.9 (0.3)	65.9 (0.3)	63.7 (0.4)			
Fs	14.9 (0.9)	15.5 (0.3)	14.8 (0.6)	31.2 (0.5)	31.1 (0.2)	33.2 (0.5)			
Mg #	73.6 (1.3)	72.8 (0.5)	73.9 (0.9)	67.8 (0.5)	67.9 (0.2)	65.7 (0.5)			

Mineral	OI	OI	Mineral	Plag	Plag	Plag	Plag	
Eruption year	2014	2014	Eruption year	1990	2007	2014	2014	
rock type/stage			rock type/stage	main-stage pumice	dome	pumice	pumice	
Texture	low-Fo	high-Fo	Texture	microlite/rim	rim	core	rim	
n	9	3	n	13	28	3	2	
SiO <sub>2</sub>	36.8 (0.2)	37.55 (0.33)	SiO <sub>2</sub>	49.1 (1.2)	50 (2)	44.6 (0.1)	51.0 (0.1)	
TiO <sub>2</sub>	0.03 (0.02)	0.02 (0.02)	TiO <sub>2</sub>		0.03 (0.03)	0.02 (0.02)	0.02 (0.01)	
$AI_2O_3$	0.01 (0.02)	0.03 (0.03)	$AI_2O_3$	31.5 (1.0)	31 (1)	33.4 (0.2)	29.3 (0.1)	
FeO*	26.6 (1.1)	23.41 (1.41)	FeO*	0.32 (0.05)	0.63 (0.06)	0.55 (0.02)	0.67 (0.01)	
MnO	0.68 (0.05)	0.52 (0.07)	MnO		0.01 (0.01)			
MgO	34.5 (0.99)	37.06 (0.87)	MgO	0.04 (0.02)	0.05 (0.02)	0.06 (0.02)	0.08 (0.01)	
CaO	0.17 (0.04)	0.2 (0.05)	CaO	14.8 (1.0)	14 (1)	18.4 (0.2)	13.42 (0.05)	
Na₂O			Na <sub>2</sub> O	3.4 (0.6)	3.5 (0.7)	1.2 (0.1)	3.9 (0.1)	
K₂O			K <sub>2</sub> O	0.06 (0.02)	0.09 (0.04)			
			Total	99.1 (0.5)	98.9 (0.5)	98.3 (0.2)	98.35 (0.01)	
$P_2O_5$	0.04 (0.02)	0.03 (0.03)						
Total	98.9 (0.31)	98.8 (0.51)						
			Plag end-member <sup>c</sup>					
Fo content <sup>e</sup>	69.8 (1.5)	73.8 (1.6)	An	70.7 (5.3)	68 (7)	89.2 (0.9)	65.8 (0.5)	
			Ab	29.0 (5.2)	31 (6)	10.8 (0.9)	34.2 (0.5)	
			Or	0.3 (0.1)	0.6 (0.3)	n.a.	n.a.	

313 Notes. n.a. = not analyzed.  $FeO^*$  or  $Fe^*$  = total Fe as  $Fe^{2+}$ . Glom = glomerocryst. See Table 2 for meaning of mineral abbreviations.

<sup>a</sup>Average amphibole composition From Utami et al. (in revision). <sup>b</sup>Pyroxene end-members: wollastonite,  $Wo = 100 \times Ca / [Mg + Fe^*]$ 

315 + Ca]; enstatite,  $En = 100 \times Mg / [Mg + Fe^* + Ca]$ ; ferrosilite,  $Fs = 100 \times Fe^* / [Mg + Fe^* + Ca]$ ;  $Mg \# = 100 \times Mg / [Mg + Fe^*]$ .

316 *Constant Constant Consta* 

317 *x K*/[*Na* + *Ca* + *K*]. <sup>*d*</sup>*Amphibole end-members according to Leake et al. (1997) as calculated in Ridolfi et al. (2010). Mg-Hst* =

318 magnesio-hastingsite; Ts-Prg = Tschermakite-Pargasite. <sup>e</sup>Olivine Fo content,  $Fo = 100 \times Mg / [Mg + Fe^*]$ .

320	We made geothermobarometric estimates for the 1990-2014 magma storage and ascent
321	conditions from mineral and glass/whole rock/matrix compositions (Table 4). For this we used
322	core-whole rock and rim-interstitial/matrix glass pairs (see also Utami et al. (in revision) for the
323	1990 eruption). The pyroxene-liquid or two pyroxene equilibria formulations of Putirka (2008)
324	give temperatures that range from about 1030°C for the crystal cores, to about 975°C (down to
325	950°C) for the crystal rims. Moreover, the mineral and glass compositions of the three eruptions
326	give pre-eruptive temperatures that are within about $\pm$ 30°C (Table 4). These temperature
327	estimates also overlaps with those obtained from amphibole-melt equilibria at about 960°C for
328	the 1990 and 2014 eruptions. Pressure estimates we obtained come from the amphibole-melt
329	geobarometer formulation of Putirka (2016), where pressures range from 450 MPa up to 700
330	MPa for the 1990 and 2014 eruptions, respectively.

331

**Table 4.** *Petrological storage conditions estimates from mineral-melt geothermobarometers,* 

333 hygrometers and H<sub>2</sub>O-CO<sub>2</sub> saturation models for the 1990, 2007, and 2014 mineral populations.

Year		1	990 <sup>a</sup>				2007			2	2014	
Crystal Zone	c	ore	rir	m			Core <sup>f</sup>		C	ore	rin	n
	n	T (°C)	n	T (°C)		n	T (°C)		n	T (°C)	n	T (°C)
Opx- melt <sup>b</sup>	75	1023 (19)	37	976 (17)		2	1032 (2)		15	1039 (15)	1	974
Cpx- melt <sup>b</sup>	122	1043 (13)	23	1045 (34)		5	1029 (3)		18	1028 (6)		
Two- px <sup>b,c</sup>	67	1023 (40)	67	1030 (43)		2	980 (9)				7	948 (8)
OI-melt <sup>b</sup>									8	1018 (5)		
Year			1990				2007				2014	
	type	n	H <sub>2</sub> O <sub>melt</sub> (wt. %)	P <sub>sat</sub> (MPa) <sup>e</sup>	zone	n	H <sub>2</sub> O <sub>melt</sub> (wt. %)	P <sub>sat</sub> (MPa)		n	H <sub>2</sub> O <sub>melt</sub> (wt. %)	P <sub>sat</sub> (MPa) g
Plag- melt <sup>d</sup>	rim	9	4.1 (0.2)	172 (1)	core	82	4.4 (0.2)	189	core	2	6.0 (0.2)	214
_	micr	9	4.1 (0.2)	170 (2)	rim	28	4.4 (0.2)	189	rim	2	4.0 (0.2)	168
Year			1990				2007				2014	
		n	T (°C)	P <sub>store</sub> (MPa)		n	T (°C)	P <sub>store</sub> (MPa)		n	T (°C)	P <sub>store</sub> (MPa)
Amph- melt <sup>h</sup>		20	960 (10)	450 (80)			not analyz	ed		8	940 (80)	700 (90)

<sup>334</sup> 

335 Note. Temperatures from pyroxene-based geothermometers were calculated using Putirka

336 (2008) (SEE for Opx-liquid =  $\pm 39^{\circ}$ C; Cpx-liquid =  $\pm 42^{\circ}$ C; two-pyroxene =  $\pm 60^{\circ}$ C) and

337 Andersen et al. (1993) ( $\pm$  37°C), assuming a constant pressure of 100 MPa. Temperatures from 338 amphibole-melt geothermometer were calculated using Putirka (2016) (SEE =  $\pm 30^{\circ}$ C), with the Al/Si equilibrium test described in Li et al. (2021) ( $^{Al/Si}K_d = 0.96 \pm 0.4$ ). Dissolved water content 339 340 of main-stage pumices were calculated using the plagioclase-melt hygrometer of Waters and 341 Lange (2015) ( $\pm$  0.35 wt. %), and the saturation pressures were calculated using the Ghiorso 342 and Gualda (2015) mixed  $H_2O$ - $CO_2$  saturation model. The saturation pressure and water content 343 calculated for 2014 pumices were calculated in the same way as the 1990 eruption, but with the 344 average estimated using the temperature calculated from two-pyroxene geothermometer of 345 Putirka (2008) (SEE  $\pm$  37°C). Temperatures from the olivine-melt geothermometer were 346 calculated using equation 21 of Putirka (2008) (SEE  $\pm$  36°C). See Table 2 for meaning of 347 abbreviations; px = pyroxene; micr = microlite. Parameters in italics are average calculated 348 values from Utami et al. (in revision) and this study. Numbers in parentheses refer to 1 standard 349 deviation (sigma), but where the 1 sigma is lower than the error, the error is reported instead. 350 <sup>a</sup>The storage temperatures and pressures, water content and saturation pressures of the 1990 351 pumices were calculated in Utami et al. (in revision) using temperatures estimated from orthopyroxene rims and interstitial glass pairs. <sup>b</sup>Ol-melt geothermometer (Eq. 21); Opx-melt 352 353 geothermometer (Eq. 28a); Cpx-melt geothermometer (Eq. 33); two-pyroxene geothermometer 354 (Eq. 37) from Putirka (2008). <sup>c</sup>QUILF two-pyroxene geothermometer (Andersen et al., 1993). 355 <sup>d</sup>Plag-melt hygrometer from Waters and Lange (2015) with minimum temperature estimates from 356 Opx-melt and two-pyroxene geothermometers from Putirka (2008). <sup>e</sup>Saturation pressure and 357 dissolved water contents for the 1990 eruption were estimated using Plag rims and microlites from Utami et al. (in revision) and the average Opx-melt temperature of  $943 \pm 48^{\circ}$ C. <sup>f</sup>For 2007 358 359 dome samples, the calculated interstitial melt was used for rim Opx-liquid geothermometer 360 calculations and Plag-melt hygrometer.<sup>8</sup>Saturation pressure is calculated with the MagmaSat 361 application using a mixed  $H_2O-CO_2$  model (Ghiorso and Gualda, 2015), and the average water content value from with the Plag-melt hygrometer. <sup>h</sup>Eq 7a (pressure) from Amph-melt 362 geothermobarometer (Putirka, 2016) with Al/Si equilibrium  $K_d$  constant proposed in Li et al. 363 364 (2020).

365

The water content in the melt for the 1990 and 2014 magmas was calculated using the plagioclase-melt hygrometer of Waters and Lange (2015) using rim-interstitial glass pairs range 368 from 3 to 4 wt. % for an averaged temperature of  $T = 975^{\circ}C$ , which is consistent with previous 369 estimates in Utami et al. (in revision). These water contents in the melt correspond to water 370 saturation pressures of 170-190 MPa, which also is similar to the range of pressures obtained in 371 Utami et al. (in revision). Surprisingly, the water content in the melt estimated for the 2007 372 samples using plagioclase rim and core and the calculated matrix glass is ~4 wt. % water, and 373 thus similar to those of the 1990 and 2014 eruptions. 374 Another important aspect to consider towards improving our understanding the volatile 375 content of these eruptions is the likely existence of an exsolved fluid phase at pre-eruptive 376 eruptions. Utami et al. (in revision) calculated the amount of S that could be degassed for the 377 1990 eruption and estimated that 0.01-0.12 Mt could reside in an excess volatile phase. We have 378 done the same calculation of the sulfur budget for the 2014 magma and satellite measurements; 379 (see Methodology section), and we find a range of excess volatile phase of 0.08-0.10 Mt of S 380 (Table 11).



Figure 1. Histograms of representative mineral compositions from the 1990-2014 eruptions: a.
pyroxenes, b. olivine, and c. plagioclase. Plagioclase composition from the 2014 eruption comes
from a combination of this study and Cassidy et al. (2016) and those of the 1990 from Utami et

*al. (in revision).* 385

### 387 4.2 General observations from the experimental results

388 Successful experimental charges range from complete glass to almost fully crystallized. 389 Glass-rich charges are devoid of vesicles (Fig. 2). Most charges (Fig. 2b-e, g-h) have a 390 porphyritic texture, with subhedral to euhedral crystals set in interstitial glass with vesicles. 391 Crystals may vary in size from phenocrysts (> 100  $\mu$ m) to microlites (< 10  $\mu$ m). Typically, Opx 392 and Ol are subhedral to euhedral, commonly with Mag and Plag inclusions (>100 µm) (Fig. 2c). 393 Some charges have clusters of Fe-Ti oxides microlites, as well as Fe-Ti oxides distributed 394 throughout the charge (Fig. 2d). Despite the large range in crystal sizes, minerals and glass 395 appears to be unzoned, according to BSE images, which is likely indicative of compositional 396 equilibrium. Due to the small size of many crystals, it was very challenging to obtain a good 397 quantitative chemical analysis, and thus we relied on mineral identification using EDS spectra 398 obtained in SEM or electron probe. We obtained glass compositions from glass-rich 399 experimental charges or for those where glass "pools" larger than about 10 µm, and we found 400 that they are homogeneous in composition (Fig. 2a-d). For charges with low water contents, 401 phases are often too small to analyze. For charges that failed i.e. where the fluid escaped the 402 capsule during the experiment, we consistently observe a very high crystal content and a quench 403 texture, with abundant microlites of Opx and Plag, and rare Ilm and glass; these were not used 404 further.

