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2 **A reassessment of the sulfur, chlorine and fluorine atmospheric loading during**
3 **the 1815 Tambora eruption**

4
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17 **Abstract**

18 The 1815 eruption of Mount Tambora (Sumbawa Island, Indonesia), largest known explosive
19 eruption in recorded history, was cataclysmic. It was responsible for a strong short-term global
20 atmospheric cooling the following year, known as “the year without a summer”. To evaluate the
21 climatic impact, an accurate quantification of volatile elements degassed during this eruption is
22 crucial. In this study, we re- evaluate the atmospheric release of sulfur, chlorine and fluorine during
23 the 1815 eruption using the petrological approach based on plagioclase-hosted melt inclusions. The
24 pre-eruptive (melt inclusions) and post-eruptive (matrix glass) volatile element concentrations of the
25 magma are measured by electron microprobe. We discuss three different outgassing scenarios and
26 conclude that 147 ± 17 Tg of SO_2 , 49 ± 5 Tg of Cl and 20 ± 2 Tg of F were degassed during the eruption,
27 considering closed system ascent and degassing. The SO_2 results take into account the dissolution of
28 sulfides which are present in melt inclusions and plagioclase crystals but not in matrix glasses. Our
29 new estimates are higher than previous estimations from petrological methods or derived from ice
30 cores but are consistent with atmospheric optical depth observations from 1816. The 1815 eruption
31 of Tambora ranks in first place in terms of volcanic SO_2 emission in the last 2000 years, higher than
32 the 1257 Samalas eruption (Lombok Island, Indonesia) if equal methodologies are applied. These
33 estimates remain nonetheless minima as they do not account for the possible additional contribution
34 of a pre-existing gas phase in the magma reservoir.

35 **Keywords** Degassing · Volatile elements · Melt inclusions · Petrological method

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42 **Conflicts of interest/Competing interests**

43 The authors declare no conflicts nor competing interest.

44 **Availability of data and material**

45 All data obtained in this study are presented in the main text and in the Supplementary Information
46 online.

47
48 **Introduction**

49
50 The 1815 eruption of Tambora is known as the largest explosive eruption of the common Era (e.g.,
51 Newhall and Self 1982; de Jong Boers 1995). With a Volcanic Explosivity Index (VEI) of 7 (Newhall
52 and Self 1982), the quantity of ash and gases released by the eruption plunged much of Indonesia into
53 darkness for more than three days (de Jong Boers 1995). The plumes formed during the Plinian
54 activity of the eruption carried gases and fine ash well into the stratosphere. Once at stratospheric
55 altitudes, sulfur gases are converted into sulphate aerosols that reflect solar radiation and cause a
56 global cooling of both the troposphere and Earth's surface. In addition, volcanogenic halogens such
57 as chlorine and fluorine have a destructive effect on stratospheric ozone (e.g., Schmidt and Robock
58 2015). After 1815, both of these effects were observed (e.g., Briffa et al. 1998; D'Arigo et al. 2009).
59 The eruption of Tambora is considered to be the cause of "the year without a summer" in 1816 (e.g.,
60 Rampino et al. 1988; Oppenheimer 2003). That year, the surface temperatures were about 0.78°C
61 colder than average worldwide and the decade 1810-1819 is considered the coldest in more than 500
62 years (Briffa et al. 1998; D'Arigo et al. 2009). In Europe, summer temperatures were 1-2 °C cooler
63 than average. During the years following 1816 spectacularly colored sunsets and twilights were
64 observed in London, a constant "dry fog" was reported in the northeastern USA and the winters were
65 stormier (e.g., Oppenheimer 2003). Such a global climate perturbation caused crop failure in
66 Indonesia and Europe, thus leading to famines, diseases, and sometimes popular uprising (e.g., de

67 Jong Boers 1995; Oppenheimer 2003). The close relationship between large explosive eruptions and
68 climate has been the subject of many studies including several on Tambora (e.g., de Jong Boers 1995;
69 Devine et al. 1984; Self et al. 2004).

70 To understand the eruption-climate link it is critical to have a reliable estimate of the bulk
71 amount of sulfur and other volatiles released by the eruption and injected into the stratosphere. The
72 sulfur quantity released by the eruption of Mount Tambora was first estimated at 17 Tg (or 34 Tg
73 SO₂) using melt inclusions contained in plagioclase crystals of the 1815 magma (Devine et al. 1984).
74 This quantity was later re-estimated at 86 Tg SO₂ and 53-58 Tg SO₂ using the same petrological
75 approach by Sigurdsson et al. (1992) and Self et al. (2004) respectively. Another method, the study
76 of volcanic sulphate deposits in Antarctic ice cores, gave an estimated output higher than 98 Tg of
77 SO₂ (Legrand and Delmas 1987), later re-evaluated to 45 Tg SO₂ (Sigl et al. 2014).

78 In this study, we re-evaluate the amounts of volatile elements released during the 1815
79 Tambora eruption by providing new analyses of S, Cl and F in plagioclase-hosted melt inclusions.
80 We propose three degassing scenarios in order to obtain estimates that are directly comparable with
81 similar studies at other volcanoes.

82 **Volcanological background**

83 **Geological setting and past activity**

84 Mount Tambora is a volcano located on Sumbawa Island (Indonesia) which is part of the Sunda Arc
85 (Fig. 1). In this subduction zone, the Indo-Australian plate subducts below the Eurasian plate at a
86 speed of 6-7 cm/y (Hamilton 1979). The Sumbawa sector of this subduction is unique in being
87 composed of young and thin crust which is flanked both to the north and south by oceanic crust
88 (Foden and Varne 1980).

89 Four older volcanic formations are exposed in the walls of the Tambora caldera, below the
90 1815 deposits (Sigurdsson and Carey 1992). The oldest one, composed of lava flows, has an age of
91 43 ka (Barberi et al. 1987). It is overlain by two pyroclastic units and a level of interbedded pyroclastic
92 surge and fall deposits which represents the latest activity of the volcano before the 1815 eruption
93 (Sigurdsson and Carey 1992). From the beginning of its activity, Tambora has produced
94 undersaturated, moderately K₂O-rich magmas ranging from nepheline-normative trachybasalt (Ne-
95 trachybasalt) to Ne-trachyandesite (Foden and Varne 1980).

96 **1815 eruption chronology and stratigraphy**

97 The 1815 eruption of Tambora lasted several days and consisted of two main phases. The first
98 includes four episodes of tephra falls F1, F2, F3 and F4 while the second consists of a succession of
99 pyroclastic flows, surges and co-ignimbrite ash fall (Sigurdsson and Carey 1989).

100 Phase 1: Tephra fall deposits (Fig. 2) (Sigurdsson and Carey 1989)

101 The first deposit of this sequence is a silt to sand size ash fall layer named F1. This layer is
102 the product of phreatomagmatic activity, and it can be divided into two units suggesting at least two
103 main explosions. It is 1-10 cm thick on the flanks of the volcano and mainly composed of blocky to
104 vesicular pale-brown-green pumice, glass shards, dark scoria fragments, crystals of feldspar, biotite,
105 clinopyroxene, magnetite and some lithics. A pale grey-green Plinian pumice fall deposit overlies the
106 F1 unit. This F2 layer was produced by a Plinian eruption which generated a plume 33 km high
107 (Sigurdsson and Carey 1989). It is 10 to 30 cm on the slopes on the volcano. It contains highly
108 vesicular, lapilli-sized pale-grey-green pumice and some glass shards. Overlying F2 is a second layer
109 of sandy-silty ash fall, similar to the F1 deposits but more extensive and thicker. This F3 layer is also
110 the product of phreatomagmatic eruptions, and three units can be divided within it, suggesting at least
111 three different explosions. The entire layer is about 10 cm thick close to the edifice and consists of
112 light-colored pumice, glass shards, dark scoria and lithics. This first phase of tephra falls ends with
113 the F4 layer, which is the thickest of the sequence. The F4 layer is the product of another Plinian
114 eruption which generated a plume 43 km high (Sigurdsson and Carey 1989). The deposits are 25 to
115 30 cm thick on the flanks of Tambora and consists mainly of highly vesicular pale-grey pumice and
116 glass shards. This first phase of phreatomagmatic and Plinian eruptions (F1 to F4) had a duration of
117 more than five days and ejected 4.6 km³ of tephra (Sigurdsson and Carey 1989).

