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Petrological insights on the last 1000 years of explosive activity at La Soufrière volcano, St. Vincent (Lesser Antilles)

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14 Abstract

15 The petrological features of scoria samples and enclosed holocrystalline xenoliths from the last 16 ~1000 years of activity of the La Soufrière volcano of St. Vincent (Lesser Antilles) are described in 17 detail here. The products of the investigated prehistoric eruptions (pre-1440, 1440, 1580 CE) are 18 relatively homogeneous basaltic andesites (with a paragenesis consisting of plagioclase, 19 clinopyroxene, orthopyroxene and opaques) commonly hosting gabbroic xenoliths. Scoria samples 20 from the historical eruptions (1718-1812 and 1902-03 CE) are mostly similar to the former in terms 21 of whole-rock composition, mineral chemistry and types of xenoliths, but include also a mafic-rich 22 scoria type (plagioclase+clinopyroxene+olivine) in the products of the 1902-03 eruption that is less 23 evolved in composition and encloses ol-gabbro and ultramafic xenoliths. Both scoria types were 24 likely affected by crystal contamination and accumulation processes, as indicated by their moderately 25 to strongly porphyritic textures and large compositional variation of the crystal populations. 26 Recycling of xenocryst, protocryst and/or antecryst phases, however, did not significantly modify the 27 original magma composition, as suggested by the overall consistency of whole-rock differentiation 28 trends with petrography, mineral chemistry, xenolith types and glass compositions (within both 29 inclusions and the groundmass) and experimental petrology observations, crystal fractionation 30 therefore still appearing as the main evolutionary process. Most of the gabbro and ol-gabbro xenoliths 31 have mineral compositions in line with that of the coexisting phenocrysts and textural features 32 consistent with an intrusive origin, i.e., representing fully-crystallised melt aliquots. Conversely, 33 ultramafic, troctolitic and noritic xenoliths (plus some ol-gabbro) are likely representing cumulitic 34 assemblages, i.e., mineral phases removed from the magma by crystal fractionation. The typical association of An-rich plagioclase and relatively Fo-poor olivine of the troctolite (and ol-gabbro) cumulates is ascribed to crystallisation from a hydrous (H₂O ~4.5-6 wt.%) high-Al₂O₃ basalt/basaltic andesite magma crystallising at shallow depths (~0.2 GPa, i.e., at ~7 km). On the other hand, the ultramafic cumulitic xenoliths are thought to derive from a less hydrous basalt magma stalling at deeper levels (~0.4 GPa, ~13 km). The tapping of such magma could mark a major rejuvenation event, which is the process that likely triggered the 1902-03 eruption.

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42 keywords: La Soufrière; St. Vincent; Lesser Antilles; scoria clasts; xenoliths; magmatic system

43

44 **1. Introduction**

The Lesser Antilles volcanic arc has been historically regarded as a reference-type location for intraoceanic volcanic arc magmatism, whose study has shed significant light on the geological processes occurring at subduction zones (e.g., Macdonald et al., 2000; Syracuse et al., 2010; Bebout, 2014; Jicha and Jagoutz, 2014). An additional element of interest for the Earth sciences community is represented by the seismic and volcanic activity presently characterizing the arc (e.g., Lindsay et al., 2005; Feuillet et al., 2011; Lindsay and Robertson, 2018), which constitute a serious source of hazard for the region.

52 Within the Lesser Antilles framework, magmatism on the island of St. Vincent is of particular 53 interest due to a number of factors, including: i) five major historical eruptions, the latest occurring 54 in 1979 (Shepherd et al., 1979; Graham and Thirlwall, 1981; Poret et al., 2017), and renewed activity in December 2020; ii) predominant eruption of weakly evolved magmas (up to primitive 55 56 compositions with MgO ~13.5 wt.%; Heath et al., 1998a); iii) whole-rock geochemistry and Sr-Nd 57 and U-Th isotope systematics resembling that of the northern Lesser Antilles islands (e.g., Turner et 58 al., 1996; Heath et al., 1998a; Toothill et al., 2007). As a consequence, in the last ~20 years or so 59 numerous studies have investigated the petrology of St. Vincent magmatic rocks, including works on 60 "classical" whole-rock and mineral chemistry characterization (Heath et al. 1998a), ²³⁸U-²³⁰Th 61 disequilibria (Heath et al. 1998b), olivine-hosted melt inclusion (Bouvier et al., 2008, 2010) and 62 experimental phase equilibria determinations (Pichavant et al., 2002b; Pichavant and Macdonald, 63 2007; Melekhova et al., 2015). Nevertheless, these have focused mainly on the earliest phases of 64 activity (mostly covering the ~690-1.3 ka age interval), whereas the latest prehistoric and historical eruptions were less investigated. 65