405

406 4.3 Phase relation, crystallinities, and mineral assemblages

407 The full experimental phase assemblage of all charges includes  $Opx \pm Cpx \pm Plag \pm Mag$ 408  $\pm$  Ilm  $\pm$  Amph  $\pm$  Ol + glass (Fig. 2a-h). We did not identify experimental apatite, but we cannot 409 rule out that it is not present. The crystal content from experimental charges varies from virtually 410 none to about 88 wt. % (Table 5), and increases as temperature and water content decreases, and 411 also as  $fO_2$  also increases. We produced liquidus or near-liquidus experiments with < 1 wt. % of 412 crystals at 1060°C, NNO at water saturated conditions (about 5-9 wt. % H<sub>2</sub>O in the melt) and at 413 200 and 400 MPa (Fig. 2a). These conditions match with the model liquidus temperature 414 calculated from rhyolite-MELTS in Utami et al. (in revision). At 100 and 200 MPa and 1000°C,

- the crystal content is varies between about 68 to 44 wt. % for 2-4 wt. % water in the melt.
- 416 Increasing the temperature to 1060°C decreases the crystal content to 52 to 31 wt. % at 3-4 wt. %
- 417 water in the melt, but it increases up to 74 wt. % for < 2 wt. % in the melt. Similar changes in
- 418 crystal content with temperature and water content in the melt for different experimental
- 419 pressures. The effect of increasing  $fO_2$  from NNO to NNO + 2 is to increase somewhat the
- 420 proportion of magnetite with comparable crystal content.
- 421



423 *Figure 2* Annotated BSE images of selected representative experimental charges in order of

424 decreasing crystallinity. a. 1060°C-200 MPa-NNO. b. 1000°C-100 MPa-NNO + 2. c. 950°C-150

425 MPa-NNO. d. 1000°C-200 MPa-NNO. e. 950°C-200 MPa-ΔNNO + 2. f. 950°C-200 MPa-NNO

426 + 2. g.  $950^{\circ}C$ -400 MPa-NNO. h. Inset of charge in g. labelled with abbreviated mineral names

427 (*Gl* = glass, *Plag* = plagioclase, *Cpx* = clinopyroxene, *Opx* = orthopyroxene, *Mag* = magnetite,

428 Amph = amphibole).

429

430 The relationship between crystallinity, temperature, and water content in the melt can be 431 used to obtain first order constraints on the pre-eruptive conditions (Fig. 3). Using a pre-eruptive 432 temperature storage of about  $975 \pm 39^{\circ}$ C for the three eruptions, as suggested by 433 geothermobarometric data of all eruptions (Table 4), we find that the 1990 and 2014 melt coul 434 contain about 4-6 wt. % water, or 4-5 wt. % at 100-200 MPa. The much higher phenocryst 435 content of the 2007 dome would imply significantly lower water content in the melt (~1 wt. %). 436 However, as we have discussed in the geothermobarometry results section above, we believe this 437 does not reflect the pre-eruptive conditions, as the plagioclase-matrix equilibria suggest similar 438 water contents in the melt for the 2007 and the 1990 and 2014 eruptions, and rather reflects re-439 equilibration and maturation during slow ascent, as further discussed below.

440

441 We find that experimental Opx and Plag are present at all conditions below the liquidus, 442 and Cpx stability field has a somewhat smaller stability field (Fig 4a-c, Fig. S4 of Supplementary 443 Material). We identified experimental amphibole at 950-1000°C and 200-400 MPa, with 4.5-6 444 wt. % water in the melt (Fig 4a-b). Despite our efforts we were not able to positively identify 445 Amph at lower pressures (< 200 MPa) and 950°C charges owing to the high crystallinity and 446 small crystal sizes, although we suspect it is stable by comparing to the results of other phase 447 equilibria studies with similar bulk compositions (e.g. Erdmann et al., 2016; Pichavant et al., 448 2002; Prouteau & Scaillet, 2003). We identified experimental Ol with 3-4 wt. % water in the 449 melt at NNO and < 150 MPa (Fig. 4b). At NNO + 2, Ol stability extends to higher pressures and 450 water content (at 200 MPa, 1000°C, and 3-6 wt. % water in the melt; Fig 4c). The stability of Fe-451 Ti oxides is also controlled by the dissolved water content and  $fO_2$ . Ilmenite is stable at lower 452 dissolved water contents than magnetite, whereas with increasing  $fO_2$  makes magnetite more 453 stable at low water contents. Given the wide stability field of pyroxenes and plagioclase, the

- 454 most diagnostic mineral to establish the pre-eruptive conditions is the presence of Amph in the
- 455 1990 and 2014, and xenocrysts and pseudomorphs in the 2007 lava domes. Amph stability
- 456 requires water contents of  $\ge$  3 wt. % in the melt, T < 1000°C and P  $\ge$  200 MPa. Cassidy et al.
- 457 (2019) conducted experiments using the 2014 Kelud rock, and the stability of anhydrous
- 458 minerals they reported are quite similar to those we found, but did not explore the P-T-water
- 459 content in the melt at which Amph is stable; thus they did not experimentally grow the mineral.

- 461 **Table 5.** Summary of experimental results, including crystal content, mineral assemblage and normalized mineral proportions and
- 462 original major element totals. Dissolved H<sub>2</sub>O and CO<sub>2</sub> concentrations were calculated using the H<sub>2</sub>O-CO<sub>2</sub> solubility model of Shiskina
- 463 *et al.* (2010).

				Calculate	ed melt vola			Phase	e propoi	tions <sup>e,f</sup>	(wt. %)				
Charge ID	log(fO <sub>2</sub> ) <sup>a</sup> (bar)	$\Delta NNO^{b}$	$XH_2O_{in}{}^c$	$XH_2O_{fluid}$	H <sub>2</sub> O <sub>melt</sub> (wt. %)	CO <sub>2-melt</sub> (wt. ppm)	Орх	Срх	Plag	Mag	llm	OI	Amph	GI	Total
T = 1000°C, I	P = 100 MPa, t	= 66 hours													
SUPL1-1	-9.5	0.8	1.00	1.0	3.5	0	6.0	2.8	30.4	1.1		3.4		56.3	100
SUPL1-2	-9.8	0.6	0.80	0.7	3.0	162	12.4	8.1	29.6	2.1				47.8	100
SUPL1-3	-10.3	0.1	0.66	0.6	2.5	267	13.2	8.7	33.9	2.7				41.5	100
SUPL1-4	-10.9	-0.6	0.56	0.5	2.3	312	14.9	8.3	42.8	2.3				31.7	100
T = 1000°C, I	P = 150 MPa, t	= 19 hours													
SUPL1-5	-8.5	1.8	1.00	1.0	4.4	0	18	25	2					55.0	100
SUPL1-6	-8.7	1.6	0.82	0.8	3.8	250	3.7	2.3	23.3	0.5		4.3		65.9	100
SUPL1-7	-9.0	1.4	0.67	0.6	3.2	414	13.1	7.6	29.7	1.8				47.8	100
SUPL1-8	-9.0	1.3	0.63	0.6	3.1	444	13.4	12.1	34.6	1.8				38.1	100
SUPL1-9	-9.5	0.9	0.41	0.3	2.3	633	13.4	5.3	55.9	3.7				21.8	100
SUPL1-10	n.a.	n.a.	0.00	0.0	0.0	888	23.7		57.8		0.7			17.9	100
T = 1000°C, I	P = 200 MPa, t	= 22 hours													
SUPL1-11	-9.1	1.2	1.00	-	-	-	-	-	-	-	-	-	-	-	-
SUPL1-12	-9.4	0.9	0.84	0.7	4.4	421	7.6	4.1	25.9	0.9				61.6	100
SUPL1-13	-9.6	0.8	0.75	0.6	3.9	573	5.1	6.8	29.2	1.0				57.9	100
SUPL1-14	-9.9	0.4	0.55	0.4	3.1	835	11.2	8.2	35.8	1.2				43.5	100
SUPL1-15	-10.7	-0.4	0.30	0.2	1.8	1090	14.6	15.3	36.0	0.6				33.5	100
SUPL1-16	n.a.	n.a.	0.00	0.0	0.0	1263	25.3		53.1		0.6			21	100
T = 1000°C, I	P = 400 MPa, t	= 23 hours													
SUPL1-17	-9.2	1.1	1.00	1.0	8.3	-	-	-	-	-	-	-	-	-	-
SUPL1-18	-9.5	0.8	0.85	0.7	6.5	1681	13.9	1.8	5.6	0.3			< 0.01	78.4	100
SUPL1-19	-10.4	0.0	0.54	0.3	3.8	2977	16.5	12.0	21.6	0.3				49.6	100
SUPL1-20	-11.4	-1.0	0.27	0.1	1.9	3438	16.0	12.0	28.5	0.8				42.7	100
SUPL1-21	n.a.	n.a.	0.00	0.0	0.0	3638	25.0		37.7		0.4			36.9	100

T = 950 °C, P = 150 MPa, t = 66 hours

				Calculated melt volatile content <sup>d</sup> P					Phase	e propor	tions <sup>e,f</sup>	(wt. %)			
Charge ID	log(fO <sub>2</sub> ) <sup>a</sup> (bar)	$\Delta NNO^{b}$	$XH_2O_{in}{}^c$	XH <sub>2</sub> O <sub>fluid</sub>	H <sub>2</sub> O <sub>melt</sub> (wt. %)	CO <sub>2-melt</sub> (wt. ppm)	Орх	Срх	Plag	Mag	llm	OI	Amph	GI	Total
SUPL1-22	-10.0	1.1	1.00	1.0	4.3	0	7.1	1.2	19.3	3.5		5.1		63.8	100
SUPL1-23	-10.2	0.9	0.84	0.8	3.7	245	14.4	3.6	29.3	4.3				48.4	100
SUPL1-24	-10.3	0.8	0.75	0.7	3.5	327	16.8	7.6	27.8	0.6				47.2	100
SUPL1-25	-10.8	0.3	0.49	0.4	2.5	582	14.0	10.8	33.8	0.8				40.6	100
SUPL1-26	-11.0	0.1	0.39	0.3	2.2	647	13.5	11.0	42.5	0.6				32.4	100
SUPL1-27	n.a.	n.a.	0.00	0.0	0.0	890	35.5		52		0.5			12	100
T = 950°C, P	P = 200 MPa, t ⊧	= 49 hours													
SUPL1-28	-10.7	0.4	1.00	1.0	5.1	0	19.9	13.2	28.6	3.0				35.2	100
SUPL1-29	-10.9	0.2	0.86	0.8	4.6	291	19.0	7.6	38.5	4.3				30.7	100
SUPL1-30	-11.1	0.0	0.68	0.6	3.8	588	17.7	20.6	31.9	0.9				28.8	100
SUPL1-31	-11.4	-0.3	0.54	0.4	3.2	797	12.8	15.4	44.0	1.6				26.3	100
T = 950°C, P	P = 400 MPa, t ⊧	= 67 hours													
SUPL1-32	-9.8	1.3	1.00	1.0	8.1	0	17.9	8.4	10.7	3.2				60.0	100
SUPL1-33	-10.3	0.8	0.78	0.6	5.8	2066	14.3	3.7	16.0	3.0			< 0.01	63.0	100
SUPL1-34	-10.6	0.6	0.71	0.4	4.9	2526	11.2	3.9	29.4	3.2				52.3	100
SUPL1-35	-11.0	0.1	0.50	0.3	4.3	3029	11.0	2.9	35.2	3.2				47.7	100
SUPL1-36	-11.5	-0.4	0.35	0.1	2.6	3322	12.9	10.2	30.5	2.7				43.6	100
SUPL1-37	n.a.	n.a.	0.00	0.0	0.0	3685	30.4		39.4		0.4			29.8	100

				Calculate	ed melt vola	atile content <sup>d</sup>			Phas	e propoi	rtions <sup>e,t</sup>	(wt. %)			
Charge ID	log(fO <sub>2</sub> ) <sup>a</sup> (bar)	$\Delta NNO^{b}$	$XH_2O_{in}{}^c$	$XH_2O_{fluid}$	H <sub>2</sub> O <sub>melt</sub> (wt. %)	CO <sub>2-melt</sub> (wt. ppm)	Орх	Срх	Plag	Mag	llm	OI	Amph	GI	Total
T = 1060°C.	P = 100 MPa. t	= 18 hours													
SUPL1-38	-8.1	1.4	1.00	1.0	3.6	0			28.7	1.9				69.3	100
SUPL1-39	-8.4	1.1	0.78	0.7	3.0	168			56.8	2.4		< 0 1		40.8	100
SUPL1-40	-8.8	0.7	0.73	0.6	2.8	220			46.0	3.6		0.9		49.5	100
SUPL1-41	-9.2	0.2	0.67	0.6	2.6	251	4.9	3.1	43.6	0.4				48.0	100
SUPL1-42	-10.3	-0.9	0.37	0.3	1.7	424	9.5	7.7	43.6	3.7				35.5	100
SUPL1-43	n.a.	n.a.	0.00	0.0	0.0	563	22.5	0.8	38.1	•	12.6			26.0	100
T = 1060°C, I	P = 200 MPa, t	= 94 hours													
SUPL1-44	-8.4	1.1	1.00	1.0	5.3	0			0.8					99.2	100
SUPL1-45	-8.9	0.5	0.72	0.5	3.6	696	16.6	2.1	16.9	2.1				62.4	100
SUPL1-46	-9.3	0.1	0.53	0.3	2.8	914	12.9	7.9	28.2	0.9				50.1	100
SUPL1-47	-10.3	-0.8	0.27	0.1	1.4	1153	20.1	1.3	37.6	2.0				39.0	100
SUPL1-48	n.a.	n.a.	0.00	0.0	0.0	1260	21.5		42.6	0.9				35.0	100
T = 1060°C, I	P = 400 MPa, t	= 49 hours													
SUPL1-49	-8.6	0.9	1.00	1.0	8.6	0								100.0	100
SUPL1-50	-10.1	-0.7	0.59	0.2	3.0	3206	2.7	1.8	13.8	0.2				81.5	100
SUPL1-51	-11.6	-2.1	0.22	0.0	1.1	3543	9.1	6.4	21.8	1.7				61.0	100
T = 1000°C, I	P = 100 MPa, t	= 77 hours													
SUPL1-52	-8.4	1.9	1.00	1.0	3.5	0	5.8	5.6	25.8	3.2		0.9		58.7	100
SUPL1-53	-8.6	1.7	0.83	0.8	3.0	143	11.0	2.5	29.2	2.8				54.5	100
SUPL1-54	-8.7	1.6	0.77	0.7	2.9	178	11.8	4.3	30.6	2.8				50.5	100
SUPL1-55	-9.0	1.3	0.57	0.5	2.3	311	13.7	4.1	38.1	3.1				41.0	100
SUPL1-56	-9.4	0.9	0.42	0.3	1.8	399	16.0	11.1	33.2	0.4				39.3	100
SUPL1-57	n.a.	n.a.	0.00	0.0	0.0	564	23.9		44.7		2.4			29.0	100
T = 1000°C,	P = 200 MPa, t	= 19 hours													
SUPL1-58	-8.5	1.8	1.00	1.0	5.2	0	14.3	3.1	15.7	0.6				66.0	100
SUPL1-59	-8.7	1.6	0.85	0.8	4.5	355	2.0	1.5	20.2	1.0		4.0		71.2	100
SUPL1-60	-8.9	1.4	0.73	0.6	3.9	575	6.4	4.7	21.4	3.3		5.9		58.4	100
SUPL1-61	-9.1	1.2	0.59	0.5	3.5	726	13.2	6.3	28.3	1.6				50.6	100
SUPL1-62	-10.0	0.3	0.29	0.2	1.9	1086	15.5	13.0	28.0	3.0				40.6	100
SUPL1-63	n.a.	n.a.	0.00	0.0	0.0	1263	22.5		39.0		9.4			29.0	100
T = 950°C, P	= 200 MPa, t =	= 71 hours													
SUPL1-64	-9.3	1.8	1.00	1.0	5.1	0	15.3	18.0	19.9	1.0				45.7	100
SUPL1-65	-9.5	1.7	0.86	0.8	4.5	303	16.7	5.2	31.4	3.7				43.0	100