118 Phase II: Pyroclastic flows, surges and distal tephra fall (Sigurdsson and Carey 1989)

119 The second main phase of the 1815 eruption corresponds to the collapse of the second (F4) Plinian
120 volcanic plume, generating pyroclastic surges and flows. The first deposit of this sequence is a
121 pinkish-grey to grey sandy-silty ash surge layer named S1 with pumices and charcoal at some
122 localities. This layer is about 15 cm thick on the volcano's flanks and the pumices reach 2-3 cm in
123 diameter. Overlying S1, is a series of at least 7 pyroclastic flow deposits (PF1, PF2...). They cover
124 almost all of the Sanggar peninsula, reaching the ocean in most directions from the volcano and
125 measure a maximum thickness of 20 m (average about 7 m). Finally, two distal ash falls (F5 and F6)
126 end this second main phase. F5 is 12-25 cm thick and composed of greyish-brown, poorly sorted,
127 silty-sandy ash with some angular pumice clasts. This layer consists of co-ignimbrite ash released by
128 the S1 phase. Unit F6 overlies the S1 unit at some localities with a thickness of 3-4 cm on the western
129 flank of Tambora.

130 **Erupted magma volume**

131 The largest uncertainty in estimates of volatile release to the atmosphere during the 1815 eruption is
132 the total erupted magma volume. Based on extensive two-month field work in Sumbawa and on the
133 many smaller nearby islands that surround it, Sigurdsson and Carey (1989) arrived at a total magma
134 volume estimate for the Tambora 1815 eruption of 51 km³ DRE (dense-rock equivalent). A thorough
135 reevaluation of the erupted volume by Kandlbauer and Sparks (2014), including some new sea floor
136 coring data, and consideration of Tambora's caldera collapse structure, provides a new revised total
137 volume estimate of 41±4 km³ DRE. The sulfur, chlorine and fluorine atmospheric loadings calculated
138 from these two estimates are shown in Table S1 in Supplementary Material 1. However, in the
139 remainder of this paper, we report all calculations using the most recent magma volume estimate of
140 41 km³ DRE (Kandlbauer and Sparks 2014).

141 **Petrology and geochemistry of the 1815 magma**

142 Since the beginning of its activity, Tambora has erupted undersaturated potassic magmas, ranging
143 from nepheline-normative trachybasalts to nepheline-normative trachyandesites (Foden and Varne
144 1980). The 1815 products have a nepheline-trachyandesite composition with high silica content (54-
145 57 wt.% SiO₂), unusual in an island-arc environment (Foden 1986). Pumices are latitic to
146 tephriphonolitic with 30-50% vesicles and contain phenocrysts in a glassy or microcrystalline matrix.
147 The mineral assemblage is plagioclase > clinopyroxene > magnetite with occasionally small amounts
148 of apatite, biotite and olivine (Gertisser et al. 2012). Foden (1986) found that plagioclase in pumice
149 showed a uniform very calcic population ranging from An₈₀ to An₉₀. However, Self et al. (2004)
150 found that the plagioclases can be separated into two distinct populations: (1) unzoned crystals of
151 almost constant composition (An_{58±6}) and (2) zoned crystals with a very calcic core (≤An₉₁) and rims
152 with a similar composition as the unzoned ones. Melt inclusions are common in both plagioclase
153 populations. The clinopyroxenes are mostly phenocrysts of Ca-rich augite (Mg₇₀-Mg₇₅) with minor
154 quantities of Ti and Al (Foden 1986; Gertisser et al. 2012).

155 Sigurdsson and Carey (1989) analysed pumice samples from all the layers of the first main
156 phase and from the seven major pyroclastic flows of the second one. Their bulk rock study showed
157 that “the eruption tapped a very homogenous magma body”. Their range in bulk rock SiO₂
158 concentrations fell “within the precision of the analytical method” (x-ray fluorescence). The matrix
159 glass analyses confirmed that “the glass composition in falls, surges and flows is identical”. A more
160 recent study (Suhendro et al. 2021) demonstrated a slight evolution in total rock and mineral
161 chemistry between the different eruption phases. Plinian fall units are characterized by a lower
162 phenocryst abundance (avg. of 5.1%) and higher silica content (bulk pumice, 58–58.5 wt.%) than

163 pyroclastic flow deposits which have a relatively higher crystal abundance (avg. of 12.1%) and a
164 lower silica content (bulk pumice, 56.7–57.9 wt.%; Suhendro et al. 2021). The analysis of the deposit
165 stratigraphy suggests phenocryst stratification in the reservoir was established prior to the 1815
166 eruption, this being responsible for the slight contrast in bulk compositions (Suhendro et al. 2021).

167 **Materials and Methods**

168 In order to quantify S, Cl and F release during the 1815 eruption, we used samples previously
169 collected by Sigurdsson and Carey (1989). The samples used here are tephra from the two Plinian
170 layers of the eruption: sample TB61 from episode F2 and sample TB65 from episode F4 (Fig. 2).
171 This F4 unit can be considered representative of the bulk of the magma, firstly because it was fed by
172 the same ignimbritic magma as unit F5, the most voluminous one (Self et al. 2014) but also because
173 the composition of the melt (matrix glass, melt inclusions) remained identical throughout the entire
174 event (Sigurdsson et al. 1989; Suhendro et al. 2021). For both F2 and F4 samples, tephra are light
175 grey, vesiculated pumices measuring 2 to 3 cm. They have a glassy matrix and contain 10% of
176 phenocrysts which are plagioclase, clinopyroxene and occasionally magnetite, biotite and apatite. We
177 collected plagioclase from these tephra in which melt inclusions are commonly found.

178 Plagioclase phenocrysts found in tephra can be divided into two subpopulations: crystals with
179 zoned rims and unzoned crystals (Fig. 3). Both populations contain abundant melt inclusions (MI).
180 They range in size from few micrometres to 150 μm in length. We selected MIs that are entirely
181 glassy, oval-shaped and contain a shrinkage bubble (Fig. 4a, c) for analysis; other types (partially
182 crystallized and/or with several bubbles are present) were excluded (Fig 4b, c, d). Sulfide globules
183 have not been reported in previous studies but were found in some of the melt inclusions and
184 plagioclase crystals (Fig. 3). Their size ranges from 1 to 50 μm and they are mostly spherical. No
185 sulfides have been found in the matrix glass.