In addition, St. Vincent rocks are known to host a large variety of xenoliths (e.g., Tollan et al., 2012), whose study gives the opportunity to obtain additional insights about the lithology of and the processes taking place within the crustal storage system (or even about the mantle sources) beneath 69 the island. Juvenile products (i.e., both lava and pyroclasts), indeed, represent only the final product 70 of the various differentiation processes acting in the crustal reservoir(s). Therefore, data on juvenile 71 material and xenoliths can be considered as complementary archives of information regarding near-72 surface, syn-eruptive, and (variable) crustal level, pre-eruptive processes.

73 The products of the last ~1000 years of activity of the La Soufrière volcano of St. Vincent 74 (referring to the historical eruptions of 1902-03 and 1718-1812, and the prehistoric eruptions of 1580 75 CE and 1440 CE) have been recently investigated by Cole et al. (2019). Here we provide a more 76 detailed petrological characterization and also present for the first time new data including: i) whole-77 rock major- and trace element composition for juvenile clasts from older undated eruptions (i.e., pre-78 1440); ii) whole-rock trace element concentrations (including Rare Earth Elements) for all the 79 juvenile clasts; iii) mineral and glass microanalyses for juvenile products (tripling the previous set of 80 available data); iv) mineral microanalyses for xenoliths embedded within the investigated juvenile 81 clasts. This expands the database on magma processes operating in St. Vincent and contributes to an 82 improved assessment of the magma dynamics driving the volcanic system. It also has positive 83 repercussions on the accuracy of models for hazard assessment and on the planning of monitoring 84 activities.

85

86 2. Geological background

87 2.1. The Lesser Antilles volcanic arc

88 The Lesser Antilles is a 750 km-long intraoceanic volcanic arc developed as a result of the 89 relatively slow subduction (~2-4 cm/year; 1986; e.g., DeMets et al., 2000) of the Atlantic/North American plate beneath the Caribbean plate (e.g., Macdonald et al., 2000; Fig. 1a). The original 90 91 volcanic arc (the so-called "Proto-arc" or "Mesozoic Caribbean arc"; Bouysse et al., 1990), active 92 from Lower Cretaceous/Upper Turonian to Lower Palaeocene, is likely to be presently represented 93 by the Aves Ridge (or Aves Swell), a mostly submerged bathymetric high west of the active arc 94 (Macdonald et al., 2000 and references therein). The present Lesser Antilles arc developed above the 95 accretionary prism of the Aves Ridge system staring from Late Oligocene (~25-20 Ma; Germa et al., 96 2011). Volcanism was almost continuous in the islands of the southern sector, whereas in the northern 97 sector a distinct westward jump (i.e., away from the trench) occurred ~5.5 Ma (Germa et al., 2011), 98 from the older, currently inactive Limestone Caribees chain (the "Old arc"), to the younger currently 99 active islands of the Volcanic Caribees chain ("Recent arc"; Fig. 1a).

100 The juvenile products of the Lesser Antilles are predominantly basalts to andesites (Macdonald et 101 al., 2000). Islands of the northern (i.e., from Saba to Montserrat, north to south) and central sectors 102 (from Guadeloupe to St. Lucia) are dominated by andesites, with low volumes of basalt and dacite