### **Table 5.** Experimental conditions and run product information (continued).

				Calculate	Calculated melt volatile content <sup>d</sup>					Phase proportions <sup>e,t</sup> (wt. %)					
Charge ID	log(fO <sub>2</sub> ) <sup>a</sup> (bar)	$\Delta NNO^{b}$	$XH_2O_{in}{}^c \\$	$XH_2O_{fluid}$	H <sub>2</sub> O <sub>melt</sub> (wt. %)	CO <sub>2-melt</sub> (wt. ppm)	Орх	Срх	Plag	Mag	llm	OI	Amph	GI	Total
SUPL1-66	-9.7	1.4	0.73	0.6	3.8	583	-	-	-	-	-	-	-	-	-
SUPL1-67	-10.0	1.1	0.54	0.4	3.1	804	15.0	11.1	34.3	3.1				36.4	100
SUPL1-68	-10.9	0.2	0.28	0.2	1.7	1104	25.5	22.2	21.4	0.5				30.5	100
SUPL1-69	n.a.	n.a.	0.00	0.0	0.0	1266	42.6		41.4		0.6			15.4	100

466 Note. <sup>*a*</sup>log(fO<sub>2</sub>) determined using water saturated Ni-Pd alloy capsules, calculated using the formulation by Pownceby and O'Neill

467 (1994). For undersaturated charges, we calculated the  $fO_2$  using the formula outlined in section 3.4. <sup>b</sup> $\Delta NNO = log(fO_2) - log(fO_2)$  at

468 *Ni-NiO buffer*.  ${}^{c}XH_{2}Oin = H_{2}O/(H_{2}O + CO_{2})$  loaded in capsules in moles. <sup>d</sup>Water and CO<sub>2</sub> content in the melt were calculated from

469 mass balance combined with the mixed  $H_2O$ - $CO_2$  solubility model of Shishkina et al. (2010) for  $H_2O$  and Dixon et al. (2008) for  $CO_2$ .

470 <sup>e</sup>Phase proportions were calculated using grayscale thresholding of BSE charge maps using ImageJ software and then converted to

471 phase proportions. <sup>f</sup>See Table 2 for meaning of abbreviations, with the addition of Gl that we refer to glasses only. For charges where

472 only  $CO_2$  was added, we could not calculate the log ( $fO_2$ ).



- 475 *Figure 3.* Dissolved water content versus crystal content of experimental charges for *a.* 200-400
- 476 *MPa*, **b**. 100-200 *MPa* at NNO, and **c**. 100-200 *MPa* at NNO + 2. Dashed lines denote
- 477 extrapolated crystal contents of experimental charges. Grey, red and blue semi-transparent
- 478 boxes represent the field bounded by the phenocryst content of natural samples, water content in
- 479 the melt for the given temperature with SEE errors (975  $\pm$  39°C) for the 1990 pumices
- 480 (explosive), 2007 dome fragments (effusive), and 2014 pumices (explosive). Dashed arrows
- 481 indicate possible changes of temperature and water in the melt during ascent for the 2007
- 482 magma. The crystal contents were obtained by Utami et al. (in revision) and this study (Table 1).
- 483 Average temperature estimates from each eruption are calculated from mineral-melt
- 484 geothermobarometry listed in Table 3.



486 *Figure 4 Phase relations for a. 200-400 MPa, b. 100-200 MPa at NNO, and c. 100-200 MPa at* 

487 *NNO* + 2. *Grey, red and blue semi-transparent boxes represent the same parameters as Fig. 3.* 

488 Dashed lines denote inferred phase stability fields at the given temperature and water content.

489 Dashed arrows indicate potential degassing pathways. The phenocryst contents were estimated

490 from Utami et al. (in revision) and this study (Table 1). Mineral name abbreviations are the

491 same as in Fig 1, with the addition of Liq for liquid and Ilm for ilmenite. Pressure specific

492 stability are specified next to the boundary line. Red line with 'liq' represents the liquidus

493 *boundary at the given pressure and temperature.* 

495 4.4 Mineral and glass compositions

496 4.4.1 Glass

497 Experimental glass compositions vary with water content and temperature (Fig 5a-h), 498 with compositions ranging from basaltic andesite to dacite as temperature decrease from 1060 to 499 950°C (Table 6). The most evolved glass that we were able to analyze has 67 wt. % SiO<sub>2</sub> is at 500 200 MPa, 1060°C, NNO at 1 wt. % water, with 50-54 wt. % crystals in the charge (Fig. 5a). The 501 glass we analyzed from the rest of the charges range from basaltic andesite to andesite. For the 502 same temperature, pressure, and  $fO_2$ , oxides such as SiO<sub>2</sub>, K<sub>2</sub>O, and CaO increases as water 503 content in the melt decreases, indicating the melt is becoming more evolved (Fig. 5a, c, e). 504 Changing the  $fO_2$  from NNO to NNO + 2 does not significantly affect the SiO<sub>2</sub> content of the 505 melt for the same pressure, temperature, and dissolved water content. The FeO\* (3.5-9 wt. %), 506 MgO (0.6-3.9 wt. %), and CaO (5-9.5 wt. %) of experimental glasses (Fig 5d-f) are controlled by 507 crystallization of ferromagnesian minerals such as pyroxenes and magnetite. The limited number 508 of natural and experimental glasses that we were able to analyze preclude direct and detailed 509 comparisons that would allow to establish the pre-eruptive conditions. The lack of agreement 510 between the matrix glass compositions and experimental glasses could be due to the effect of 511 syn-eruptive crystallization. These typically manifests as Plag microlites in natural rocks. We 512 observe microlites in one 1990 pumice and the 2007 dome, which would explain why the natural 513 rocks have a higher SiO<sub>2</sub> content than the experimental glasses.

514

#### 515 **Table 6**. Experimental glass chemistry.

T (°C)	1000 SUDI	1000	1000	1000	1000	1000	1000	1000	1000	1000	950
Charge <sup>a</sup>	1-1	1-2	50PL1- 6	SUPL1-7	50PL1- 8	12	13	16	17	19	21
n:	1	1	4	2	3	1	1	4	2	1	1
wt. %											
SiO <sub>2</sub>	52.8	56.1	57.4 (0.6)	60.1 (0.6)	58 (2)	56.5	59.6	56.7 (0.7)	56.1 (0.7)	55.8	61.2
TiO <sub>2</sub>	0.5	0.4	0.7 (0.1)	0.9 (0.2)	0.8 (0.2)	0.7	0.8	0.6 (0.1)	0.9 (0.3)	0.5	0.7
$AI_2O_3$	20.8	23.4	19 (2)	16.40 (0.01)	20 (3)	20.5	17.0	22 (1)	18.8 (0.7)	23.0	17.1
FeO*	6.3	4.7	6.9 (0.5)	8.3 (0.2)	7 (2)	7.4	8.6	6.3 (0.5)	9.2 (0.5)	5.2	7.8
MnO	0.1	0.1	0.17 (0.01)	0.20 (0.01)	0.17 (0.03)	0.2	0.2	0.13 (0.02)	0.18 (0.01)	0.1	0.3
MgO	1.9	1.6	2.8 (0.3)	2.7 (0.1)	2.0 (0.5)	2.1	2.2	1.4 (0.2)	3.1 (0.2)	1.2	2.3

CaO	9.0	9.7	8.6 (0.9)	6.69 (0.01)	8 (2)	8.7	6.5	8.7 (0.7)	7.6 (0.3)	9.6	6.3
Na₂O	3.0	3.3	3.2 (0.1)	3.1 (0.1)	3.37 (0.09)	3.0	3.7	3.4 (0.1)	3.2 (0.03)	3.5	3.1
K₂O	0.6	0.7	0.8 (0.1)	1.22 (0.01)	0.9 (0.2)	0.8	1.1	0.9 (0.1)	0.80 (0.04)	0.7	0.2
P <sub>2</sub> O <sub>5</sub>	0.1	0.2	0.17 (0.02)	0.27 (0.03)	0.19 (0.06)	0.1	0.2	0.17 (0.03)	0.17 (0.02)	0.2	1.1
Total <sup>b</sup>	95.0	96.2	89.8 (0.3)	87.2 (1.0)	90.2 (0.8)	95.3	94.0	95.2 (1.0)	91.9 (0.8)	93.7	85.8
TAS content	3.7	4.0	4.0	4.3	4.3	3.8	4.9	4.3	4.0	4.2	4.9
H₂O content <sup>c</sup>	3.5	3	3.8	3.2	3.1	4.4	3.9	8.3	6.5	3.8	4.3
T (°C)	1060	1060	1060	1060	1060	1060	1060	1060	1060	1060	1060
Charge <sup>a</sup>	SUPL 1-37	SUPL 1-38	SUPL1- 39	SUPL1- 40	SUPL1- 41	SUPL1- 43	SUPL1- 44	SUPL1- 45	SUPL1- 46	SUPL1- 48	SUPL1- 49
n:	3	5	4	4	1	5	2	3	1	4	4
wt. %											
SiO	56.4 (0.04)	56.5 (0.6)	55.9 (0.9)	56.7 (0.4)	59.6	55.0 (0.6)	60 (2)	57.8 (0.3)	66.6	56.0 (0.4)	57.6 (0.2)
T:O	0.66	0.7	0.6	0.8 (0.1)	1.0	0.7	1.0 (0.1)	0.80	1.7	0.6	0.70
	(0.06) 18.5	(0.1) 19 (2)	(0.1)	19 (2)	16.7	(0.1) 18.8	18 (3)	(0.03) 21.9	14 9	(0.1) 19.2	(0.02) 17.5
$AI_2O_3$	(0.3) 7.4	6.7	20 (2) 5.9	10 (2)		(0.1) 8.5	10 (0)	(0.1) 4.6	14.0	(0.03) 7.4	(0.1) 7.5
FeO*	(0.2)	(0.8)	(0.9)	6.2 (0.4)	7.1	(0.3)	8 (1)	(0.01)	3.5	(0.2)	(0.1)
MnO	(0.03)	(0.03)	(0.03)	(0.03)	0.3	(0.03)	0.2 (0.1)	(0.01)	0.1	(0.05)	(0.01)
MgO	4.2 (0.1)	3.88 (0.04)	3.6 (0.5)	3.5 (0.3)	3.4	4.0 (0.1)	1.6 (0.3)	0.68 (0.04)	2.0	3.8 (0.1)	4.1 (0.02)
CaO	8.6 (0.2)	8.9 (0.8)	9.5 (0.9)	9.4 (0.7)	7.7	9.03 (0.03)	7 (2)	9.1 (0.1)	5.4	9.1 (0.1)	8.2 (0.1)
Na <sub>2</sub> O	3.1 (0.1)	3.1 (0.1)	3.1 (0.1)	3.0 (0.1)	3.0	3.0	3.6 (0.2)	3.6 (0.1)	3.7	2.8 (0.05)	3.1 (0.04)
K 0	0.75	0.8	0.6	0.8 (0.1)	1.0	0.69	1.2 (0.2)	1.00	1.9	0.6	0.8
K <sub>2</sub> O	(0.02) 0.15	(0.1) 0.15	(0.1) 0.14	0.14	0.0	(0.01) 0.13		(0.02) 0.21	0.2	(0.04) 0.1	(0.01) 0.2
$P_2O_5$	(0.02)	(0.01)	(0.04)	(0.03)	0.2	(0.01)	0.2 (0.1)	(0.03)	0.3	(0.02)	(0.01)
Total <sup>2</sup>	(0.2)	94 (1)	(0.06)	93 (1)	93.6	(0.6)	(0.3)	(0.3)	94.8	(0.4)	(0.4)
TAS											
content	3.8	3.8	3.7	3.8	4.0	3.7	4.9	4.6	5.6	3.4	3.9
H <sub>2</sub> O content <sup>3</sup>	3.6	3.0	2.8	2.6	1.7	5.3	3.6	2.8	1.4	8.6	3.0
T (°C)	1000	1000	1000	1000	1000	1000	1000				
Charge <sup>a</sup>	SUPL 1-52	SUPL 1-53	SUPL1- 54	SUPL1- 55	SUPL1- 58	SUPL1- 59	SUPL1- 60				
n:	1	1	2	1	5	1	3				
wt. %											
SiO <sub>2</sub>	54.8	56.2	56.0 (0.4)	59.6	56.7 (0.2)	57.8	59.5 (0.09)				
TiO <sub>2</sub>	0.3	0.5	0.70 (0.02)	0.6	0.68 (0.05)	16.8	1.0 (0.04)				
$AI_2O_3$	25.5	22.1	21.8 (0.1)	18.9	18.3 (0.6)	7.3	15.99 (0.09)				
FeO*	3.2	5.7	5.8 (0.4)	6.6	8.1 (0.2)	3.2	9.1 (0.2)				
MnO	0.1	0.1	0.14	0.2	(0.20)	3.2	0.23				
			(3.02)		(0.02)		(3.01)				