186 **Analytical methods**

187 **Electron microprobe**

188 Major and volatile elements were analysed in selected melt inclusions, matrix glass (MG) and
189 plagioclases by electron microprobe (Cameca SXFive-TACTIS of LMV, Clermont-Ferrand, France)
190 following standard procedure (e.g., Le Voyer et al. 2008; Rose-Koga et al. 2020). Major elements in
191 plagioclase crystals were analysed using a focused beam at 15 nA and 15 kV, whereas melt inclusions
192 and matrix glasses were analysed with a defocused 20 μm beam at 8 nA and 15 kV. For S, Cl, F
193 contents in melt inclusions and matrix glass, a defocused 10 or 20 μm beam at 40 nA and 15 kV was
194 used. Typical errors on measurement for each element are given in 1σ for plagioclase crystals and 3σ

195 for melt inclusions and matrix glasses: <1% SiO₂, Al₂O₃, FeO, MgO, CaO, K₂O; <2% Na₂O; <4%
196 TiO₂; <20% MnO, P₂O₅; <10% S, Cl and <15% F. Procedure characteristics are summarized in
197 Tables S2 and S3 in Supplementary Material 1.

198 Scanning electron microscope (SEM)

199 After EMPA analysis, a scanning electron microscope (JEOL JSM-5910LV of LMV, Clermont-
200 Ferrand, France) was used to image, in backscattered electrons, the sulfides and plagioclase zoning.
201 Spot analyses were carried out on sulfides with a defocused 56 nm beam at 80 μA and 15 kV.

202 Petrological method

203 The petrological method used here was described by Devine et al. (1984), Sigurdsson et al. (1992)
204 and Self et al. (2004) for Tambora and applied to several other eruptions (e.g., Thordarson 2003 for
205 1783 Laki eruption; Mandeville et al. 1996 for 1883 Krakatau eruption; Iacovino et al., 2016 for 946
206 Peaktu eruption; Vidal et al. 2016 for 1257 Samalas eruption; Peccia et al. in press for 43 BCE Okmok
207 eruption). It consists of comparing the pre-eruptive (melt inclusions) and post-eruptive (matrix glass)
208 sulfur, chlorine and fluorine concentrations to estimate by difference the degassed quantities of the
209 respective volatile elements. We used the following mass balance equations to estimate the total
210 masses of SO₂, Cl and F released:

$$211 \quad E_{SO_2} = 2 \times M_v \times (1 - W_{xls}) \times (C_{incl} - C_{matrix})/100 \quad (1)$$

$$212 \quad E_{Cl} = M_v \times (1 - W_{xls}) \times (C_{incl} - C_{matrix})/100 \quad (2)$$

$$213 \quad E_F = M_v \times (1 - W_{xls}) \times (C_{incl} - C_{matrix})/100 \quad (3)$$

214 where E_{SO_2} is the SO₂ emission in kg, M_v the mass of erupted magma in kg (1.01×10^{14}), W_{xls} the
215 mass fraction of crystals in the magma (0.115; mean of 0.10 (Self et al. 2004) and 0.13 (Sigurdsson
216 and Carey 1989)) and $C_{incl} - C_{matrix}$ the difference between the average concentrations of the glass
217 inclusion and the matrix glass in wt.%.

218 We develop three different scenarios in which the C_{incl} value changes. Our goal is to provide
219 estimates that can be directly compared with similar studies that each follow their own set of
220 assumptions. The first scenario uses the volatile concentrations (S, Cl and F) measured in the melt
221 inclusions (MI) that are closest in composition to the bulk rock, considering that it is the most
222 representative of the volatile content of the system prior to eruption. In the second scenario we use
223 the S, Cl and F concentrations of the melt inclusions which have the same FeO content as the matrix
224 glass. This scenario considers that the best estimate of the pre-eruption volatile content is that of the
225 melt at the same level of differentiation as the matrix glass. The third scenario uses the S, Cl and F
226 average concentrations measured in melt inclusions. This scenario considers that variations in MI

227 volatile content can be viewed as statistical distribution centred on a mean or average which best
228 represents the volatile content of the system prior to eruption. This last scenario is mainly used for
229 comparison purpose as it is the method employed by previous studies of the 1815 Tambora eruption.

230 **Results**

231 **Chemical composition of plagioclases**

232 Plagioclases from TB61 and TB65 samples showed similar compositions (Table S4 in Supplementary
233 Material 1) falling in the range An₇₉-An₉₅ (Fig. 5). This compositional range agrees with the
234 plagioclase cores compositional range of An₈₀-An₉₀ reported by Foden (1986), and that of \leq An₉₁
235 reported by Self et al. (2004) although higher anorthite contents were measured in this study. This
236 range also agrees with the An₄₀-An₉₅ range reported by Suhendro et al. (2021).

237 **Chemical composition of melt inclusions (MI) and matrix glasses (MG)**

238 Major elements

239 Major oxides (SiO₂, TiO₂, Al₂O₃, FeO, MnO, MgO, CaO, Na₂O, K₂O, P₂O₅) of 31 melt inclusions
240 and five matrix glasses were analysed by electron microprobe (Table S5 in Supplementary Material
241 1). Among these 31 inclusions, 11 showed a non-homogeneous texture in SEM, which we interpreted
242 as evidence of post-entrapment crystallization. Only the 20 other, completely glassy melt inclusions
243 are considered in the following.

244 These 20 glassy melt inclusions show homogeneous major element compositions with no
245 significant difference between samples TB61 and TB65 (Table 1). The melt inclusions contained
246 between 52 and 57 wt.% SiO₂, between 18 and 20 wt.% Al₂O₃ and more than 6 wt.% K₂O. Therefore,
247 they plot in the tephri-phonolitic field in a TAS diagram (Na₂O+K₂O vs. SiO₂; Fig. 6). The matrix
248 glasses – being more differentiated and therefore richer in SiO₂ (57-59 wt.%) - lie between phonolite,
249 trachyte and trachyandesite compositions (Fig. 6).

250 The evolution of silica contents in the melt inclusions and matrix glasses highlights the
251 depletion of iron in the magma during differentiation (Fig. 7). CaO, MgO and TiO₂ also show a
252 restricted <1 wt.% decrease.

253 Volatile elements

254 When the volatile element content (S, Cl, and F) in the melt inclusions and matrix glasses is greater
255 than 300 ppm, it can be accurately measured by electron microprobe (e.g., Rose-Koga et al., 2020).
256 The analysis of melt inclusions showed sulfur content ranging from 428±28 to 1118±47 ppm S (Fig.

257 8a), chlorine ranging from 1537 ± 38 to 2253 ± 46 ppm (Fig. 8b) and fluorine ranging from 514 ± 107 to
258 1253 ± 114 ppm (Fig. 8c). Matrix glasses showed, on average, lower volatile content than melt
259 inclusions (Tables 1 and 2). For sulfur, there was a difference of about 750 ppm between the
260 maximum content in the inclusions and the mean content in the matrix glasses (Table 2). For chlorine,
261 this difference was about 800 ppm. For fluorine the difference was less than 600 ppm (Table 2).

262 **Sulfides**

263 Sulfide globules ($3.6-10.3\pm0.2$ μm) were found in melt inclusions and as inclusions in plagioclase
264 (Fig. 3c, d). The composition of one of these sulfides was measured by SEM and contained 65.7 wt.%
265 Fe, 32.7 wt.% S and 1.7 wt.% Cu (no nickel measurements were made), corresponding to an iron-
266 sulfide composition (pyrrhotite; the copper concentration is too low to be considered a copper-
267 sulfide). Sulfide was neither observed in the matrix glass nor reported in previous studies.