103 and rare rhyolite, while in the islands of the southern sector (St. Vincent, the Grenadines and Grenada) 104 andesite is subordinate with respect to less evolved rocks (Macdonald et al., 2000). The latter are 105 represented by basalt and basaltic andesite at St. Vincent, Mg-rich, silica-undersaturated picrobasalt and ankaramitic basalt (i.e., porphyritic melanocratic basalt) at the Grenadines and Grenada (giving 106 107 rise to the MgO-richer M-series and the CaO-richer C-series, respectively; e.g., Macdonald et al., 108 2000; Heath et al., 1998a; Stamper et al., 2014a, 2014b; Camejo-Harry et al., 2018). A similar regular 109 distribution has also been reported for rock serial affinity (e.g., Macdonald et al., 2000 and references 110 therein): 1) low-K tholeiitic for the northern islands; 2) low/medium-K calcalkaline in the central 111 islands; 3) medium-K calcalkaline in the southern islands. However, the serial affinity of the Lesser 112 Antilles rocks, and especially that of St. Vincent, has been the subject of debate (e.g., Smith et al., 113 1996; Heath et al., 1998a; Macdonald et al., 2000 and references therein). Moreover, it has become 114 increasingly evident that the along-arc trends are an oversimplification, with large compositional 115 variations at single volcanic centres (especially in the central sectors; e.g., Davidson and Wilson, 2011). 116

117 Nevertheless, the reported chemical variabilities have suggested some kind of relation with the 118 structural features of the arc, as indicated by the segmentation of the Benioff zone (Wadge and 119 Shepherd, 1984). This is also reflected in the isotopic composition of the emplaced products, 120 displaying a homogeneous depleted MORB-like signature in the north, whereas the larger variability 121 observed in the central and southern islands is suggestive of a stronger involvement of a crustal 122 component (recycled in the mantle source or/and contaminating ascending magmas; e.g., Macdonald 123 et al., 2000; Toothill et al., 2007; Carpentier et al., 2008; Bezard et al., 2014; Allen et al. 2019).

124 As a final remark, the volcanic products from all islands of the Lesser Antilles arc (possibly with 125 the only exception of Nevis and Guadeloupe) are commonly characterised by cumulate-textured 126 xenoliths (from 1 up to 50 cm in diameter; Arculus and Wills, 1980). These include adcumulitic, 127 heteradcumulitic and orthocumulitic varieties, and occasionally feature rhythmic igneous layering, 128 slump structures and auto-intrusive features such as plagioclase-rich veins (Macdonald et al., 2000; 129 Kiddle et al., 2010; Tollan et al., 2012; Stamper et al., 2014a; Cooper et al., 2016, 2019; Melekhova 130 et al., 2017, 2019; Camejo-Harry et al., 2018). Plagioclase and amphibole are generally the most abundant mineral phases, except in Grenada, where clinopyroxene and amphibole±olivine prevail 131 132 (with few or no plagioclase; Arculus and Wills, 1980; Stamper et al., 2014a).

133

134 **2.2.** The island of St. Vincent and the La Soufrière volcano

Volcanic activity at St. Vincent is suggested to have started much more recently with respect to
the neighbouring islands, as St. Vincent lacks exposed Miocene rocks (Heath et al., 1998a), unlike St

Lucia, Martinique, Grenada and many of the Grenadine islands. The island of St. Vincent is 137 138 dominated by the La Soufrière volcano, a 1204 m high active stratovolcano rising at the northernmost 139 edge of the island (Fig. 1b) as the product of the most recent phase of activity (<0.69 Ma, K-Ar ages; 140 Briden et al., 1979). Older pre-La Soufrière activity occurred at several other volcanic centres, the 141 remnants of which are found in the central (Morne Garu and Grand Bonhomme volcanic centres) and 142 southern sectors (the "South-east volcanic centres"), defining a crude N-S increase of eruption ages 143 from ~0.69 to 2.74 Ma (K-Ar; Briden et al., 1979). Knowledge of the pre-La Soufrière activity is 144 scant, mainly due to poor exposure that characterise the central and southern sectors of the island 145 (Robertson, 2005). Volcanic sequences are made of alternations of weakly evolved pyroclastic 146 deposits and lava flows (plus minor domes and some intruding dykes), commonly overlain by fine-147 grained yellow ash deposits (Robertson, 2005).