MgO	1.0	1.5	1.5 (0.1)	1.4	3.5 (0.1)	0.8	2.64 (0.08)
CaO	10.7	9.6	9.6 (0.1)	7.7	8.5 (0.3)	9.5	6.79 (0.07)
Na₂O	3.7	3.2	3.5 (0.1)	3.5	3.0 (0.1)	0.3	3.27 (0.03)
K₂O	0.5	0.8	0.73 (0.02)	1.2	0.74 (0.04)	0.2	1.17 (0.02)
$P_2O_5$	0.1	0.2	0.21 (0.02)	0.3	0.16 (0.03)	1.0	0.22 (0.02)
Total <sup>a</sup>	93.9	96.6	96.7	98.0	94.5 (0.4)	94.8	94.7 (0.4)
TAS content	4.2	4.0	4.2	5.2	3.8	4.0	4.4
H₂O content <sup>°</sup>	3.5	3.0	2.9	1.8	4.5	3.9	3.5

518 Notes. <sup>a</sup>See Table 5. <sup>b</sup>Original total in wt. %. <sup>c</sup>H<sub>2</sub>O calculated by mass balance and mixed H2O-

519 CO2 solubility model of Shishkina et al. (2010) and Dixon et al. (2008). FeO\* = total Fe

520 expressed as  $Fe^{2+}$ . Numbers in parentheses refer to 1s of the mean of multiple analyses. Where

521 the 1s is less than the relative error, the relative error is reported instead. TAS = Total Alkali-

522 *Silica content.* 





Figure 5 Major element oxide compositions of experimental glasses (in wt. %) vs. dissolved
water content in the melt for a. SiO<sub>2</sub>, b. Al<sub>2</sub>O<sub>3</sub>, c. CaO, d. MgO, e. K<sub>2</sub>O, f. FeO\*. Solid coloured
boxes denote the range of whole rock and glass compositions reported in Utami et al. (in
revision) and this study for all three eruptions. The glass composition for the 2007 dome sample
was estimated by mass balance calculations (Albarede, 1995). The dashed blue square denotes
the pre-eruptive water content in the melt.

### 531 4.4.2 Plagioclase

532 It was difficult to obtain "pure" analyses of experimental plagioclase due to its elongated 533 shape (e.g. very thin in one direction) and the presence of various mineral inclusions, so we 534 report the recalculated analyses after subtraction of the effect of mineral inclusions (see 535 methodology section). We first filtered the experimental Plag analyses for their FeO\* and TiO<sub>2</sub> 536 contents to remove Mag inclusions, but the MgO remained higher than expected for a 537 plagioclase (~1 wt. %). We thus suspect that there may be some analysis that are mixtures of 538 Plag and Opx, which we have not attempt to correct further. However, this should not affect the 539 relative Ca-Na-K concentrations and thus determination of the stoichiometric Plag end-members. 540 Experimental plagioclase ranges in composition from about An<sub>57</sub> to An<sub>71</sub> (Table 7; Fig 6a), and 541 varies according to the water content in the melt and to a lesser extent, by temperature. For the 542 same T, P and  $fO_2$ , we find that water content positively correlates with An content. We have not 543 been able to analyze experimental Plag for near liquidus charges, nor from those with very high 544 crystal content. Some of the experimental Plag compositions overlap with natural Plag microlites 545 and rims from the 1990, 2007, and 2014 eruptions as described in Utami et al. (in revision) and 546 in this study with  $An_{59.71}$  (Fig. 6a-c; Table S1). Specifically, microlites and phenocryst rims from 547 the 1990-2014 eruptions (An from 65 to 68) overlap with experimental Plag crystallized at 200 548 MPa, 1000°C and  $\Delta$ NNO with ~4 wt. % water. No experimental Plag composition that we 549 analyzed have An content > 71. However, we find that Plag cores and some oscillatory growth 550 zones from all three eruptions with An content of > 71 in this study, Utami et al. (in revision), 551 and in Cassidy et al. (2016) (See Fig. 1 and Fig. S3 of Supplementary Data). Thermodynamic 552 modelling with rhyolite-MELTS at 975°C and 4 wt. % water shows that Plag with An content of 553 85-90 can be produced within the range of experimental temperatures (962-1062 $^{\circ}$ C). 554

555 **Table 7:** *Experimental plagioclase chemistry.* 

T (%C)	1000	1000	1000	050	050	1000	1000	1000	1000	1000	1000	050	050	050	050
T (°C)	SUPI 1-	SUPI 1-	SUPI 1-	950 SUPI 1-	950 SUPI 1	SUPI 1-	SUPI 1-	SUPI 1-	SUPI 1-	SUPI 1-	SUPI 1-	900 SUPI 1-	950 SUPI 1	900 SUPI 1-	950 SUPI 1
Charge	11N	12N	13N	28N	-30	44N	45N	58N	59N	60N	61N	63	-64	65N	-66
n	2	1	2	1	3	1	1	1	2	3	2	3	3	2	1
wt. %															
SiO <sub>2</sub>	57.9	59.0	57.5	59.9	54.8 (0.1)	59.2	58.7	58.4	59.9	57.9	62.0	58 (1)	53 (1)	58.6	55.7
TiO <sub>2</sub>					0.2 (0.1)	0.2					0.01	0.2 (0.1)	0.06 (0.02)		0.1
Al <sub>2</sub> O <sub>3</sub>	25.6	24.1	26.3	23.1	26.4 (0.6)	23.5	25.6	24.6	23.0	25.9	22.4	26 (1)	29.7 (0.6)	24.3	27.3
FeO*	0.2	0.6	0.1	1.1	1.9 (1)	1.1	0.2	0.8	1.1	0.03	0.6	1.4 (0.5)	1.1 (0.1)	1.2	1.1
MnO	0.1	0.2	0.0	0.2	0.05 (0.01)	0.1	0.1	0.1	0.1	0.05	0.1	0.03 (0.02)	0.02 (0.01)	0.1	0.02
MgO	1.4	1.4	0.9	3.3	0.6 (0.2)	1.1	0.5	1.6	1.6	0.8	1.2	0.3 (0.1)	0.14 (0.3)	1.4	0.2
CaO	11.1	10.5	11.4	9.5	10.7 (0.4)	10.5	11.0	11.0	10.2	11.4	8.7	10 (1)	13.2 (0.7)	10.3	11.1
Na₂O	3.3	3.6	3.7	3.6	3.6 (0.1)	3.5	3.9	3.5	3.8	3.9	3.8	3.5 (0.3)	3.09 (0.03)	3.6	3.5
K₂O	0.5	0.7	0.5	0.4	0.3 (0.1)	0.9	0.7	0.7	0.8	0.6	1.2	0.5 (0.2)	0.14 (0.04)	0.5	0.4
Total <sup>ª</sup>	97.5 (0.5)	97.9	98.4 (0.1)	98.3	98.4 (0.5)	97.1	98.0	98.7	98 (1)	98.2 (0.5)	98.4 (0.6)	99.9 (1)	100.5 (0.4)	99.2 (0.6)	99.4
Plag end- member															
An	63.9 (0.5)	58.8	61 (2)	58	61 (2)	60.4	58.5	61.0	57 (1)	59.3 (0.4)	53 (5)	59 (2)	70 (1)	59 (2)	62.0
Ab	32.6 (0.4)	36.6	36 (1)	39	37 (1)	34.2	36.8	34.9	37.8 (0.5)	37 (1)	39 (3)	38 (1)	30 (1)	37.0 (0.8)	35.4
Or	3.5 (0.1)	4.5	4 (1)	3	2 (0.4)	5.4	4.7	4.1	5.3 (0.1)	4 (1)	8 (2)	3 (1)	0.8 (0.3)	3.6 (0.9)	2.6

557

Notes.  $FeO^* = total Fe expressed as Fe^{2+}$ . Numbers in parentheses refer to 1s of the mean of multiple analyses. n = calculated and normalized plagioclase composition after subtraction of Fe-Ti oxide composition. <sup>a</sup>Original total oxide in wt. %. See Table 3 for endmember abbreviations and calculations.

561

# 564 4.4.3 Orthopyroxene

565	Experimental Opx composition mainly varies with temperature and water content in the
566	melt (Mg # 57-77; Fig 6b, Table 8). The Wo component of some of the experimental Opx is high
567	(Wo <sub>6-11</sub> ), and is classified as pigeonite according to Deer et al. (1992). For the same $P$ and $fO_2$ , a
568	temperature increase from 950°C to 1000°C results in an increase in the Mg # of about 58 to 70;
569	a decrease in water content in the melt also results in an increase in Mg #. A strong influence of
570	water content in the Opx-melt Mg # partitioning was reported in previous experimental studies
571	(Waters & Lange, 2017). The 1990, 2007, and 2014 Opx cores and glomerocrysts range from
572	Mg # 66 to 76 (average of 66), with a bimodal distribution. This Mg # range overlaps with
573	experimental orthopyroxene compositions at 1000°C, 200 MPa and at NNO for all eruptions,
574	although we have not identified pigeonite in the natural samples.
575	
576	4.4.4 Clinopyroxene
577	Experimental Cpx is ubiquitous in most charges (Mg # 61-76, Wo33-40En39-46Fs15-25) (Fig
578	6c, Table 8). The Mg # of experimental Cpx increases from 61 to 76 with increasing
579	temperature. There appears to be some effect of water content on composition: at 950°C, the
580	experimental Cpx at 200 MPa have higher Mg # (68-69) than at 400 MPa (61-63). As we could
581	not obtain good quantitative analysis of Cpx for all charges, we have no clear indication of the
582	effect of fO <sub>2</sub> on clinopyroxene composition. The 1990, 2007, and 2014 clinopyroxene cores and
583	glomerocrysts composition Mg # (70-76, average ~ 74) overlap with experimental clinopyroxene
584	at 1000°C, 200 MPa, and NNO + 2 at 4 wt. % water.
585	