268 **Discussion**

269 Syn-eruptive degassing is widely confirmed both by the few eyewitnesses of the eruption and its gas
270 emissions (e.g., de Jong Boers, 1995). Moreover, climate reports associate the abnormally cold
271 temperatures of 1816 with the presence of a volcanic sulphate aerosol in the atmosphere (Stothers
272 1984, Rampino et al. 1988, Briffa et al. 1998, D'Arrigo et al. 2009). The melt inclusions richest in S,
273 Cl and F are also the richest in iron and lowest in SiO_2 and are therefore the least evolved (Fig. 8). In
274 contrast, the matrix glasses are the most evolved end-member and have the lowest volatile content
275 (Table 2, Fig. 8). The difference in concentrations, especially of sulfur and chlorine, between the melt
276 inclusions and the matrix glasses gives a first estimate of the total amount of degassed volatiles.

277 **Volatile degassing budget estimates**

278 Estimates of the quantities of SO_2 , Cl and F degassed are calculated from mass balance equations (1),
279 (2) and (3). For each volatile element, three degassing scenarios are considered.

280 The first scenario uses the melt inclusions with compositions closest to that of the F2 and F4
281 Plinian Fall pumice deposits (Table 3; Sigurdsson and Carey, 1989) as starting point. These melt
282 inclusions have some of the highest S, Cl and F concentrations measured (reported in Table 2)
283 suggesting that they represent the original volatile content of the magma before degassing and
284 concomitant crystallisation. In this scenario, it is assumed that the magma rises and degasses in a
285 closed system. It is therefore considered that all the gases exsolved during ascent and crystallization
286 are conserved in the system and participate in the syn-eruptive degassing.

287 Scenario 2 uses the values of the melt inclusion which have 4.6 wt.% FeO (Table 2). This FeO
288 composition is the one that most closely resembles the composition of the matrix glasses which have

289 an average FeO content of 4.5 wt.% (Table 1). This scenario assumes rising and degassing of the
290 magma in an open system where the exsolved gases can escape before the magma erupts, up until the
291 melt has fully evolved to the composition of the matrix glasses (4.5-4.6 wt.% FeO). Only then does
292 this scenario assumes degassing takes place in a closed system.

293 Scenario 3 uses the average concentrations of melt inclusions (Table 2). It is not associated
294 with any rising/degassing model but corresponds to the approach of Devine et al. (1984) and Self et
295 al. (2004) and will therefore be used mainly for comparison with literature estimates. The amounts
296 of volatile elements degassed depending on the three scenarios during the eruption are reported in
297 Table 4.

298 The scenario we judge most appropriate is scenario 1, as it uses melt inclusion compositions
299 most similar to that of the bulk Plinian Fall deposits (Table 3). The assumption we make therefore, is
300 that from this point onward in the melt compositional evolution, all exsolved volatiles remained in
301 the system until the eruption. In this context, we hypothesize a closed-system scenario for magma
302 evolution and ascent, a supposition supported by the exceptional violence of the 1815 Tambora
303 eruption, which achieved a VEI of 7. Scenario 2 by contrast would make the hypothesis of magma
304 evolution and ascent in an open system, a framework more apt for describing volatile emissions from
305 a slow lava flow originating from an open vent volcano. Scenario 1's approach is the same as the one
306 used by Peccia et al. (in press) to estimate the S emission during the 43 BCE eruption of Okmok
307 volcano. Using scenario 1, SO₂ degassing during the 1815 eruption is estimated at 135±14 Tg (68±7
308 Tg S; Table 4). Also 49±5 Tg of chlorine and 20±2 Tg of fluorine were released (Table 4). These
309 results remain minima for S, as the contribution of sulfides dissolution during the eruption and the
310 possible presence of a deeper gas phase initially present in the magma chamber are not considered in
311 these calculations (Oppenheimer et al. 2011). The pyroclastic flows represent 18±6 km³ DRE of the
312 total magma volume emitted during the 1815 Tambora eruption (Kandlbauer and Sparks 2014).
313 Pyroclastic flows typically only send gases to the troposphere, although large pyroclastic flows such
314 as in the Okmok II eruption can send gas clouds to the stratosphere. Here, if we consider that the PDC
315 only emits sulfur to the troposphere, only 56% of our estimated SO₂ load might have made it to the
316 stratosphere.

317 Even without considering sulfide dissolution and a pre-existing gas phase, these new Tambora
318 estimates are higher than those previously estimated by the "classic" petrological method (Table 5).
319 They are two times higher than the estimates of Self et al. (2004) and four times those of Devine et
320 al. (1984). This can mainly be explained by the fact that these authors used the mean values of the
321 melt inclusions in their calculation. Indeed, their results are closer to our scenario 3 (Table 4). In
322 addition, Self et al (2004) based their calculations on an emitted magma volume of between 30 and

323 33 km³ DRE, which is lower than the 41 km³ DRE used here, hence partially explaining their lower
324 estimations of SO₂ emitted. Devine et al. (1984) and Sigurdsson and Carey (1992) on the other hand,
325 used magma volumes of 87.5 and 51 km³ DRE respectively. The estimate of Legrand and Delmas
326 (1987) based on ice records (SO₂>98 Tg) is closer to our estimates (Table 5) in contrast to the estimate
327 of 45 Tg SO₂ of Sigl et al. (2014), also based on ice core data but using a model calibrated to the
328 quantities of sulfur emitted during the 1991 Pinatubo eruption. Sigl et al. (2014) used the 1991
329 Pinatubo eruption, for which the quantity of SO₂ degassed had been directly measured by satellite, as
330 a single point reference to establish a function linking the quantity of sulfate aerosols found in ice
331 cores to the quantities emitted during an eruption. The estimate of Legrand and Delmas (1987) by
332 contrast is based on the sulfate aerosol deposition flux per km² found in ice cores, which are then
333 converted into a quantity of SO₂ emitted. This discrepancy between estimates derived from ice core
334 data reflects the complexity of volcanic emission reconstruction based on distal sulfate deposits. Our
335 calculated estimate of 135±14 Tg of SO₂ emission aligns remarkably well with the optical depth
336 assessments derived from astronomical observations presented by Stothers et al. (1984). Stothers et
337 al. (1984) estimated a release of 200 Tg of H₂SO₄ into the atmosphere, equivalent to 120 Tg of SO₂,
338 using a compilation of astronomical observations. Their approach encompassed the analysis of cloud
339 shape and colour in locations such as London, New York, and other northern hemisphere regions
340 during the months following the eruption in addition to the observed darkening of the moon during a
341 total lunar eclipse. In contrast to our results, prior estimates of SO₂ release based on petrological
342 analyses by Devine et al. (1984), Sigurdsson et al. (1989), and Self et al. (2014), ranging from 34 to
343 86 Tg are irreconcilable with the optical depth estimation provided by Stothers et al. (1984).

344 **Sulfur degassing and sulfide contribution**

345 Sulfur concentrations measured in lavas or in quench and glassy volcanic products are not necessarily
346 characteristic of their magma source as volatiles degassing and sulfide precipitation or dissolution
347 can occur during ascent and eruption. The presence of sulfides in melt inclusions and in plagioclase crystals
348 indicates that a fraction of the sulfur originally dissolved in the melt partitioned into these sulfides. It also
349 indicates that the magma was saturated in sulfides during the inclusion-trapping stage and that an equilibrium
350 was established between these sulfides and the magma.