The volcanic history of the La Soufrière has been roughly subdivided into four main phases of activity by Sigurdsson and Carey (1991), resulting in the deposition of the Pre-Somma Lavas Formation (0.69 Ma - 10 ka), the Yellow Tuff Formation (~14 ka), the Crater Lavas Formation and finally the Pyroclastic Formation (dating back to at least 5 ka, ¹⁴C ages; see Robertson, 2005). The lack of clear field relations and precise absolute dates for periods >5 ka results in considerable uncertainty about the stratigraphy of this period.

154 La Soufrière has been the site of at least five main historical eruptions (dated 1718, 1812, 1902-155 03, 1971 and 1979) during which basaltic andesite domes (in the crater) and pyroclastic density 156 current deposits were emplaced, along with subordinate fallout components (Aspinall et al., 1973; Shepherd et al., 1979; Graham and Thirlwall, 1981; Pyle et al., 2018; Cole et al., 2019). Recently, 157 158 two major prehistoric eruptions predating the 1718 event were identified by Cole et al. (2019), who 159 reported ¹⁴C ages pointing to 1440 and 1580 CE. On this basis, the authors proposed for the last ~600 160 years of activity at La Soufrière a crude periodicity with repose periods ranging between 77 and ~140 161 years and systematically decreasing in more recent times. A new eruption began on 27th December 162 2020 involving the passive effusion of a lava dome.

163

164 2.2.1. Petrology of juvenile products and xenoliths at St. Vincent

165 Rock compositions at St. Vincent range from basalt (both microphyric, with 166 olivine+spinel±clinopyroxene microphenocrysts, and coarsely porphyritic with 167 plagioclase+clinopyroxene+olivine+spinel phenocrysts) to basaltic andesite (strongly porphyritic 168 with phenocrysts of plagioclase+clinopyroxene+orthopyroxene+Ti-magnetite±olivine), and fewer 169 and esite (only as a higher-SiO₂ component of rocks displaying evidence for magma mixing; Heath et 170 al., 1998a). According to Heath et al. (1998a), the products from the oldest Pre-Somma Lavas and

171 Yellow Tuff formations span a wide compositional range, including the MgO-richest concentrations 172 (i.e., from ~2 up to 10-13.5 wt.%), whereas the rocks of the youngest Crater Lavas and Pyroclastic 173 formations are more homogeneous and evolved (e.g., MgO ~2-6 wt.%, only occasionally up to ~8 174 wt.%). Time-related geochemical gradients have also been reported within single eruptions, like that 175 of 1902-03, for which the existence of a zoned magma chamber was suggested (Roobol and Smith, 176 1975; Cole et al., 2019). The products of the historical eruptions of 1902-03 and 1979 bear also 177 evidence for mixing phenomena (i.e., banded and mingled juvenile clasts) between basaltic andesite 178 and dacite magmas (Carey and Sigurdsson, 1978; Graham and Thirlwall, 1981).

179 The evolution of St. Vincent primitive magmas has been investigated by whole-rock and mineral 180 petrochemical studies (Heath et al., 1998a) and by experimental works at various P and H₂O content 181 conditions (with NNO + 1-4 for oxygen fugacity; Pichavant et al., 2002b; Pichavant and Macdonald, 182 2007; Melekhova et al., 2015). The results indicate that differentiation processes were notably 183 complex, driven by polybaric fractionation (from 1.3 to <0.4 GPa) starting from primitive, variably 184 hydrated high-MgO basalts (HMB, MgO ≥ 10 wt.%), giving rise to at least four different liquid lines 185 of descent. High-level differentiation (1.3-1.0 GPa) of H₂O-rich (2.3-4.5 wt.%) magmas produced low-MgO basalts (LMB, 6 wt.% < MgO < 10 wt.%), whereas lower-P (1.0-0.7 GPa) differentiation 186 187 of H₂O-poorer magmas (0.6-2.3 wt.%) generated basaltic andesites. The fewer high-Al₂O₃ basalts 188 (HAB, MgO <6 wt.%, Al₂O₃ >19 wt.%) and andesites are ascribed respectively to i) shallow-depth 189 differentiation (<0.4 GPa) of primitive magmas with high H₂O (>4 wt.%, delaying plagioclase 190 crystallisation), and ii) partial melting of water-poor (H₂O <0.6 wt.%) HMB that solidified at depth 191 (~1 GPa) to a hornblende gabbro. In addition, high-CaO (>12 wt.%) low-MgO basalt compositions, 192 reported only for La Soufrière olivine-hosted melt inclusions (Bouvier et al., 2008), can be obtained 193 through low-P differentiation (≤ 0.7 GPa), preventing the early crystallisation of clinopyroxene.