## **Table 8.** *Experimental pyroxene chemistry*

Manual	Dat	Onv	Onv	Dat	Onv	Dat	00%	Dat	Dat	Onv	Onv	Onv
Mineral	1000	0px	0px	Pgi 1000	0px 1000	1000	0px 1000	Pgi 1000	Pgi 1000	0px	0px	050
T (°C)	SUPI	SUPI	SUPI	SUPI	SUPL1	SUPI	SUPL1	SUPI	SUPI	SUPI	SUPI	SUP
Charge	1-02	1-11	1-12	1-12	-13	1-13	-16	1-16	1-17	1-18	1-19	1-31
n:	3	6	14	1	4	1	5	1	1	1	3	3
wt. %												
0.0	52.0	52.6	52.5	52.1	54.0	52.7	51.0	49.8	51.5	50.1	51.1	51.0
SiO <sub>2</sub>	(1.1)	(0.8)	(0.5)	02.1	(1.0)	02	(0.8)		0110		(0.6)	(0.3)
TiO <sub>2</sub>	(0.02)	(0.04)	(0.04)	0.3	(0.03)	0.2	(0.07)	0.3	0.2	0.3	(0.07)	(0.01)
-	`1.6´	<b>`</b> 2.3 <sup>´</sup>	2.0	2.6	2.7 <sup>′</sup>	07	4 (1)	6.0	12	2.6	`2.1 <i>´</i>	<b>2.6</b>
$AI_2O_3$	(0.5)	(0.9)	(0.3)	2.0	(0.6)	0.1	0.40	0.0		2.0	(0.8)	(0.2)
Cr <sub>2</sub> O <sub>2</sub>	(0.03)	(0.03)	(0.05)	0.1	(0.1)	0.03	(0.03)	0.1	0.05	b.d.	(0.04)	(0.03)
	20.9	18.9	20.0	18.2	14.1	22.3	20.9	20.2	22.0	24.8	22.2	24.6
FeO*	(0.4)	(0.7)	(0.6)	10.2	(0.1)	22.5	(0.6)	20.2	22.0	24.0	(0.3)	(0.2)
MnO	0.7	0.58	0.63	0.6	0.47	0.7	0.58	0.6	0.7	0.6	0.64	0.65
Nin O	18.6	22.7	21.9	21.0	25.7	10.4	19.4	10 7	10 /	10.5	19.1	18.1
MgO	(0.5)	(0.7)	(0.3)	21.0	(0.6)	19.4	(0.6)	10.7	10.4	19.5	(0.6)	(0.4)
C20	5.4	2.2	2.0	3.9	2.3	3.8	2.6	3.1	4.4	2.2	3 (1)	1.8
CaO	0.2(0.2)	(0.5)	(0.2)		0.04	0.05	0.2			1. J	0.05	0.03
Na <sub>2</sub> O	)	D.d.	D.d.	0.1	(0.03)	0.05	(0.1)	0.4	0.1	D.d.	(0.02)	(0.01)
КO	0.05	0.02	b.d.	b.d.	b.d.	0.01	0.01	0.0	0.01	b.d.	b.d.	b.d.
R <sub>2</sub> U	(0.08) 99.7	(0.02) 99.6	99.5		100.0		(0.01) 99.1				98.5	99.1
Total	(1.4)	(1.2)	(0.5)	98.8	(0.6)	100.0	(0.6)	99.1	98.4	100.1	(0.3)	(0.6)
Ma #a	61.4	68 (2)	66.2	67.2	76.5	60.8	62.3	62.3	59.9	58.3	60.5	56.7
ivig #	(1.1)	( )	(0.9)		(0.5)		(0.7)				(0.5)	(0.7)
End-												
member												
ŭ	11 4		42		47							3.8
Wo	(0.3)	5 (1)	(0.4)	8.3	(0.5)	7.9	6 (1)	7.0	9.3	4.4	6 (3)	(0.2)
_	54.4	65 (2)	63.4	61.6	72.9	56.0	58.7	58.0	54.3	55 7	57 (2)	54.5
En	(0.8)	00 (2)	(0.9)	01.0	(0.8)	00.0	(0.9)	00.0	01.0	00.1	27.0	(0.8)
Fs	(1.1)	30 (1)	32.4									41.0
	· · ·	00 (1)	(0.9)	30.1	(0.4)	36.1	35.5	35.0	36.4	39.8	(0.6)	(0.6)
		00(1)	(0.9)	30.1	(0.4)	36.1	35.5 (0.8)	35.0	36.4	39.8	(0.6)	(0.6)
Mineral	Орх	Opx	(0.9) Opx	30.1 Pgt	(0.4)	36.1 Opx	(0.8)	35.0 Opx	36.4 Opx	39.8 Opx	(0.6)	(0.6) Opx
Mineral T (°C)	Opx 950	Opx 950	(0.9) Opx 950	30.1 Pgt 1060	(0.4) Pgt 1060	36.1 Opx 1060	35.5 (0.8) Opx 1060	35.0 Opx 1060	36.4 Opx 1060	39.8 Opx 1000	(0.6) Opx 1000	(0.6) Opx 1000
Mineral T (°C)	Opx 950 SUPL	Opx 950 SUPL	(0.9) Opx 950 SUPL	30.1 Pgt 1060 SUPL	(0.4) Pgt 1060 SUPL1	36.1 Opx 1060 SUPL	35.5 (0.8) Opx 1060 SUPL1	35.0 Opx 1060 SUPL	36.4 Opx 1060 SUPL	39.8 Opx 1000 SUPL	0.6) Opx 1000 SUPL	(0.6) Opx 1000 SUPL
Mineral T (°C) Charge	Opx 950 SUPL 1-32	Opx 950 SUPL 1-33	(0.9) Opx 950 SUPL 1-34 7	30.1 Pgt 1060 SUPL 1-44	(0.4) Pgt 1060 SUPL1 -45	36.1 Opx 1060 SUPL 1-46	35.5 (0.8) Opx 1060 SUPL1 -46 1	35.0 Opx 1060 SUPL 1-49	36.4 Opx 1060 SUPL 1-50	39.8 Opx 1000 SUPL 1-51	0px 1000 SUPL 1-57	(0.6) Opx 1000 SUPL 1-59
Mineral T (°C) Charge n:	Opx 950 SUPL 1-32 10	Opx 950 SUPL 1-33 6	(0.9) Opx 950 SUPL 1-34 7	30.1 Pgt 1060 SUPL 1-44 1	(0.4) Pgt 1060 SUPL1 -45 1	36.1 Opx 1060 SUPL 1-46 1	35.5 (0.8) Opx 1060 SUPL1 -46 1	35.0 Opx 1060 SUPL 1-49 9	36.4 Opx 1060 SUPL 1-50 1	39.8 Opx 1000 SUPL 1-51 26	0,06) Opx 1000 SUPL 1-57 1	(0.6) Opx 1000 SUPL 1-59 3
Mineral T (°C) Charge n: wt. %	Opx 950 SUPL 1-32 10	Opx 950 SUPL 1-33 6	(0.9) Opx 950 SUPL 1-34 7	30.1 Pgt 1060 SUPL 1-44 1	(0.4) Pgt 1060 SUPL1 -45 1	36.1 Opx 1060 SUPL 1-46 1	35.5 (0.8) Opx 1060 SUPL1 -46 1	35.0 Opx 1060 SUPL 1-49 9	36.4 Opx 1060 SUPL 1-50 1	39.8 Opx 1000 SUPL 1-51 26 53.0	0.6) Opx 1000 SUPL 1-57 1	(0.6) Opx 1000 SUPL 1-59 3
Mineral T (°C) Charge n: wt. % SiO <sub>2</sub>	Opx 950 SUPL 1-32 10 51.3 (0.6)	Opx 950 SUPL 1-33 6 51.5 (0.4)	(0.9) Opx 950 SUPL 1-34 7 51.8 (0.2)	30.1 Pgt 1060 SUPL 1-44 1 52.9	22.4 (0.4) Pgt 1060 SUPL1 -45 1 53.7	36.1 Opx 1060 SUPL 1-46 1 53.7	35.5 (0.8) Opx 1060 SUPL1 -46 1 53.9	35.0 Opx 1060 SUPL 1-49 9 52.7 (1.0)	36.4 Opx 1060 SUPL 1-50 1 53.1	39.8 Opx 1000 SUPL 1-51 26 53.0 (0.9)	0,0,6) Opx 1000 SUPL 1-57 1 52.0	(0.6) Opx 1000 SUPL 1-59 3 53.1 (0.9)
Mineral T (°C) Charge n: wt. % SiO <sub>2</sub>	Opx 950 SUPL 1-32 10 51.3 (0.6) 0.36	Opx 950 SUPL 1-33 6 51.5 (0.4) 0.37	(0.9) Opx 950 SUPL 1-34 7 51.8 (0.2) 0.38	30.1 Pgt 1060 SUPL 1-44 1 52.9 0.3	22.4 (0.4) Pgt 1060 SUPL1 -45 1 53.7 0.1	36.1 Opx 1060 SUPL 1-46 1 53.7 0.1	35.5 (0.8) Opx 1060 SUPL1 -46 1 53.9 0.2	35.0 Opx 1060 SUPL 1-49 9 52.7 (1.0) 0.20	36.4 Opx 1060 SUPL 1-50 1 53.1 0.3	39.8 Opx 1000 SUPL 1-51 26 53.0 (0.9) 0.23	0,000 0px 1000 SUPL 1-57 1 52.0 0,3	(0.6) Opx 1000 SUPL 1-59 3 53.1 (0.9) 0.23
Mineral T (°C) Charge n: wt. % SiO <sub>2</sub> TiO <sub>2</sub>	Opx 950 SUPL 1-32 10 51.3 (0.6) 0.36 (0.05) 2.4	Opx 950 SUPL 1-33 6 51.5 (0.4) 0.37 (0.03) 2 3	(0.9) Opx 950 SUPL 1-34 7 51.8 (0.2) 0.38 (0.04) 25	30.1 Pgt 1060 SUPL 1-44 1 52.9 0.3	22.4 (0.4) Pgt 1060 SUPL1 -45 1 53.7 0.1	36.1 Opx 1060 SUPL 1-46 1 53.7 0.1	35.5 (0.8) Opx 1060 SUPL1 -46 1 53.9 0.2	35.0 Opx 1060 SUPL 1-49 9 52.7 (1.0) 0.20 (0.02) 3.6	36.4 Opx 1060 SUPL 1-50 1 53.1 0.3	39.8 Opx 1000 SUPL 1-51 26 53.0 (0.9) 0.23 (0.04) 2.1	0,000 0px 1000 SUPL 1-57 1 52.0 0.3	(0.6) Opx 1000 SUPL 1-59 3 53.1 (0.9) 0.23 (0.05)
Mineral T (°C) Charge n: wt. % SiO <sub>2</sub> TiO <sub>2</sub> Al <sub>2</sub> O <sub>3</sub>	Opx 950 SUPL 1-32 10 51.3 (0.6) 0.36 (0.05) 2.4 (0.5)	Opx 950 SUPL 1-33 6 51.5 (0.4) 0.37 (0.03) 2.3 (0.4)	(0.9) Opx 950 SUPL 1-34 7 51.8 (0.2) 0.38 (0.04) 2.5 (0.5)	30.1 Pgt 1060 SUPL 1-44 1 52.9 0.3 2.4	22.4 (0.4) Pgt 1060 SUPL1 -45 1 53.7 0.1 1.1	36.1 Opx 1060 SUPL 1-46 1 53.7 0.1 1.1	35.5 (0.8) Opx 1060 SUPL1 -46 1 53.9 0.2 1.8	35.0 Opx 1060 SUPL 1-49 9 52.7 (1.0) 0.20 (0.02) 3.6 (0.7)	36.4 Opx 1060 SUPL 1-50 1 53.1 0.3 2.6	39.8 Opx 1000 SUPL 1-51 26 53.0 (0.9) 0.23 (0.04) 2.1 (0.7)	37.0 (0.6) Opx 1000 SUPL 1-57 1 52.0 0.3 2.4	(0.6) Opx 1000 SUPL 1-59 3 53.1 (0.9) 0.23 (0.05) 3 (1)
Mineral T (°C) Charge n: wt. % SiO <sub>2</sub> TiO <sub>2</sub> Al <sub>2</sub> O <sub>3</sub>	Opx 950 SUPL 1-32 10 51.3 (0.6) 0.36 (0.05) 2.4 (0.5) 0.11	Opx 950 SUPL 1-33 6 51.5 (0.4) 0.37 (0.03) 2.3 (0.4) 0.13	(0.9) Opx 950 SUPL 1-34 7 51.8 (0.2) 0.38 (0.04) 2.5 (0.5) 0.10	30.1 Pgt 1060 SUPL 1-44 1 52.9 0.3 2.4 0.0	Pgt 1060 SUPL1 -45 1 53.7 0.1 1.1	36.1 Opx 1060 SUPL 1-46 1 53.7 0.1 1.1	35.5 (0.8) Opx 1060 SUPL1 -46 1 53.9 0.2 1.8 0.2	35.0 Opx 1060 SUPL 1-49 9 52.7 (1.0) 0.20 (0.02) 3.6 (0.7) 0.4	36.4 Opx 1060 SUPL 1-50 1 53.1 0.3 2.6 0.5	39.8 Opx 1000 SUPL 1-51 26 53.0 (0.9) 0.23 (0.04) 2.1 (0.7) 0.04	37.0 (0.6) Opx 1000 SUPL 1-57 1 52.0 0.3 2.4 0.2	(0.6) Opx 1000 SUPL 1-59 3 53.1 (0.9) 0.23 (0.05) 3 (1) 0.29
Mineral T (°C) Charge n: wt. % SiO <sub>2</sub> TiO <sub>2</sub> Al <sub>2</sub> O <sub>3</sub> Cr <sub>2</sub> O <sub>3</sub>	Opx 950 SUPL 1-32 10 51.3 (0.6) 0.36 (0.05) 2.4 (0.5) 0.11 (0.04) 22 9	Opx 950 SUPL 1-33 6 51.5 (0.4) 0.37 (0.03) 2.3 (0.4) 0.13 (0.01) (0.01)	(0.9) Opx 950 SUPL 1-34 7 51.8 (0.2) 0.38 (0.04) 2.5 (0.5) 0.10 (0.03) 22.4	30.1 Pgt 1060 SUPL 1-44 1 52.9 0.3 2.4 0.0	Pgt 1060 SUPL1 -45 1 53.7 0.1 1.1 0.2	36.1 Opx 1060 SUPL 1-46 1 53.7 0.1 1.1 0.2	35.5 (0.8) Opx 1060 SUPL1 -46 1 53.9 0.2 1.8 0.2	35.0 Opx 1060 SUPL 1-49 9 52.7 (1.0) 0.20 (0.02) 3.6 (0.7) 0.4 (0.4)	36.4 Opx 1060 SUPL 1-50 1 53.1 0.3 2.6 0.5	39.8 Opx 1000 SUPL 1-51 26 53.0 (0.9) 0.23 (0.04) 2.1 (0.7) 0.04 (0.03) 47.2	37.0         (0.6)         Opx         1000         SUPL         1-57         1         52.0         0.3         2.4         0.2	(0.6) Opx 1000 SUPL 1-59 3 53.1 (0.9) 0.23 (0.05) 3 (1) 0.29 (0.07) (0.07) 47.2
Mineral T (°C) Charge n: wt. % SiO <sub>2</sub> TiO <sub>2</sub> Al <sub>2</sub> O <sub>3</sub> $Cr_2O_3$ FeO*	Opx 950 SUPL 1-32 10 51.3 (0.6) 0.36 (0.05) 2.4 (0.5) 0.11 (0.04) 23.8 (0.2)	Opx 950 SUPL 1-33 6 51.5 (0.4) 0.37 (0.03) 2.3 (0.4) 0.13 (0.01) 24.0 (0.2)	(0.9) Opx 950 SUPL 1-34 7 51.8 (0.2) 0.38 (0.04) 2.5 (0.5) 0.10 (0.03) 23.4 (0.3)	30.1 Pgt 1060 SUPL 1-44 1 52.9 0.3 2.4 0.0 22.0	22.4 (0.4) Pgt 1060 SUPL1 -45 1 53.7 0.1 1.1 0.2 18.9	36.1 Opx 1060 SUPL 1-46 1 53.7 0.1 1.1 0.2 18.9	35.5 (0.8) Opx 1060 SUPL1 -46 1 53.9 0.2 1.8 0.2 1.8 0.2 19.1	35.0 Opx 1060 SUPL 1-49 9 52.7 (1.0) 0.20 (0.02) 3.6 (0.7) 0.4 (0.4) 14.7 (0.8)	36.4 Opx 1060 SUPL 1-50 1 53.1 0.3 2.6 0.5 17.9	39.8 Opx 1000 SUPL 1-51 26 53.0 (0.9) 0.23 (0.04) 2.1 (0.7) 0.04 (0.03) 17.3 (0.5)	37.0 (0.6) Opx 1000 SUPL 1-57 1 52.0 0.3 2.4 0.2 19.5	(0.6) Opx 1000 SUPL 1-59 3 53.1 (0.9) 0.23 (0.05) 3 (1) 0.29 (0.07) 17.3 (0.2)
Mineral T (°C) Charge n: wt. % SiO <sub>2</sub> TiO <sub>2</sub> Al <sub>2</sub> O <sub>3</sub> $Cr_2O_3$ FeO*	Opx 950 SUPL 1-32 10 51.3 (0.6) 0.36 (0.05) 2.4 (0.5) 0.11 (0.04) 23.8 (0.2) 0.65	Opx 950 SUPL 1-33 6 51.5 (0.4) 0.37 (0.03) 2.3 (0.4) 0.13 (0.01) 24.0 (0.2) 0.66	(0.9) Opx 950 SUPL 1-34 7 51.8 (0.2) 0.38 (0.04) 2.5 (0.5) 0.10 (0.03) 23.4 (0.3) 0.67	30.1 Pgt 1060 SUPL 1-44 1 52.9 0.3 2.4 0.0 22.0	22.4 (0.4) Pgt 1060 SUPL1 -45 1 53.7 0.1 1.1 0.2 18.9 0.6	36.1 Opx 1060 SUPL 1-46 1 53.7 0.1 1.1 0.2 18.9 0.6	35.5 (0.8) Opx 1060 SUPL1 -46 1 53.9 0.2 1.8 0.2 1.8 0.2 19.1	35.0 Opx 1060 SUPL 1-49 9 52.7 (1.0) 0.20 (0.02) 3.6 (0.7) 0.4 (0.4) 14.7 (0.8) 0.46	36.4 Opx 1060 SUPL 1-50 1 53.1 0.3 2.6 0.5 17.9 0.6	39.8 Opx 1000 SUPL 1-51 26 53.0 (0.9) 0.23 (0.04) 2.1 (0.7) 0.04 (0.03) 17.3 (0.5) 0.58	37.0 (0.6) Opx 1000 SUPL 1-57 1 52.0 0.3 2.4 0.2 19.5 0.6	(0.6) Opx 1000 SUPL 1-59 3 53.1 (0.9) 0.23 (0.05) 3 (1) 0.29 (0.07) 17.3 (0.2) 0.55
Mineral T (°C) Charge n: wt. % SiO <sub>2</sub> TiO <sub>2</sub> Al <sub>2</sub> O <sub>3</sub> $Cr_2O_3$ FeO* MnO	Opx 950 SUPL 1-32 10 51.3 (0.6) 0.36 (0.05) 2.4 (0.5) 0.11 (0.04) 23.8 (0.2) 0.65 (0.02)	Opx 950 SUPL 1-33 6 51.5 (0.4) 0.37 (0.03) 2.3 (0.4) 0.13 (0.01) 24.0 (0.2) 0.66 (0.02)	(0.9) Opx 950 SUPL 1-34 7 51.8 (0.2) 0.38 (0.04) 2.5 (0.5) 0.10 (0.03) 23.4 (0.3) 0.67 (0.03) 0.03	30.1 Pgt 1060 SUPL 1-44 1 52.9 0.3 2.4 0.0 22.0 0.7	22.4 (0.4) Pgt 1060 SUPL1 -45 1 53.7 0.1 1.1 0.2 18.9 0.6	36.1 Opx 1060 SUPL 1-46 1 53.7 0.1 1.1 0.2 18.9 0.6	35.5 (0.8) Opx 1060 SUPL1 -46 1 53.9 0.2 1.8 0.2 19.1 0.7	35.0 Opx 1060 SUPL 1-49 9 52.7 (1.0) 0.20 (0.02) 3.6 (0.7) 0.4 (0.4) 14.7 (0.8) 0.46 (0.03) 0.46 (0.03)	36.4 Opx 1060 SUPL 1-50 1 53.1 0.3 2.6 0.5 17.9 0.6	39.8 Opx 1000 SUPL 1-51 26 53.0 (0.9) 0.23 (0.04) 2.1 (0.7) 0.04 (0.03) 17.3 (0.5) 0.58 (0.03)	37.0         (0.6)         Opx         1000         SUPL         1-57         1         52.0         0.3         2.4         0.2         19.5         0.6	(0.6) Opx 1000 SUPL 1-59 3 53.1 (0.9) 0.23 (0.05) 3 (1) 0.29 (0.07) 17.3 (0.2) 0.55 (0.05) 0.25 (0.05)
Mineral T (°C) Charge n: wt. % SiO <sub>2</sub> TiO <sub>2</sub> Al <sub>2</sub> O <sub>3</sub> $Cr_2O_3$ FeO* MnO	Opx 950 SUPL 1-32 10 51.3 (0.6) 0.36 (0.05) 2.4 (0.5) 0.11 (0.04) 23.8 (0.2) 0.65 (0.02) 18.6 (0.2)	Opx 950 SUPL 1-33 6 51.5 (0.4) 0.37 (0.03) 2.3 (0.4) 0.13 (0.01) 24.0 (0.2) 0.66 (0.02) 18.5 (0.2)	(0.9) Opx 950 SUPL 1-34 7 51.8 (0.2) 0.38 (0.04) 2.5 (0.5) 0.10 (0.03) 23.4 (0.3) 0.67 (0.03) 18.9 (0.4)	30.1 Pgt 1060 SUPL 1-44 1 52.9 0.3 2.4 0.0 22.0 0.7 19.9	22.4 (0.4) Pgt 1060 SUPL1 -45 1 53.7 0.1 1.1 0.2 18.9 0.6 22.9	36.1 Opx 1060 SUPL 1-46 1 53.7 0.1 1.1 0.2 18.9 0.6 22.9	35.5 (0.8) Opx 1060 SUPL1 -46 1 53.9 0.2 1.8 0.2 19.1 0.7 20.6	35.0 Opx 1060 SUPL 1-49 9 52.7 (1.0) 0.20 (0.02) 3.6 (0.7) 0.4 (0.4) 14.7 (0.8) 0.46 (0.03) 25.6 (0.4)	36.4 Opx 1060 SUPL 1-50 1 53.1 0.3 2.6 0.5 17.9 0.6 21.9	39.8 Opx 1000 SUPL 1-51 26 53.0 (0.9) 0.23 (0.04) 2.1 (0.7) 0.04 (0.03) 17.3 (0.5) 0.58 (0.03) 24.2 (0.9)	37.0         (0.6)         Opx         1000         SUPL         1-57         1         52.0         0.3         2.4         0.2         19.5         0.6         22.1	(0.6) Opx 1000 SUPL 1-59 3 53.1 (0.9) 0.23 (0.05) 3 (1) 0.29 (0.07) 17.3 (0.2) 0.55 (0.05) 23.5 (0.5)
Mineral T (°C) Charge n: wt. % SiO <sub>2</sub> TiO <sub>2</sub> Al <sub>2</sub> O <sub>3</sub> $Cr_2O_3$ FeO* MnO MgO	Opx 950 SUPL 1-32 10 51.3 (0.6) 0.36 (0.05) 2.4 (0.5) 0.11 (0.04) 23.8 (0.2) 0.65 (0.02) 18.6 (0.2) 1.9	Opx 950 SUPL 1-33 6 51.5 (0.4) 0.37 (0.03) 2.3 (0.4) 0.13 (0.01) 24.0 (0.2) 0.66 (0.02) 18.5 (0.2) 2.1	(0.9) Opx 950 SUPL 1-34 7 51.8 (0.2) 0.38 (0.04) 2.5 (0.5) 0.10 (0.03) 23.4 (0.3) 0.67 (0.03) 18.9 (0.4) 2.0	30.1 Pgt 1060 SUPL 1-44 1 52.9 0.3 2.4 0.0 22.0 0.7 19.9 2.0	22.4 (0.4) Pgt 1060 SUPL1 -45 1 53.7 0.1 1.1 0.2 18.9 0.6 22.9	36.1 Opx 1060 SUPL 1-46 1 53.7 0.1 1.1 0.2 18.9 0.6 22.9 2.2	35.5 (0.8) Opx 1060 SUPL1 -46 1 53.9 0.2 1.8 0.2 19.1 0.7 20.6	35.0 Opx 1060 SUPL 1-49 9 52.7 (1.0) 0.20 (0.02) 3.6 (0.7) 0.4 (0.4) 14.7 (0.8) 0.46 (0.03) 25.6 (0.4) 2.2	36.4 Opx 1060 SUPL 1-50 1 53.1 0.3 2.6 0.5 17.9 0.6 21.9 2.2	39.8 Opx 1000 SUPL 1-51 26 53.0 (0.9) 0.23 (0.04) 2.1 (0.7) 0.04 (0.03) 17.3 (0.5) 0.58 (0.03) 24.2 (0.9) 2.1	37.0         (0.6)         Opx         1000         SUPL         1-57         1         52.0         0.3         2.4         0.2         19.5         0.6         22.1         2.4	(0.6) Opx 1000 SUPL 1-59 3 53.1 (0.9) 0.23 (0.05) 3 (1) 0.29 (0.07) 17.3 (0.2) 0.55 (0.05) 23.5 (0.5) 2.1