351 The sulfur concentration at sulfide saturation (SCSS), was calculated using the parameters of
352 Fortin et al (2015) with a temperature of 950°C (Self et al, 2004) and a pressure of 0.2 GPa for
353 inclusions (Gertisser et al., 2012) and of 0.1 MPa for matrix glasses (i.e., atmospheric pressure; Fig.
354 8). The sulfur contents measured in all the melt inclusions are higher than the theoretical SCSS
355 suggesting that the magma was supersaturated in sulfides. This discrepancy could be due to the fact
356 that water is not accounted for in the calculations. However, the SCSS calculated with 1.5, 2.5, 3.5

357 and 4.5 wt.% H₂O (using the same parameters but only for the MIs) are always lower than the sulfur
358 contents measured in melt inclusions (Fig. 8). Considering more than 4.5 wt.% water in the 1815
359 magma would not be realistic, with water estimates between 1.5 and 4.5 wt.% H₂O (by difference to
360 100% in EMPA analyses, this study) or 2-2.5 wt.% H₂O (Sigurdsson et al., 1989). In addition, we
361 tested whether the presence of copper (present in sulfides) and nickel (not measured but potentially
362 present), could play a role in increasing the SCSS. According to tests carried out using the model of
363 Li and Zhang (2022), the addition of copper and/or nickel would decrease the SCSS. Thus, none of
364 the models tested predicts the observed sulfide saturation in the melt inclusions of this study. This
365 could be related to the fact that these models are calibrated mainly on basaltic compositions far from
366 the tephriphonolitic composition of the 1815 magma.

367 The sulfur concentration at anhydrite saturation (SCAS) line was also calculated (Fig. 8). All
368 our data lies below this line and no sulphate saturation is expected nor observed. This SCAS was
369 calculated according to the parameters of Chowdhury and Dasgupta (2019) and at the same pressures
370 and temperatures as used for the SCSS. The calculations to obtain the SCSS and SCAS lines are
371 presented in Supplementary material 2.

372 In contrast to the melt inclusions, no sulfides were observed in the matrix glass. This absence
373 indicates that during ascent and eruption, sulfides in matrix glass - that were not trapped in the melt
374 inclusions or in crystals - decomposed, and the lower S concentrations in the matrix imply that this
375 additional S was released to the gas phase and contributed to the degassing. The contribution of
376 sulfides was estimated by calculating the relative volume occupied by sulfide globules in melt
377 inclusions. We assume here that sulfides and inclusions are randomly trapped by growing crystals
378 and that their volumetric proportion is hence representative of the melt at the time of entrapment. We
379 surveyed a total of 62 glassy melt inclusions of which 13 were found to contain sulfides. From 2D
380 images we assumed that sulfides and melt inclusions were spherical in shape to calculate the total
381 volume of each. Using our measured sulfide composition, a sulfide density of 4g/cm³ (Robertson et
382 al. 2016) and a total volume of magma emitted of 41±4 km³ DRE (Kandlbauer and Sparks 2014), we
383 estimated the contribution of sulfide dissolution to a total degassing budget of 12±3 Tg SO₂.

384 Considering this, the initial estimate of SO₂ degassing of 135±14 Tg in the first scenario,
385 chosen here, now reaches 147±17 Tg. Placed in the context of the last 2000 years of large eruptions
386 worldwide, these new estimates accounting for sulfide dissolution put the 1815 eruption of Tambora
387 in first place in terms of the amount of SO₂ degassed, higher than the 1257 Samalas eruption (Fig. 9).
388 The amount of SO₂ degassed during the latter is re-estimated at 81±3 Tg. This estimate was
389 recalculated based on data from Vidal et al. (2016) and Métrich et al., (2017), following scenario 1
390 of this study to make it directly comparable. For this, an emitted magma mass of 1.10¹⁴ kg (40 km³

391 DRE) was considered in mass balance equations (1) and (2), as well as a crystal mass fraction of 0.1
392 (average value from Métrich et al. 2017). We used the S content reported for the most primitive
393 inclusions from the 1257 Samalas eruption (500 ppm), which most closely match the composition of
394 the bulk deposit, as well as the S content of matrix glasses (50 ppm). The 1815 Tambora eruption and
395 the 1257 Samalas eruption have erupted similar volumes of magma and have the same VEI. However,
396 the melt compositions differ. The 1815 Tambora melt inclusions have a tephriphonolitic composition
397 and record dissolved S up to 1118 ppm while at the 1257 Samalas melt inclusions have a trachydacitic
398 composition and only record up to 500 ppm of dissolved S. It is therefore unsurprising to find that
399 the 1815 Tambora eruption emitted much larger quantities of S to the atmosphere compared to the
400 1257 Samalas. The same calculation was done for the quantity of chlorine degassed which is
401 estimated at 219 ± 26 Tg.

402 **Halogens degassing and impact on ozone**

403 As chlorine partitions preferentially into the gas phase compared to the melt (e.g., Webster et al.
404 2009a, 2009b; Beermann et al. 2015), the process of gas exsolution effectively depletes the chlorine
405 content of the melt. This chlorine exsolution process is clearly observed in the magmatic inclusions
406 of the 1815 eruption, which show a difference of about 800 ppm between the most primitive and the
407 most differentiated end members (Fig. 8). For fluorine, whose solubility is less well known, the
408 degassing trend is less obvious except for the TB65 samples which seem to show a strong decrease
409 in fluorine content in the matrix glasses (Fig. 8). The release of halogens, especially chlorine, into the
410 stratosphere has impacts on its properties and as a result on climate. Once in the stratosphere,
411 halogenated compounds (in the form of HCl, HF or HBr) induce the catalytic destruction of
412 stratospheric ozone (e.g., Textor et al., 2003; Krüger et al. 2015; Schmidt and Robock, 2015). This
413 can lead to a partial destruction of the ozone layer which protects the Earth's surface from UV
414 radiation, harmful to many lifeforms. Although there are no data after the 1815 eruption to verify this
415 ozone destruction, Vupputuri (1992) made calculations showing that ozone depletion following the
416 eruption may have been as high as 7% but their calculations did not include chlorine nor fluorine and
417 might hence have been underestimated. The first direct observations of ozone depletion in the
418 stratosphere were made following the eruption of El Chichón in 1882 (DeLuisi et al. 1984). Ozone
419 depletion caused by release of halogens during eruptions is thought to be, in general, more important
420 in subduction zones volcanic systems than in hot spot or rifting zones given their magmas are richer
421 in chlorine (e.g., Scaillet et al. 2003). Some eruptions in arc settings however emit very little to no
422 Cl, with an estimated ≈ 0 Tg of Cl and F released during the 43 BCE eruption Okmok eruption for

423 instance (Peccia et al. in press). Our estimated 49 ± 5 Tg Cl and 20 ± 2 Tg F released during the 1815
424 Tambora eruption appear therefore significant.

425 **Conclusions**

426 The 1815 Tambora eruption is well known for its global impact on the climate because of the large
427 quantities of volatile elements released to the stratosphere via plumes 33 and 43 km high (Sigurdsson
428 and Carey 1989). In this study, the analysis of melt inclusions and matrix glasses in the products of
429 the Plinian phases (F2 and F4) of the eruption allow us to estimate the amounts of volatile elements
430 (S, Cl and F) degassed during this eruption. Considering degassing and a magma ascent in a closed
431 system, we estimate that 147 ± 17 Tg of SO_2 (74 Tg S) were degassed as well as 49 ± 5 Tg of Cl and
432 20 ± 2 Tg of F. The SO_2 estimate includes 12 ± 3 Tg coming from the dissolution of sulfides during the
433 eruption. In view of these values, and if comparable methodologies are applied, the 1815 eruption of
434 Tambora appears to be the largest emitter of volcanic SO_2 in the common era. While highly
435 significant, the values obtained in this study could still be considered as minimum values since the
436 contribution of a possible pre-existing exsolved gas phase is not accounted for.