194 Additional information about magma evolution has been obtained from the abundant xenoliths 195 hosted in St. Vincent lavas and pyroclasts, mainly represented by holocrystalline gabbros, troctolites 196 and rarer hornblende-clinopyroxenites (plus much lesser metavolcanic and calc-silicate sedimentary 197 lithotypes; Heath et al., 1998a; Tollan et al., 2012). Experimental studies, in accordance with 198 geothermobarometric and isotopic evidence on natural samples (Tollan et al., 2012), suggest that 199 these xenoliths are the product of shallow level fractional crystallisation of (relatively water-rich) 200 differentiated magmas deriving from the evolution of primary basaltic melts. This is supported by the 201 peculiar composition of olivine (Fo-poor) and plagioclase (remarkably An-rich), the scarcity of 202 orthopyroxene and the relative abundance of amphibole, never observed in the paragenesis of St. 203 Vincent lavas and pyroclasts (Tollan et al., 2012).

204

205 **3. Analytical techniques**

206 Single scoria samples from the main outcropping units of the La Soufrière volcano of St. Vincent 207 have been analysed in this study. Sample location and stratigraphy of the deposits are given in Cole 208 et al. (2019) for the samples of the 1902-03 (n = 14) and 1718-1812 (6) historical eruptions and of 209 the 1580 CE (6) and 1440 CE (4) prehistoric eruptions. In addition, 2 single scoria samples were 210 collected from pyroclastic density current deposits in lower stratigraphic position with respect to the 211 1440 CE deposits (i.e., "pre-1440"): SVG21 and 70, respectively from locations 14 and 64 of Cole et 212 al. (2019). Samples were analysed for major and trace element concentrations by mixed ICP-OES 213 (Inductively-Coupled Optical Emission Spectrometry) and ICP-MS (Inductively-Coupled Plasma 214 Mass Spectrometry) at ActLabs (Ontario, Canada).

215 The composition of the main mineral and glass phases was obtained for a selection of 11 samples, 216 representative of all the investigated eruptions and the recognised scoria types (see section 4), by EDS 217 (energy dispersive spectrometry) at DiSTAR laboratories, using an Oxford Instruments 218 Microanalysis Unit equipped with an INCA X-act detector and a JEOL JSM-5310 microscope. The 219 dataset previously presented by Cole et al. (2019) was significantly expanded as follows: 293 data 220 points for plagioclase (vs. previous 62), 147 for clinopyroxene (vs. 56), 95 for olivine (vs. 33), 105 221 for orthopyroxene (vs. 39), 107 for opaques (vs. 33), 95 for glasses (vs. 37). Additional new data 222 include a total of 96 data points for the pre-1440 scoria and 337 data points for xenoliths found within 223 juvenile clasts. Full procedural and analytical details can be found in Cole et al. (2019) and at 224 www.actlabs.com. Results and quality control data are reported in the Electronic Appendix as Tables 225 SM1-10.

226

4. Petrography and phase chemistry

228 **4.1. Petrographic features of the scoria clasts**

The investigated La Soufrière scoria samples display a strongly to moderately porphyritic texture (locally glomeroporphyritic) with abundant plagioclase (pl) and clinopyroxene (cpx) phenocrysts, plus lesser olivine (ol) and/or orthopyroxene (opx) set into a weakly to moderately vesicular glassy groundmass. All the main crystal phases commonly feature glass inclusions (especially pl and cpx) and inclusions of the other mineral phases. Some small though significant paragenetic differences can be envisaged between scoria clasts from different eruptions (Fig. 2). These have been already described in Cole et al. (2019), so they are briefly summarised here.