Na₂O	0.03 (0.01)	0.06 (0.04)	0.06 (0.05)	0.3	0.1	0.1	0.2	b.d.	0.05	b.d.	0.01	0.04 (0.02)
K₂O	b.d.	0.01 (0.01)	0.01 (0.01)	0.03	0.01	0.01	0.01	b.d.	0.01	b.d.	0.03	0.01 (0.01)
Total	99.1 (0.3)	99.5 (0.5)	99.8 (0.4)	101.5	101.0	101.0	101.3	100.2 (0.6)	100.1	100 (1)	99.4	99.9 (0.3)
Mg # <sup>a</sup>	58.2 (0.3)	57.8 (0.2)	59.0 (0.3)	61.7	68.4	68.4	65.8	75.6 (1.0)	68.6	71.4 (0.9)	66.9	70.8 (0.3)
End- member ª												
Wo	4.0 (0.5)	4.5 (0.3)	4.3 (0.5)	6.1	6.5	6.5	9.5	4.5 (0.6)	6.9	4.3 (0.8)	4.4	4 (2)
En	55.8 (0.4)	55.2 (0.2)	56.5 (0.6)	57.9	63.9	63.9	59.5	72.1 (0.9)	63.9	68 (1)	64.0	68 (1)
Fs	40.1 (0.2)	40.3 (0.2)	39.2 (0.2)	36.0	29.6	29.6	31.0	23.3 (1.0)	29.3	27.4 (0.8)	31.6	28.0 (0.4)
Mineral	Орх	Орх	Pat	Срх	Срх	Срх	Срх	Срх	Срх	Срх	Орх	Орх
T (°C)	1000	950	950	1000	950	950	950	1000	1000	950	1060	1000
Charge	SUPL	SUPL	SUPL	SUPL	SUPL1	SUPL	SUPL1	SUPL	SUPL	SUPL	SUPL	SUPL
n:	1	7	1-23	3	-20	1	-54	3	3	1	1	26
wt. %												
SiO <sub>2</sub>	52.2	53.6 (0.7)	52.6	48.9 (0.6)	52.6 (0.9)	50.1	50.7 (0.6)	52 (1)	51.7 (0.6)	53.3	53.1	53.0 (0.9)
T:0	0.3	0.24	0.3	0.62	3.1	0.7	0.72	0.4	0.5	0.4	0.3	0.23
HO <sub>2</sub>	26	(0.05) 2.2	2.4	(0.09) 5.0	(0.6)	2.2	(0.06) 3.3	(0.1) 3.0	(0.1) 3.1	4.4	26	(0.04) 2.1
$AI_2O_3$	2.0	(0.5)	2.4	(0.3)	19 (2)	3.2	(0.2)	(0.9)	(0.3)	4.4	2.0	(0.7)
$Cr_2O_3$	0.3	(0.02)	0.01	(0.06)	b.d.	0.3	(0.01)	(0.1)	(0.1)	0.02	0.5	(0.04)
FeO*	19.4	18.1 (0.2)	17.7	10.1 (0.3)	14 (1)	14.3	13.1 (0.9)	9.2 (0.1)	10 (2)	10.8	17.9	17.3 (0.5)
MnO	0.6	0.70 (0.05)	0.7	0.40 (0.05)	0.25 (0.01)	0.5	0.4 (0.1)	0.4 (0.1)	0.4 (0.1)	0.5	0.6	0.58 (0.03)
MaQ	21.3	22.0 (0.4)	21.5	14.4	0.4	12.7	12.8	15.9	15 (2)	13.2	21.9	24.2
Nigo	2.7	3.1	5.0	18.3	(0.1)	16.5	(0.1)	18.4	18 (3)	17.6	3.3	2.1
CaO		(0.3) 0.06	0.0	(0.8) 0.28	0.255		0.24	(0.5) 0.19	0.20		0.0	(0.4)
Na <sub>2</sub> O	0.1	(0.01)	0.1	(0.02)	(0.01)	0.2	(0.03)	(0.02)	(0.04)	0.3	0.05	b.d.
K₂O	0.02	(0.02	0.01	0.03 (0.02)	(0.1)	0.01	(0.04)	0.03 (0.01)	b.d.	0.1	0.01	b.d.
Total	99.5	100.1 (0.5)	100.3	98.1 (0.2)	101.3 (0.3)	98.4	99.3 (0.8)	100.0 (0.6)	99.7 (0.5)	100.7	100.1	100 (1)
Mg # <sup>a</sup>	66.3	68.4 (0.4)	68.4	71.9 (0.9)	69 (1)	61.3	63.4 (0.7)	76 (1)	73 (1)	68.6	68.6	71.4 (0.9)
End- member ª												
Wo	5.8	6.5 (0.6)	10.3	40 (1)	39 (3)	36	39 (3)	39 (1)	38 (7)	39.6	6.9	4.3 (0.8)
En	62.4	64.0 (0.7)	61.4	43 (1)	42 (3)	39	39 (1)	46 (2)	45 (4)	41.5	63.9	68 (1)
Fs	31.8	29.5 (0.4)	28.3	17 (1)	19 (1)	25	22 (1)	15.0 (0.3)	17 (3)	19.0	29.3	27.4 (0.8)
Mineral <sup>1</sup>	Орх	Pat	Орх	Орх	Орх	Срх	Срх	Срх	Срх	Срх	Срх	Срх
T (°C)	1000	1000	1000	1000	950	1000	950	950	950	1000	1000	950
Charge	SUPL 1-57	SUPL 1-57	SUPL 1-59	SUPL 1-61	SUPL1 -64	SUPL 1-18	SUPL1 -28	SUPL 1-31	SUPL 1-34	SUPL 1-58	SUPL 1-59	SUPL 1-64
					~ '							