437 **References**

- 438 Barberi S, Bigioggero B, Boriani A, et al (1987) The Island of Sumbawa; a major structural
439 discontinuity in the Indonesian Arc. 547–620
- 440 Beermann O, Botcharnikov RE, Nowak M (2015) Partitioning of sulfur and chlorine between
441 aqueous fluid and basaltic melt at 1050°C, 100 and 200MPa. *Chemical Geology* 418:132–157.
442 <https://doi.org/10.1016/j.chemgeo.2015.08.008>
- 443 Briffa KR, Jones PD, Schweingruber FH, Osborn TJ (1998) Influence of volcanic eruptions on
444 Northern Hemisphere summer temperature over the past 600 years. *Nature* 393:450–455.
445 <https://doi.org/10.1038/30943>
- 446 Chowdhury P, Dasgupta R (2019) Effect of sulfate on the basaltic liquidus and Sulfur Concentration
447 at Anhydrite Saturation (SCAS) of hydrous basalts – Implications for sulfur cycle in subduction
448 zones. *Chemical Geology* 522:162–174. <https://doi.org/10.1016/j.chemgeo.2019.05.020>
- 449 Cooper CL, Swindles GT, Savov IP, et al (2018) Evaluating the relationship between climate change
450 and volcanism. *Earth-Science Reviews* 177:238–247.
451 <https://doi.org/10.1016/j.earscirev.2017.11.009>
- 452 D'Arrigo R, Wilson R, Tudhope A (2009) The impact of volcanic forcing on tropical temperatures
453 during the past four centuries. *Nature Geosci* 2:51–56. <https://doi.org/10.1038/ngeo393>
- 454 de Jong Boers B (1995) Mount Tambora in 1815: A Volcanic Eruption in Indonesia and Its Aftermath.
455 *Indonesia* 60:37. <https://doi.org/10.2307/3351140>
- 456 DeLuisi, J. J., Mateer, C. L., Komhyr, W. D.: 1984, Effects of the E1 Chichón stratospheric aerosol
457 cloud on Umkehr measurements at Mauna Loa, Hawaii, in C. S. Zerefos and A. Ghaz (eds.),
458 *Atmospheric Ozone*, D. Reidel, Dordrecht.
- 459 Devine JD, Sigurdsson H, Davis AN, Self S (1984) Estimates of sulfur and chlorine yield to the
460 atmosphere from volcanic eruptions and potential climatic effects. *J Geophys Res* 89:6309–6325.
461 <https://doi.org/10.1029/JB089iB07p06309>
- 462 Foden J (1986) The petrology of Tambora volcano, Indonesia: A model for the 1815 eruption. *Journal*
463 *of Volcanology and Geothermal Research* 27:1–41. [https://doi.org/10.1016/0377-](https://doi.org/10.1016/0377-0273(86)90079-X)
464 [0273\(86\)90079-X](https://doi.org/10.1016/0377-0273(86)90079-X)
- 465 Foden JD, Varne R (1980) The petrology and tectonic setting of Quaternary—Recent volcanic centres
466 of Lombok and Sumbawa, Sunda arc. *Chemical Geology* 30:201–226.
467 [https://doi.org/10.1016/0009-2541\(80\)90106-0](https://doi.org/10.1016/0009-2541(80)90106-0)
- 468 Fortin M-A, Riddle J, Desjardins-Langlais Y, Baker DR (2015) The effect of water on the sulfur
469 concentration at sulfide saturation (SCSS) in natural melts. *Geochimica et Cosmochimica Acta*
470 160:100–111 <https://doi.org/10.1016/j.gca.2015.03.022>

- 471 Gertisser R, Self S, Thomas LE, et al (2012) Processes and Timescales of Magma Genesis and
472 Differentiation Leading to the Great Tambora Eruption in 1815. *Journal of Petrology* 53:271–
473 297. <https://doi.org/10.1093/petrology/egr062>
- 474 Hamilton WB (1979) *Tectonics of the Indonesian Region*
- 475 Iacovino K, Ju-Song K, Sisson T, et al (2016) Quantifying gas emissions from the “Millennium
476 Eruption” of Paektu volcano, Democratic People’s Republic of Korea/China. *Sci Adv*
477 2:e1600913. <https://doi.org/10.1126/sciadv.1600913>
- 478 Kandlbauer J, Sparks RSJ (2014) New estimates of the 1815 Tambora eruption volume. *Journal of*
479 *Volcanology and Geothermal Research* 286:93–100.
480 <https://doi.org/10.1016/j.jvolgeores.2014.08.020>
- 481 Krüger K, Kutterolf S, Hansteen TH (2015) Halogen release from Plinian eruptions and depletion of
482 stratospheric ozone. In: Schmidt A, Fristad KE, Elkins-Tanton LT (eds) *Volcanism and Global*
483 *Environmental Change*, 1st edn. Cambridge University Press, pp 244–259
- 484 Le Voyer M (2009) *Rôle des fluides dans la genèse des magmas d’arc : analyses in situ des éléments*
485 *volatils et des isotopes du bore dans les inclusions magmatiques des olivines primitives.*
486 *Université Blaise Pascal - Clermont Ferrand II*
- 487 Legrand M, Delmas RJ (1987) A 220-year continuous record of volcanic H₂SO₄ in the Antarctic ice
488 sheet. *Nature* 327:671–676. <https://doi.org/10.1038/327671a0>
- 489 Li H, Zhang L (2022) A thermodynamic model for sulfur content at sulfide saturation (SCSS) in
490 hydrous silicate melts: With implications for arc magma genesis and sulfur recycling.
491 *Geochimica et Cosmochimica Acta* 325:187–204. <https://doi.org/10.1016/j.gca.2022.03.008>
- 492 Mandeville CW, Carey S, Sigurdsson H (1996) Magma mixing, fractional crystallization and volatile
493 degassing during the 1883 eruption of Krakatau volcano, Indonesia. *Journal of Volcanology and*
494 *Geothermal Research* 74:243–274. [https://doi.org/10.1016/S0377-0273\(96\)00060-1](https://doi.org/10.1016/S0377-0273(96)00060-1)
- 495 Métrich N, Vidal CM, Komorowski J-C, et al (2017) New Insights into Magma Differentiation and
496 Storage in Holocene Crustal Reservoirs of the Lesser Sunda Arc: the Rinjani–Samalas Volcanic
497 Complex (Lombok, Indonesia). *Journal of Petrology* 58:2257–2284.
498 <https://doi.org/10.1093/petrology/egy006>
- 499 Moussallam Y, Oppenheimer C, Scaillet B, et al (2014) Tracking the changing oxidation state of
500 Erebus magmas, from mantle to surface, driven by magma ascent and degassing. *Earth and*
501 *Planetary Science Letters* 393:200–209. <https://doi.org/10.1016/j.epsl.2014.02.055>
- 502 Newhall CG, Self S (1982) The volcanic explosivity index (VEI) an estimate of explosive magnitude
503 for historical volcanism. *J Geophys Res* 87:1231. <https://doi.org/10.1029/JC087iC02p01231>