The samples from the 1718-1812 and 1902-03 historical eruptions include i) a more common scoria type dominated by large pl phenocrysts with generally smaller cpx, opx, few ol microphenocrysts and accessory opaque microphenocrysts/microcrysts, and ii) a less common 239 "mafic-rich" scoria type present only in 1902-03 products, characterised by approximately similar 240 abundances of pl and colourless/light green cpx, plus ol phenocrysts. Anhedral pl (only in the main 241 scoria type, particularly abundant and large in sample SVG14; Cole et al., 2019) and ol grains (larger than the euhedral/subhedral microphenocrysts, in both scoria types) are also occasionally observed. 242 243 The scoria samples from the 1440 and 1580 CE prehistoric eruptions, as well as those from the pre-244 1440 eruption reported here, are relatively homogeneous and share many similarities with the main 245 historical scoria clast type (including occasional subhedral/anhedral ol). Glomerocrysts generally 246 appear as clots of euhedral (occasionally intergrown) crystals with similar average size with respect 247 to phenocrysts and showing no evidence for textural disequilibrium. These usually consist of few 248 crystals and include both monomineralic (pl- or cpx- only) and polymineralic varieties 249 (pl+cpx±opx±ol±opaques), some also including small glass batches.

250

4.2. Petrographic features of the embedded xenoliths

252 A large variety of crystal clusters are present in all the scoria types, typically appearing as well-253 defined rock fragments with granular to intergranular textures (therefore being distinguishable from 254 the glomerocrysts), interpreted as gabbroic (lato sensu) and ultramafic xenoliths (Fig. 3). These are 255 generally well-recognizable even in the field, though they are only ~5-6 mm in diameter on average. 256 In the historical scoria samples xenoliths are mainly mm-sized (micro)gabbro assemblages 257 (cpx+pl±ol±opx±opaques), occasionally with few brown amphibole (in the 1902-03 main scoria 258 type), and sporadic coarse- to medium-grained (~5-7 mm) troctolite (1718-1812 eruption; Fig. 3d). 259 The 1902-03 mafic-rich scoria xenoliths include: 1) mm-sized granular dunite made of anhedral ol 260 (with opaques inclusions and few small pl attached on the borders; Fig. 3a); 2) mm-sized 261 microgranular wehrlite (Fig. 3b); 3) mm- to cm-sized ol-bearing microcrystalline (mela)gabbro, with 262 larger cpx and smaller interstitial pl and opaques (Fig. 3c). In the prehistoric scoria, gabbro xenoliths 263 are the most common (Fig. 3e), with largely variable proportions of the main phases 264 (pl+cpx+opaques±ol±opx), including both melanocratic cpx-dominated, and leucocratic pl-265 dominated varieties. Microgranular norite (pl+opx+opaques; Fig. 3f) and opx-rich websterite 266 (opx+cpx) xenoliths were also observed, respectively in 1440 CE and pre-1440 prehistoric scoria 267 samples. Microcrystalline pl+cpx-phyritic lava fragments were occasionally observed in the historical 268 (1718-1812) and prehistoric (1580 CE) eruption scoria. A synthesis of the main petrographic features 269 of the observed xenoliths is reported in Table 1.

270

In the following section, phase compositions are described separately for phases in textural equilibrium (euhedral/subhedral pheno/microphenocrysts and in glomerocrysts) and for phases from

- the xenoliths and those occurring as anhedral grains, out of textural equilibrium. Data on mineral inclusions are presented together with phases in textural equilibrium.
- 275

4.3. Composition of phases in textural equilibrium and mineral inclusions

277 4.3.1. Plagioclase

Plagioclase crystals from the historical eruptions cover a large compositional range from bytownite to labradorite (plus one andesine outlier; Fig. 4), with no significant difference between the main scoria type (i.e., $An_{49-96}Ab_{5-50}Or_{0-3}$) and the mafic-rich variety ($An_{59-93}Ab_{7-40}Or_{0-3}$). The same was observed for pl from the prehistoric eruptions ($An_{52-96}Ab_{4-48}Or_{0-2}$), including the pre-1440 (An_{57-} 91 $Ab_{8-42}Or_{0-2}$). Plagioclase in glomerocrysts are well within the compositional range of the coexisting phenocrysts.