n:	1	1	3	1	7	3	4	1	5	3	3	1
wt. %												
SiO <sub>2</sub>	52.0	52.6	53.1 (0.9)	52.2	53.6 (0.7)	48.9 (0.6)	52.6 (0.9)	50.1	50.7 (0.6)	52 (1)	51.7 (0.6)	53.3
TiO <sub>2</sub>	0.3	0.3	0.23 (0.05)	0.3	0.24 (0.05)	0.62 (0.09)	3.1 (0.6)	0.7	0.72 (0.06)	0.4 (0.1)	0.5 (0.1)	0.4
Al <sub>2</sub> O <sub>3</sub>	2.4	1.5	3 (1)	2.6	2.2 (0.5)	5.0 (0.3)	19 (2)	3.2	3.3 (0.2)	3.0 (0.9)	3.1 (0.3)	4.4
$Cr_2O_3$	0.2	0.2	0.29 (0.07)	0.3	0.02 (0.01)	0.10 (0.06)	b.d.	0.3	0.22 (0.01)	0.4 (0.1)	0.5 (0.1)	0.02
FeO*	19.5	18.7	17.3 (0.2)	19.4	18.1 (0.2)	10.1 (0.3)	14 (1)	14.3	13.1 (0.9)	9.2 (0.1)	10 (2)	10.8
MnO	0.6	0.7	0.55 (0.05)	0.6	0.70 (0.05)	0.40 (0.05)	0.25 (0.01)	0.5	0.4 (0.1)	0.4 (0.1)	0.4 (0.1)	0.5
MgO	22.1	20.3	23.5 (0.5)	21.3	22.0 (0.4)	14.4 (0.5)	0.4 (0.1)	12.7	12.8 (0.1)	15.9 (0.8)	15 (2)	13.2
CaO	2.1	5.0	2.1 (0.9)	2.7	3.1 (0.3)	18.3 (0.8)	19 (2)	16.5	18 (1)	18.4 (0.5)	18 (3)	17.6
Na <sub>2</sub> O	0.01	0.06	0.04 (0.02)	0.1	0.06 (0.01)	0.28 (0.02)	0.255 (0.01)	0.2	0.24 (0.03)	0.19 (0.02)	0.20 (0.04)	0.3
K₂O	0.03	0.01	0.01 (0.01)	0.02	0.02 (0.01)	0.03 (0.02)	0.5 (0.1)	0.01	0.04 (0.02)	0.03 (0.01)	b.d.	0.1
Total	99.4	99.2	99.9 (0.3)	99.5	100.1 (0.5)	98.1 (0.2)	101.3 (0.3)	98.4	99.3 (0.8)	100.0 (0.6)	99.7 (0.5)	100.7
Mg # <sup>a</sup>	66.9	65.9	70.8 (0.3)	66.3	68.4 (0.4)	71.9 (0.9)	69 (1)	61.3	63.4 (0.7)	76 (1)	73 (1)	68.6
End- member ª												
Wo	4.4	10.5	4 (2)	5.8	6.5 (0.6)	40 (1)	39 (3)	36	39 (3)	39 (1)	38 (7)	39.6
En	64.0	59.0	68 (1)	62.4	64.0 (0.7)	43 (1)	42 (3)	39	39 (1)	46 (2)	45 (4)	41.5
Fs	31.6	30.5	28.0 (0.4)	31.8	29.5 (0.4)	17 (1)	19 (1)	25	22 (1)	15.0 (0.3)	17 (3)	19.0

590 Notes. <sup>a</sup>Pyroxene end-members and  $Mg \neq calculations$  are as in Table 3. FeO\* = total Fe

*expressed as*  $Fe^{2+}$ . *Pgt: pigeonite. b.d. = below detection level. Numbers in parentheses refer to* 

*Is of the mean of multiple analyses. Where the 1s is less than the relative error, the relative error* 

*is reported instead.* 

### 597 4.4.5 Olivine

598 Experimental OI (100-500  $\mu$ m) is present in charges at 950-1000°C, P = 100-150 MPa at NNO, 599 and 1000°C, 200 MPa, at NNO + 2 (Fig. 6d, Table 9). Its Fo content ranges from 59 to 76, and 600 increases with temperature for a water content of 3-5 wt. %. There is no systematic change in Fo 601 with increasing  $fO_2$  from NNO to NNO + 2. We can only compare the experimental Ol 602 compositions to Ol from the 2014 eruption, because the Ol from the 1990 and 2007 eruptions 603 have completely reacted to symplectites. There are two compositional groups of Ol from the 604 2014 eruption, with one at Fo = 72-75 and another at Fo = 67-69. The high-Fo group of Ol 605 overlaps with experimental OI from 100 MPa, 1060°C and NNO and ~3 wt. % water, and the low-Fo group overlap with experimental Ol from 60°C lower. 606 607 608 4.4.6 Amphibole

609 It was particularly difficult to identify Amph in the experimental charges, and we were 610 able to obtain good quantitative analysis of one experimental Amph (magnesio-hornblende; Mg 611 # = 69; Table 9) in the charge at 950°C, 400 MPa, and 6 wt. % dissolved water in the melt (Fig. 612 2g-h). We also identified additional Amph crystals in other charges using principal component 613 analyses of mineral compositions obtained with the electron microprobe analysis (Fig. S2). The 614 geothermobarometry results from the natural Amph of the 1990 and 2014 eruptions agree with 615 the experimental Amph stability field we have found (Fig. 4). We reconstructed the composition 616 melt from natural Amph from 1990 and 2014 according to the methodology of Li et al. (2021) to 617 compare the equilibrium  $SiO_2$  content of the parental melt of Amph with that of our glasses. We 618 find that the Al/Si ratio of Amph from 1990 correspond to 53-60 wt. %, which is close to the 619 SiO<sub>2</sub> content of the starting material glass. The equilibrium melt of Amph from 2014 is more 620 silica-rich, with 68-70 wt. % SiO<sub>2</sub>, which overlaps with the interstitial glasses. This suggests that 621 the Amph from 1990 and 2014 came from different melts, where the 1990 Amph was sampled 622 from a less differentiated melt than the 2014 Amph.

- 623
- 624

### 625 4.4.7 Fe-Ti oxides

Magnetite and ilmenite crystallized in experimental charges depending on the water content in the melt, and magnetite is more common (Table 10). Experimental magnetite ( $X_{usp} =$ 0.24-0.60) is present in almost all water-saturated charges. Ilmenite occurs in water poor charges. Although we have identified ilmenite in charges through its EDS spectrum, we were not able to quantitatively determine their compositions.

631

Mineral <sup>a</sup>	Amph	Amph	Amph	Amph	OI	OI	OI	OI	OI	OI	OI
T (°C)	1000	1000	950	950	1000	1000	950	1060	1060	1000	1000
Charge	SUPL1- 16	SUPL1- 17	SUPL1- 32	SUPL1- 28	SUPL1- 1	SUPL 1-6	SUPL1- 21	SUPL1- 38	SUPL1- 39	SUPL1- 58	SUPL1- 59
n:	1	1	1	1	7	1	1	1	2	3	3
wt. %											
SiO <sub>2</sub>	48.5	48.9	48.6	52.2	37.2 (0.4)	36.3	36.2	37.5	37.9 (0.5)	38.0 (0.4)	37.5 (0.4)
TiO <sub>2</sub>	0.6	0.9	0.5	0.5	0.02 (0.01)	0.02	0.02	0.05	0.02 (0.01)	0.02 (0.01)	0.02 (0.01)
$AI_2O_3$	4.5	10.1	5.3	4.0	0.2 (0.2)	0.02	0.02	0.03	0.04 (0.01)	0.03 (0.01)	b.d.
Cr <sub>2</sub> O <sub>3</sub>	0.1			0.1	b.d.	0.01	b.d.	0.02	0.04 (0.02)	b.d.	b.d.
FeO*	14.6	11.9	15.3	18.4	27.8 (0.2)	27.7	34.1	21.8	21.6 (0.2)	25.1 (0.2)	28.5 (0.3)
MnO	0.4	0.4	0.5	0.6	0.65 (0.02)	0.6	0.8	0.5	0.51 (0.01)	0.53 (0.02)	0.58 (0.01)
MgO	13.1	11.0	15.9	11.9	33.6 (0.2)	32.7	28.1	38.1	38.4 (0.4)	35.7 (0.1)	32.7 (0.3)
CaO	15.7	16.4	12.1	12.7	0.3 (0.1)	0.2	0.2	0.3	0.28 (0.03)	0.23 (0.01)	0.27 (0.4)
Na <sub>2</sub> O	0.7	1.2	0.3	0.3	b.d.	0.001	0.01	0.01	0.01 (0.01)	b.d.	b.d.
K <sub>2</sub> O	0.2	0.1	0.0	0.1	b.d.	0.03	0.01	0.01	0.01 (0.01)	b.d.	b.d.
Total	98.5	100.7	98.5	100.8	99.4 (0.5)	97.4	99.2	98.1	98.6 (0.7)	99.4 (0.04)	99.4 (0.1)
End- member <sup>a</sup>	Mg-hbl	und.	Mg-hbl	Mg-hbl							
Mg # <sup>a</sup> Fo content <sup>a</sup>	61.5	62.2	68.7	53.6	68.3 (0.2)	67.8	59.5	75.7	76 (1)	71.7 (0.2)	66.9 (0.3)

### 632 **Table 9**. *Experimental amphibole and olivine compositions*

633

 $634 \qquad Notes. See Table 2 for meaning of abbreviations. Mg-hbl = Magnesio-hornblende. FeO* = total$ 

635 Fe expressed as  $Fe^{2+.}Mg \# = 100 \times Mg / (Mg + Fe^*)$ . b.d. = composition below detection level.

- *und.* = *undetermined. Numbers in parentheses refer to 1s of the mean of multiple analyses.*
- 637 Where the 1s is less than the % error, the % error is reported instead.

**Table 10:** *Experimental Fe-Ti oxides chemistry.* 

T (°C)	1000	1000	1000	1000	1000	1000	1000	
Charge	SUPL1-1	SUPL1-2	SUPL1-3	SUPL1-12	SUPL1-13	SUPL1-14	SUPL1-15	
n:	1	1	3	3	7	8	3	
wt .%								
SiO <sub>2</sub>	0.5	0.3	0.30 (0.07)	0.30 (0.08)	0.21 (0.03)	0.3 (0.1)	0.3 (0.1)	
TiO <sub>2</sub>	5.2	11.5	10.9 (0.5)	8.1 (0.1)	9.0 (0.3)	13.2 (0.1)	16.9 (0.1)	
$AI_2O_3$	5.7	3.3	3.5 (0.1)	5.3 (0.1)	4.8 (0.1)	3.8 (0.1)	3.1 (0.1)	
FeO*	67.0	71.8	70.8 (0.7)	71.4 (0.2)	70.5 (0.7)	70.0 (0.2)	63.8 (0.2)	
MnO	0.5	0.4	0.47 (0.02)	0.42 (0.01)	0.46 (0.01)	0.46 (0.01)	0.51 (0.01)	
MgO	3.7	2.4	2.37 (0.04)	3.05 (0.02)	2.8 (0.1)	2.43 (0.03)	2.1 (0.1)	
CaO	0.2	0.3	0.24 (0.03)	0.2 (0.1)	0.15 (0.01)	0.20 (0.04)	0.25 (0.02)	
Total	82.8	90.0	88.6 (0.8)	88.8 (0.3)	88.0 (0.8)	90.4 (0.2)	86.93 (0.02)	
FeO <sup>c</sup>	54.0	59.2	58.4 (0.5)	57.7 (0.1)	57.5 (0.5)	58.3 (0.1)	55.3 (0.1)	
Fe <sub>2</sub> O <sub>3</sub>	14.5	14.1	13.8 (0.2)	15.2 (0.1)	14.5 (0.2)	13.0 (0.1)	9.5 (0.1)	
X <sub>usp</sub>	0.24	0.38	0.37(0.01)	0.28 (0.01)	0.31 (0.01)	0.44 (0.01)	0.58 (0.01)	
·								
T (°C)	950	950	950	950	950	950	950	950
Charge	SUPL1-28	SUPL1-29	SUPL1-30	SUPL1-31	SUPL1-32	SUPL1-64	SUPL1-65	SUPL1-67
n:	1	1	1	1	1	1	1	2
wt .%								
SiO <sub>2</sub>	0.3	0.4	0.6	0.4	0.3	0.3	0.3	0.2 (0.1)
TiO <sub>2</sub>	15.8	16.2	16.7	15.1	16.0	16.3	7.5	12.0 (0.7)
$AI_2O_3$	2.9	2.6	2.7	2.7	4.5	2.6	3.8	3.00 (0.05)
FeO*	70.9	68.6	64.8	69.9	63.9	67.0	75.9	72.4 (0.9)
MnO	0.3	0.4	0.4	0.5	0.5	0.4	0.5	0.41 (0.02)
MgO	0.7	1.4	1.4	1.7	2.2	1.4	1.8	1.52 (0.01)
CaO	0.3	0.2	0.3	0.4	0.3	0.3	0.3	0.30 (0.03)
Total	91.2	89.9	87.0	90.7	87.8	88.4	90.3	90 (1)
FeO <sup>c</sup>	57.7	58.8	56.4	59.1	55.0	60.5	61.4	60.2 (0.6)
Fe <sub>2</sub> O <sub>3</sub>	10.4	10.9	9.3	12.1	9.9	11.5	16.2	13.7 (0.3)
V	0.50	0.53	0.57	0.49	0.55	0.54	0.25	0.39 (0.01)

642 Notes.  $Fe_2O_3$  calculated using the approach by Droop (1987), where  $Fe_2O_3 = FeO^* x (Fe^{3+} / Pe^{3+})$ 

 $Fe^{2+} + Fe^{3+}$  x 1.1113.  $FeO^c = FeO$  calculated using the approach by Droop (1987), where

 $FeO^{c} = FeO^{*} x (Fe^{2+} / Fe^{2+} + Fe^{3+})$ . Numbers in parentheses refer to 1s of the mean of multiple

645 *analyses.* Where the 1s is less than the relative error, the relative error is reported instead. b.d.