- 504 Oppenheimer C (2003) Climatic, environmental and human consequences of the largest known
505 historic eruption: Tambora volcano (Indonesia) 1815. *Progress in Physical Geography: Earth and*
506 *Environment* 27:230–259. <https://doi.org/10.1191/0309133303pp379ra>
- 507 Oppenheimer C, Scaillet B, Martin RS (2011) Sulfur Degassing From Volcanoes: Source Conditions,
508 Surveillance, Plume Chemistry and Earth System Impacts. *Reviews in Mineralogy and*
509 *Geochemistry* 73:363–421. <https://doi.org/10.2138/rmg.2011.73.13>
- 510 Peccia A, Moussallam Y, Plank T (2023) Melt inclusion and matrix glass data from the 43 BCE
511 eruption of Okmok Volcano, in press.
- 512 Rampino MR, Self S, Stothers RB (1988) Volcanic Winters. *Annu Rev Earth Planet Sci* 16:73–99.
513 <https://doi.org/10.1146/annurev.ea.16.050188.000445>
- 514 Reubi O, Nicholls IA (2004) Magmatic evolution at Batur volcanic field, Bali, Indonesia: petrological
515 evidence for polybaric fractional crystallization and implications for caldera-forming eruptions.
516 *Journal of Volcanology and Geothermal Research* 138:345–369.
517 <https://doi.org/10.1016/j.jvolgeores.2004.07.009>
- 518 Robertson JC, Barnes SJ, Le Vaillant M (2015) Dynamics of Magmatic Sulphide Droplets during
519 Transport in Silicate Melts and Implications for Magmatic Sulphide Ore Formation. *J Petrology*
520 56:2445–2472. <https://doi.org/10.1093/petrology/egv078>
- 521 Rose-Koga EF, Koga KT, Devidal J-L, et al (2020) In-situ measurements of magmatic volatile
522 elements, F, S, and Cl, by electron microprobe, secondary ion mass spectrometry, and heavy ion
523 elastic recoil detection analysis. *American Mineralogist* 105:616–626.
524 <https://doi.org/10.2138/am-2020-7221>
- 525 Scaillet B, Cl  mente B, Evans BW, Pichavant M (1998) Redox control of sulfur degassing in silicic
526 magmas
- 527 Schmidt A, Carn S (2022) Volcanic emissions, aerosol processes, and climatic effects. In: *Aerosols*
528 *and Climate*. Elsevier, pp 707–746
- 529 Schmidt A, Robock A (2015) Volcanism, the atmosphere and climate through time. In: *Volcanism and*
530 *Global Environmental Change*. Cambridge University Press, pp 195–207
- 531 Self S (2004) Magma volume, volatile emissions, and stratospheric aerosols from the 1815 eruption
532 of Tambora. *Geophys Res Lett* 31:L20608. <https://doi.org/10.1029/2004GL020925>
- 533 Sigl M, McConnell JR, Toohey M, et al (2014) Insights from Antarctica on volcanic forcing during
534 the Common Era. *Nature Clim Change* 4:693–697. <https://doi.org/10.1038/nclimate2293>
- 535 Sigurdsson H, Carey S (1989) Plinian and co-ignimbrite tephra fall from the. *Bull Volcanol* 51:243–
536 270. <https://doi.org/10.1007/BF01073515>

- 537 Sigurdsson H, Carey S (1992) The Eruption of Tarnbora in 1815: Environmental Effects and Eruption
538 Dynamics. In Harington, I.R., editor, The year without a summer? World climate in 1816.
539 Ottawa: Canadian Museum of Nature, 16–45
- 540 Stothers RB (1984) The Great Tambora Eruption in 1815 and Its Aftermath. *Science* 224:1191–1198.
541 <https://doi.org/10.1126/science.224.4654.1191>
- 542 Suhendro I, Toramaru A, Miyamoto T, et al (2021) Magma chamber stratification of the 1815
543 Tambora caldera-forming eruption. *Bull Volcanol* 83:63. [https://doi.org/10.1007/s00445-021-](https://doi.org/10.1007/s00445-021-01484-x)
544 [01484-x](https://doi.org/10.1007/s00445-021-01484-x)
- 545 Textor C, Graf HF, Herzog M, Oberhuber JM (2003) Injection of gases into the stratosphere by
546 explosive volcanic eruptions. *Journal of Geophysical Research: Atmospheres*. 16;108(D19).
- 547 Thordarson T (2003) Atmospheric and environmental effects of the 1783–1784 Laki eruption: A
548 review and reassessment. *J Geophys Res* 108:4011. <https://doi.org/10.1029/2001JD002042>
- 549 Vidal CM, Métrich N, Komorowski J-C, et al (2016) The 1257 Samalas eruption (Lombok,
550 Indonesia): the single greatest stratospheric gas release of the Common Era. *Sci Rep* 6:34868.
551 <https://doi.org/10.1038/srep34868>
- 552 Vupputuri RKR (1992) The Tambora eruption in 1815 provides a test on possible global climatic and
553 chemical perturbations in the past. *Nat Hazards* 5:1–16. <https://doi.org/10.1007/BF00127136>
- 554 Webster JD, Sintoni MF, De Vivo B (2009a) The partitioning behavior of Cl, S, and H₂O in aqueous
555 vapor- ±saline-liquid saturated phonolitic and trachytic melts at 200 MPa. *Chemical Geology*
556 263:19–36. <https://doi.org/10.1016/j.chemgeo.2008.10.017>
- 557 Webster JD, Tappen CM, Mandeville CW (2009b) Partitioning behavior of chlorine and fluorine in
558 the system apatite–melt–fluid. II: Felsic silicate systems at 200MPa. *Geochimica et*
559 *Cosmochimica Acta* 73:559–581. <https://doi.org/10.1016/j.gca.2008.10.034>

560 **Figure captions**

561 **Fig. 1** Map of the Sunda Arc, showing the tectonic setting and the distribution of volcanoes, modified
562 from Gertisser et al. (2012). The enlarged map shows the location of Mount Tambora on Sumbawa
563 island.

564 **Fig. 2** Stratigraphic log of the deposits of the 1815 eruption at 25 km west of the volcano, modified
565 from Sigurdsson and Carey (1989).

566 **Fig. 3 a** Plagioclase crystal with zoned rims (TB65_pl15), **b** Unzoned plagioclase crystal
567 (TB65_pl11), **c** Sulfide globule in a plagioclase-hosted melt inclusion (TB65_pl11), **d** Sulfide globule
568 in a plagioclase crystal.

569 **Fig. 4 a** Glassy, oval-shaped melt inclusions with a shrinkage bubble (TB61_pl19 MI26&27), **b** Melt
570 inclusion containing a sulfide globule (TB61_pl15 MI21), **c** Glassy melt inclusion with several
571 bubbles (TB61_pl5 MI9), **d** Melt inclusion partially crystallized (TB61_pl16 MI10).

572 **Fig. 5** Feldspar ternary diagram showing the average concentrations of plagioclases in the 1815 tephra
573 from Tambora and data (average) from Samalas and Batur, volcanoes of the same arc.

574 **Fig. 6** Total alkali vs silica plot of average alkali concentrations of melt inclusions (MI) and matrix
575 glasses (MG) from four studies (error bars are 1σ standard deviations). All data are normalised to
576 100%.

577 **Fig. 7** Trends of major element concentrations in the Tambora magma during its differentiation. All
578 data are normalised. Grey squares are the bulk rock compositions of the F2 and F4 layers from
579 Sigurdsson and Carey (1989). The blue squares are from Suhendro et al. (2021).

580 **Fig. 8** Trends of volatile element concentrations as a function of FeO (error bars are 1σ standard
581 deviation). The lines are the SCSS (Fortin et al., 2015) with 0, 2.5 and 4.5 wt.% H₂O and the SCAS
582 (Chowdhury and Dasgupta, 2019) (1.5 and 3.5 wt.% H₂O are not represented for reasons of clarity).
583 The vertical dotted line is the average FeO content of the bulk rocks of F2 and F4 layers from
584 Sigurdsson and Carey (1989). All data are normalised to 100%.