Plagioclase inclusions are overall compositionally indistinguishable from the coexisting phenocryst/microphenocrysts (Fig. SM1), likely suggesting contemporaneous segregation. Inclusions within ol are generally less evolved ($An_{90-94}Ab_{6-10}Or_{<1}$, except for two more evolved crystals from the mafic-rich scoria with An_{56-75}) with respect to those in cpx ($An_{60-87}Ab_{9-41}Or_{0-1}$, except for two less evolved crystals from the 1902-03 scoria with An_{90-91}), opx ($An_{65-86}Ab_{11-35}Or_{0-1}$, except for one less evolved crystal from the 1902-03 scoria with An_{89}) and opaque minerals ($An_{59}Ab_{31}Or_0$).

290

4.3.2. Clinopyroxene

292 Titanium-poor (Ti <0.051 apfu) cpx is the main ferromagnesian phase of all the investigated 293 samples. In the samples from the historical eruptions, cpx is mostly augitic and covers the range Wo₃₆₋ 294 $_{45}En_{41-45}Fs_{13-19}$ in the main scoria type [Mg# 69-77; Mg# = 100*Mg/(Mg+Fe)], Wo₃₅₋₄₅En₄₀₋₄₆Fs₁₃₋₁₉ 295 in the mafic-rich scoria (Mg# 69-78; Fig. 5; Fig. SM3). The few (aluminian, Al >0.1 apfu) diopsidic 296 cpx crystals are mainly in the mafic-rich scoria crystal cores (Wo₄₅₋₅₁En₃₉₋₄₄Fs₈₋₁₃, Ca 0.871-0.951 297 apfu, Al 0.139-0.332 apfu, Mg# 76-84). Compositions follow a regular decrease of Ca (from 0.951 298 to 0.674 apfu) and Al (from 0.227 to 0.064 apfu) with decreasing Mg#. In the main scoria type, less 299 evolved compositions are mainly in crystal cores, differently from the mafic-rich scoria, where cores 300 feature both the most- and the least evolved compositions. The cpx found as inclusions within pl 301 (Wo₃₈₋₄₀En₄₁₋₄₃Fs₁₈₋₁₉, Al 0.052-0.089 apfu, Mg# 70-72) or opx crystals (Wo₄₀En₄₂Fs₁₈, Al 0.081 302 apfu, Mg# 70) display a composition comparable with that of the most evolved coexisting phenocryst 303 phases. As a whole, glomerocrysts cpx are indistinguishable from pheno- or microphenocrysts cpx.

Clinopyroxene from the prehistoric eruptions essentially covers the same range of that of the historical eruption main scoria type. Composition is mostly augite (rarely aluminian), spanning the range $Wo_{38-44}En_{41-45}Fs_{15-19}$ in the 1440 and 1580 CE (Al 0.059-0.126 apfu, Ca 0.748-0.843 apfu, Mg# 307 69-75), Wo₃₇₋₄₄En₄₁₋₄₃Fs₁₄₋₂₁ in the pre-1440 CE scoria clasts (Al = 0.070-0.121 apfu, Ca 0.730-0.845308 apfu, Mg# 68-75). Sporadic diopside compositions (Wo₄₅₋₄₆En₄₁₋₄₂Fs₁₃₋₁₄, Al 0.146-0.201 apfu, Ca 309 0.872-0.893 apfu, Mg# 76-77) are also found in the pre-1440 scoria samples.

310

311 4.3.3. Olivine

Olivine is found as a relevant phenocryst in textural equilibrium only in the mafic-rich 1902-03 scoria samples, showing a relatively large compositional variation, with crystal cores generally Mgricher (Mg# 71-86) with respect to both rims (72-83) and microphenocrysts (73-77). The fewer ol pheno/microphenocrysts analysed in the remaining samples are generally more evolved and cover the Mg# range of 61-76 in the historical eruptions, 72-77 in the prehistoric.

317

318 4.3.4. Orthopyroxene

319 Orthopyroxene is a common mineral phase that is lacking only in the mafic-rich scoria samples, 320 where it has been occasionally found only as inclusions within cpx (with Mg# in the 65-71 range). 321 Compositions are overall constant, with slightly Mg-richer crystals in the scoria from the historical 322 eruptions (Mg# 64-72), with respect to those from the 1440 and 1580 CE (Mg# 63-67) and the pre-323 1440 prehistoric events (Mg# 64-66). The few opx in glomerocrysts have a composition that is 324 perfectly consistent with these (Fig. SM3). Some opx grains have also been found included within 325 cpx and pl from the historical eruption scoria clasts, displaying a homogeneous composition with 326 Mg# 67, in line with that of the coexisting phenocryst counterparts.