Figure 6. Variations in experimental mineral compositions of experimental charges compared to natural ones (Plag, Cpx, Opx, and Ol) of the crystal cargo of 1990, 2007, and 2014 eruptions: a. An in Plag, b. Mg # in Opx, c. Mg # in Cpx, and d. Fo content in Ol. Coloured solid boxes denote the observed ranges of mineral compositions from the three eruptions, with the same colours as outlined in Fig. 5. Open boxes represent rim compositions of the minerals (Utami et al. (in revision) and Table 2 of this study).

### 655 **5 Discussion**

656 5.1 Geothermobarometry and experimental constraints on magma storage conditions of sub-

657 Plinian eruptions and dome extrusions

658 Combining the geothermobarometric and experimental results, we find that the pre-659 eruptive magma storage conditions that match the phenocryst content, natural phase assemblage, 660 and some of the mineral compositions for the three eruptions are at  $975 \pm 39^{\circ}$ C,  $175 \pm 25$  MPa, 661 with 4-6 wt. % water in the melt and a fO<sub>2</sub> of NNO. These conditions correspond to about 7 km of storage below the crater if we use a crustal density of 2640 kg m<sup>-3</sup>. However, this does not 662 mean that all of the crystal cargo grew at this depth, as amphibole geobarometry indicates 663 664 pressures of up to 400 MPa (up to ~15 km), and hence some these crystals were probably 665 entrained from deeper parts of the magmatic system (Fig. 4a; Utami et al., in revision). A key 666 difference between the 1990 and 2014 eruptions and those of the 2007 dome is the much hugher 667 phenocryst content of the latter (Table 2). This could be due to crystal accumulation in the dome, 668 but the major and trace element compositions of the three eruptions are similar, including 669 incompatible trace elements such as Zr (this study; Jeffery et al., 2013), meaning that the 670 observed difference in crystal content is due to in situ crystallization. According to experimental 671 results, the lava dome's higher phenocryst content (Fig 3) can be reproduced with ~1 wt. % 672 water in the melt for a temperature of about 975°C. Thus we propose that the low apparent water 673 content is likely the result of additional storage at shallower pressures and/or transport towards 674 the surface that led to water loss and cooling, rather than a difference in the initial pre-eruptive 675 conditions. The presence of the pseudomorphs and symplectites that used to be Amph and Ol in 676 the 2007 rocks also indicate that the magma was originally stored at similar conditions to the 677 1990 and 2014 eruptions, although its slower ascent resulted in phenocryst proportions and 678 compositions to re-equilibrate to shallower depths.

679

680 5.2 The importance of excess volatiles in determining eruption style

681 The explosive → effusive → explosive eruption style transition shown by the 1990-2007682 2014 events remains some of the most intriguing aspects of Kelud volcano. Given that the pre683 eruptive or "initial" (e.g. before ascent) conditions of temperature, pressure, and water content in

684 the melt are broadly the same for the three events, other variables have played a role in 685 producing the different eruption styles. In Utami et al. (in revision), we proposed that the 686 explosive eruptions of the 1990 magma was likely due to excess fluid accumulation, and we 687 calculated that up to 0.12 Mt SO<sub>2</sub> could have resided as a separate fluid phase. Our mass balance 688 calculations for the SO<sub>2</sub> budget of the 2014 eruption also indicate the presence of an exsolved 689 volatile phase at pre-eruptive conditions, with up to 0.1 Mt SO<sub>2</sub> (Table 11). Such excess fluid can 690 be developed via progressive magma crystallization and volatile buildup to supersaturation (e.g. 691 Arzilli et al., 2019; Cassidy et al., 2019; Tramontano et al., 2017), with additional fluids can also 692 be due to fluid fluxing and degassing from a deep-seated basaltic magma reservoir and/or 693 limestone in the case of Kelud (e.g. Caricchi et al., 2018; Giuffrida et al., 2017; Utami et al., in 694 revision). Although we can only calculate the amount of SO<sub>2</sub>, the fluid phase would have 695 certainly included water and  $CO_2$ . The presence of an exsolved fluid phase at pre-eruptive 696 conditions means that the magma buoyancy is greatly increased, as fluids can expand from the 697 moment the magma starts its ascent and lithospheric pressure decreases (Gonnermann & Manga, 698 2013). The additional buoyancy could lead to significantly faster magma ascent, depending on 699 the distribution of the fluid in the magma and whether there is a one- or two-phase flow, and also 700 lead to much higher explosivity.

701

702 Our hypothesis differs from that of Cassidy et al. (2019), who proposed that the 703 difference in eruption style is dependent on the degree of water saturation in the magma. They 704 further proposed that the 2007-2008 dome extrusion was triggered by an injection of hot, CO<sub>2</sub>-705 rich magma that suppressed the water content, and the 2014 explosive eruption was driven by 706 water saturated magma. On the other hand, Jeffery et al. (2013) invoked additional processes 707 such as crustal contamination, inter-crustal magma mixing, xenolith incorporation, and magma 708 migration, where the volatile content degassed from 8 wt. % to 2-3 wt. % during magma 709 migration. We have as yet no definitive geochemical or textural evidence to suggest that magma 710 mixing is a dominant process at Kelud based on mineral chemistry and textures interpretations 711 presented in Utami et al. (in revision).

- 712
- 713

714	<b>Table 11.</b> Excess volatile estimates for the 2014 magma using the petrologic method of Devine et
715	al. (1984).

	14-PUMI-2	Units
Erupted volume (DRE) <sup>a</sup>	0.28	km <sup>3</sup>
Rock density	2404	ka/m <sup>3</sup>
Mass of magma	673	Mt
Max. concentration of SO <sub>2</sub> in melt inclusion	547-582	ppm
Max concentration of SO <sub>2</sub> in interstitial glass	95-126	ppm
Glass proportion	0.66-0.71	
SO <sub>2</sub> degassed from the melt	0.20-0.22	Mt
Atmospheric SO <sup>b</sup>	0.30	Mt
Excess SO <sub>2</sub>	0.08-0.10	Mt

### 717

719

720 The accumulation of an excess fluid also requires that the plumbing system behaves as a 721 closed system, or with the mass flux of exsolved fluids that was produced being larger than that 722 which is degassed, implies that there was limited passive degassing between eruptions. Kelud 723 volcano is considered semi-plugged by Whelley et al. (2015). Prior to 1990 eruption, degassing 724 from depth was likely blocked by a plug of crystalline magma from the previous event in 1966 725 and ~4 million  $m^3$  of lake water (Lesage & Surono, 1995; Vandemeulebrouck et al., 2000), 726 whereas before the 2014 eruption, the volcano was plugged by the 2007 dome (Global 727 Volcanism Program, 2014). The 1990 plug and 2007 dome likely provided little permeability for 728 the fluids to escape, in addition to exerting relatively high lithostatic/hydrostatic pressure. These 729 conditions could have effectively decreased the rock's permeability and allowed fluids and 730 overpressure to accumulate before eruption. Minerals such as cristobalite likely formed at 731 shallow pressures (< 50 MPa) during and after dome emplacement and contribute reducing the 732 permeability of the dome (Horwell et al., 2013; Martel & Schmidt, 2003). In contrast, after the 733 1990 eruption and up until 2007, there was significant outgassing from arcuate fractures 734 observed, and an "embryonic" dome sealed the conduit (Global Volcanism Program, 1990b). 735 The observed fumarolic activity and outgassing likely stemmed from active degassing of a 736 shallow magma body from a residual magma intrusion, similar to a resurgent dome (Acocella et 737 al., 2000; Global Volcanism Program, 1990b). Between 1990 and 2007, there were also several 738 crises involving two heating episodes, when the lake water increased in temperature from 40°C 739 and 35°C in 1996 and 2001, respectively to ~50°C (Bernard & Mazot, 2004). The observed post-

<sup>718</sup> *Notes.* <sup>a</sup>Erupted volume from Maeno et al. (2019). <sup>b</sup>SO<sub>2</sub> from Carn (2019).

and inter-eruptive crises between 1990 to 2007 suggest the magma system was partially open,

allowing the shallow magma body to further degas volatiles out of the system and thus was not

able to accumulate a significant amount of excess fluids.

743

### 744 5.3 Relating pre-eruptive processes to unrest signals

745 Volcanic unrest at Kelud has been monitored using a range of observations, including 746 seismicity, lake chemistry and its temperature, and deformation. Unrest signals preceding all 747 three events started around 2-3 months before eruption, and included seismic swarms with 748 seismic unrest signals spanning volcano-tectonic, shallow VB, low frequency, tremor (e.g. 749 Hidayati, 2009; Hidayati et al., 2018; Lesage & Surono, 1995). Seismicity escalated rapidly (e.g. 750 < 24 hours) before the 1990 and 2014 eruptions, and earthquake hypocenters are mostly 751 concentrated around shallow depths beneath the crater (< 7 km) (Hidayati et al., 2018; Lesage & 752 Surono, 1995). Sharp increases in lake temperature and input of magmatic volatiles such as Cl<sub>2</sub> 753 and CO<sub>2</sub> into the lake have also been proposed for the unrest prior to the 1990 and 2007 754 eruptions (Bernard & Mazot, 2004; Caudron et al., 2012; Kunrat, 2009; Vandemeulebrouck et 755 al., 2000). Tiltmeters and interferometric synthetic aperture radar (InSAR) detected changes in 756 local and regional deformation from inflation to rapid deflation preceding 2014 eruption and 757 2007 dome extrusion, respectively (Hidayati et al., 2018; Lubis, 2014). Despite the diverse 758 monitoring data available, it is still difficult to anticipate eruptions at Kelud, including whether 759 the ensuing eruption will be effusive or explosive (Nakamichi et al., 2017).

760

761 Results of our study suggest that the accumulation of an excess volatile phase prior to the 762 eruptions of 1990 and 2014 was a key parameter to their explosivity (Fig. 7). Contrasting this 763 with the inter-eruptive period between the 1990 and 2007, where the system was able to degas 764 more freely, which then resulted in the 2007 eruption being effusive. If our hypothesis is correct, 765 identifying monitoring signals that could be related to fluid accumulation and magma degassing 766 or transfer within the plumbing system could be useful in anticipating explosive eruptions. One 767 possibility is to monitor deep long period seismicity using broadband seismometers as this has 768 been interpreted as degassing of volatile-rich magma at depth (Melnik et al., 2020). Another 769 possibility are microgravity surveys or continuous gravity monitoring, as we can expect that

significant fluid accumulation would lead to subtle changes in density of the plumbing system

771 over time, as demonstrated in Masaya volcano (Nicaragua) (e.g. Williams-Jones et al., 2003).

772



- 773
- 774

775 Figure 7. Conceptual model of the magmatic conditions and processes occurring for the 1990-776 2014 Kelud eruptions. Black rectangles represent the crystal cargo, or crystallization in the 777 conduit. White flattened circles represent accumulation of the pre-eruptive excess fluid phase 778 present before explosive eruptions. Solid white arrows represent fluid fluxing through the system 779 either from a deep-seated degassing basaltic magma, or from limestone assimilation. Dashed 780 white arrows represent fluid release through a semi-permeable conduit leading up to dome 781 extrusion. The 1990 and 2014 magmas were able to accumulate an excess volatile phase and 782 hence the higher explosivity of their eruptions, where as a higher permeability of the system 783 between 1990 and 2007 allowed for extended degassing and slower ascent of the 2007 as a 784 dome. Pressures are converted to depth below the crater using the density calculated in Smyth et 785 al. (2007). The volcaniclastic and carbonate layers are also in Smyth et al. (2007), as 786 represented in Jeffery et al. (2013). Volcano Explosivity Indices (VEI) reported come from 787 Global Volcanism Program (2013). Storage depth constraints come from Cassidy et al. (2019)

- 788 (2014 eruption) [1], Jeffery et al. (2013) (2007 dome) [2], and Utami et al. (in revision) (1990
- ruption) [3]. Plume height were reported in Bourdier et al. (1997) for 1990; Global Volcanism
- 790 *Program (2008) for 2007; and Kristiansen et al. (2015) for 2014.*
- 791

### 792 **6 Conclusions**

793 We present petrological and experimental constraints on the pre-eruptive storage 794 conditions of the basaltic andesite magma feeding the 1990, 2007, and 2014 eruptions of Kelud 795 volcano, which alternated between explosive  $\rightarrow$  effusive  $\rightarrow$  explosive eruption styles. The three 796 events share similar whole rock compositions, mineralogy, implying that their magma storage 797 conditions were also similar. We found that the natural phenocryst conten and mineral 798 assemblage can be experimentally reproduced at 975  $\pm$  39°C, 175  $\pm$  25 MPa (~7 km below the 799 crater level), NNO, and with 4-6 wt. % water in the melt. We also found that the 1990 and 2014 800 magmas coexisted with an exsolved volatile phase at pre-eruptive conditions, and we propose 801 that this played a main role to their explosive style. The lava dome was likely a residual magma 802 batch from the 1990 eruption or earlier that had no excess fluids, as the conduit was partially 803 open after the 1990 eruption that allowed gases to be released. Monitoring techniques such as 804 continuous microgravity surveys could help to identify changes in density from fluid 805 accumulation and thus may be able to anticipate eruption styles.

806

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- 818 4057-88e2-19c0480fa99e), and available as online supplementary material (Datasets S1 and S2).
- 819 We are in the process of archiving our data. There are no financial conflicts of interests between
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