585 **Fig. 9** Quantities of volatiles degassed during the major eruptions of the last two millennia, modified
586 from Vidal et al. (2016). On the Tambora estimates are included the results of previous studies: A
587 Devine et al. (1984), B Self et al. (2004), C Sigurdsson and Carey (1992). The different methods of
588 estimate are given in Table S6 in Supplementary Materials 1.

589 **Table captions**

590 **Table 1** Major elements (wt.%) and volatile (ppm) contents of melt inclusions and matrix glasses.

591 **Table 2** Summary of volatile concentrations (ppm) in melt inclusions and matrix glasses (with 1σ
592 standard deviation).

593 **Table 3** Major element composition of the melt inclusions with maximum S and F contents, second
594 highest Cl content (with 1σ standard deviation) and average major element composition of the F2 and
595 F4 bulk rocks from Sigurdsson and Carey (1989) and Suhendro et al. (2021).

596 **Table 4** Volatile quantities emitted during the 1815 eruption of Tambora according to the three
597 scenarios (Tg). The total error on the result takes into account the error on the volume of magma
598 emitted, on the fraction of crystals and on the accuracy of the microprobe analyses. The respective
599 errors of each parameter in equation (1) were propagated through the different calculation according

600 to $\sigma_x = \sqrt{\sigma_a^2 + \sigma_b^2}$ if $x = a \pm b$ and to $\frac{\sigma_x}{x} = \sqrt{\left(\frac{\sigma_a}{a}\right)^2 + \left(\frac{\sigma_b}{b}\right)^2}$ if $x = a / \times b$.

601 **Table 5** Estimates of the SO₂ amount degassed during the 1815 eruption of Tambora according to
602 different method and studies.

603

	Mean TB61 (n=12)		Mean TB65 (n=12)	
SiO₂	47.05	(± 1.47)	47.14	(± 0.44)
MgO	0.04	(± 0.01)	0.05	(± 0.01)
FeO	0.55	(± 0.07)	0.59	(± 0.06)
Na₂O	1.76	(± 0.62)	1.88	(± 0.17)
Al₂O₃	33.13	(± 1.06)	33.2	(± 0.29)
K₂O	0.19	(± 0.12)	0.18	(± 0.03)
CaO	16.58	(± 1.16)	16.42	(± 0.25)
Total	99.3		99.46	
% An	88.9	(± 4.38)	88.39	(± 1.13)

	Melt Inclusions			Matrix Glasses		
	TB61 (n=11)	TB65 (n=12)	Mean	TB61 (n=4)	TB65 (n=2)	Mean
SiO₂	51.67 (± 0.90)	54.39 (± 0.80)	54.54 (± 0.84)	57.95 (± 0.39)	57.17 (± 0.11)	57.69 (± 0.50)
MgO	1.61 (± 0.22)	1.52 (± 0.15)	1.57 (± 0.19)	1.41 (± 0.02)	1.3 (± 0.01)	1.37 (± 0.06)
FeO	5.11 (± 0.63)	4.82 (± 0.17)	4.98 (± 0.49)	4.49 (± 0.23)	4.47 (± 0.12)	4.48 (± 0.19)
Na₂O	5.21 (± 0.16)	5.28 (± 0.22)	5.24 (± 0.19)	5.41 (± 0.21)	5.52 (± 0.28)	5.45 (± 0.21)
Al₂O₃	18.9 (± 0.39)	19.28 (± 0.25)	19.07 (± 0.38)	19.94 (± 0.24)	19.84 (± 0.07)	19.9 (± 0.19)
K₂O	6.27 (± 0.22)	6.27 (± 0.32)	6.27 (± 0.26)	6.24 (± 0.24)	6.41 (± 0.02)	6.3 (± 0.21)
CaO	3.29 (± 0.38)	3.28 (± 0.51)	3.29 (± 0.43)	3.27 (± 0.11)	3.15 (± 0.03)	3.23 (± 0.10)
TiO₂	0.74 (± 0.13)	0.74 (± 0.05)	0.74 (± 0.10)	0.53 (± 0.03)	0.5 (± 0.02)	0.52 (± 0.03)
P₂O₅	0.56 (± 0.18)	0.61 (± 0.11)	0.58 (± 0.15)	0.37 (± 0.05)	0.46 (± 0.01)	0.4 (± 0.06)
MnO	0.18 (± 0.06)	0.22 (± 0.05)	0.19 (± 0.06)	0.15 (± 0.18)	0.18 (± 0.03)	0.16 (± 0.14)
Total	96.52	96.41	96.47	99.76	99.01	99.51
S	775 (± 214)	775 (± 142)	775 (± 181)	369 (± 46)	354 (± 16)	364 (± 37)
Cl	1986 (± 1986)	2052 (± 213)	2016 (± 219)	1725 (± 101)	1756 (± 35)	1735 (± 81)
F	790 (± 132)	854 (± 196)	819 (± 163)	636 (± 83)	779 (± 90)	683 (± 106)

	Melt Inclusions			Matrix Glasses
	Mean	Maximum	At 4.6 wt.% FeO	Mean
S	775 (±37)	1118 (±47)	586 (±32)	364 (±37)
CI	2016 (±42)	2553 (±37)	1975 (±42)	1735 (±81)
F	819 (±107)	1253 (±114)	840 (±104)	683 (±106)

	Melt inclusions used for						Bulk rocks			
	S in Scenario 1 (=1118 ppm)		Cl in Scenario 1 (=2282)		F in Scenario 1 (=908 ppm)		Sigurdsson and Carey (1989)		Suhendro et al. (2021)	
SiO₂	54.36	(±0.18)	54.34	(±0.17)	55.72	(±0.17)	54.57	(±0.39)	58.2	(±0.19)
TiO₂	0.76	(±0.02)	0.79	(±0.02)	0.91	(±0.02)	0.64	(±0.01)	0.68	(±0.01)
Al₂O₃	18.14	(±0.07)	18.73	(±0.07)	19.60	(±0.07)	19.14	(±0.23)	18.57	(±0.06)
FeO	5.68	(±0.15)	5.65	(±0.15)	5.82	(±0.15)	6.05	(±0.07)	5.94	(±0.11)
MnO	0.24	(±0.05)	0.19	(±0.06)	0.08	(±0.06)	0.20	(±0.00)	0.19	(±0.00)
MgO	1.82	(±0.03)	1.74	(±0.02)	1.92	(±0.03)	1.42	(±0.12)	2.06	(±0.05)
CaO	3.38	(±0.04)	3.85	(±0.04)	3.87	(±0.04)	4.23	(±0.31)	3.87	(±0.13)
Na₂O	4.91	(±0.06)	5.06	(±0.06)	5.30	(±0.06)	3.75	(±1.39)	4.46	(±0.07)
K₂O	6.17	(±0.07)	6.04	(±0.07)	6.27	(±0.07)	5.83	(±0.09)	5.85	(±0.10)
P₂O₅	0.64	(±0.03)	0.65	(±0.03)	0.51	(±0.03)	0.42	(±0.02)	0.32	(±0.01)
Total	96.09		97.02		94.75		96.23		100.14	

	Scenario 1	Scenario 2	Scenario 3
S	135±14	40±4	74±7
Cl	49±5	22±2	25±3
F	20±4	14±2	12±2

Method	SO₂ degassed (Tg)	Reference
Petrological	34	Devine et al., 1984
	53-58	Self et al., 2004
	86	Sigurdsson et al., 1992
	147±17	This study
Ice cores	> 98	Legrand et Delmas, 1987
	45	Sigl et al., 2014

