327

328 4.3.5. Opaque minerals

Titano-magnetite is the main opaque microphenocryst/microcryst phase in all the investigated samples, except for the 1902-03 mafic-rich scoria (where it occasionally occurs as inclusion within ol and cpx). Compositions are variable, with Usp in the range of 19-42 mol.%, Mg# 8.7-20, Cr# <5.0 [Cr# = 100*Cr/(Cr+Al)], Al 0.121-0.276 apfu (Fig. SM5). Titanium-rich ilmenite compositions (Usp 87 mol.%) have been occasionally found only in the groundmass of pre-1440 CE scoria samples. Opaque grains from the glomerocrysts are compositionally consistent with coexisting microphenocrysts/microcrysts.

Opaque minerals occurring as inclusions do not deviate significantly from the general composition of the coexisting crystals in textural equilibrium, although they also feature some least evolved, Mg-Cr-Al-richer and Ti-poorer varieties. These are found in ol (Usp down to 1.8-8.7 mol.%, Mg# 18-59, Cr# 16-67, Al up to 0.535-0.880 apfu) and cpx (Usp 13-38 mol.%, Mg# 12-37, Cr# 9.1-44, Al 0.129-0.562 apfu) from the 1902-03 mafic-rich scoria. In the more evolved historical and prehistoric scoria,

- 341 opaque inclusions are generally Mg-poorer and Ti-richer moving from crystals hosted in ol (Usp 16-
- 342 22 mol.%, Mg# 14-22) and cpx (Usp 21-42 mol.%, Mg# 7.6-22), to those in opx (Usp 35-41 mol.%,
- 343 Mg# 8.7-15) and pl (Usp 38 mol.%, Mg# 10).
- 344

345 **4.4.** Composition of phases in the xenoliths and of anhedral phases

346 4.4.1. Plagioclase

In the 1902-03 mafic-rich scoria, plagioclase is mainly represented by labradorite-bytownite crystals from ol-gabbro xenoliths (An₆₈₋₇₉Ab₂₁₋₃₂Or₀₋₁; Fig. SM2). Additional sparse pl crystals have also been found on the borders of dunite (An₇₄₋₇₆Ab₂₄₋₂₆Or₀₋₁) and wehrlite xenoliths (An₆₆₋₈₀Ab₂₀₋₃₂Or₀₋₂).

351 Plagioclase from the gabbroic xenoliths in the historical scoria clasts include anorthite (An₉₀₋₉₅Ab₅₋ 352 $_{9}Or_{0-1}$) in 1718-1812 troctolite, and bytownite-labradorite in 1902-03 ol-gabbro (An₆₈₋₈₉Ab₁₁₋₃₁Or₀₋₂), 353 1902-03 gabbro (An₆₈₋₇₉Ab₂₁₋₃₂Or₀₋₁) and 1718-1812 gabbro (An₅₆₋₇₆Ab₂₄₋₄₃Or₀₋₁), the latter 354 displaying slightly Na-richer compositions. Similarly, pl from xenoliths in the prehistoric samples 355 include bytownite in 1580 ol-gabbro (An₈₄₋₈₉Ab₁₁₋₁₆Or₀₋₁), labradorite-bytownite in 1440 gabbro 356 (An₅₃₋₇₉Ab₂₀₋₄₇Or₀₋₁) and relatively evolved andesine in 1440 norite (An₃₆₋₄₇Ab₅₂₋₆₃Or₀₋₂). Finally, pl 357 from gabbro xenoliths found in pre-1440 scoria samples are mostly An₅₅₋₇₃Ab₂₇₋₄₅Or₀₋₂, with the only 358 exception of a bytownite crystal core (An₈₅Ab₁₄Or₁).

Anhedral pl crystals found in the historical eruptions scoria cover a similar compositional spectrum with respect to their counterparts in textural equilibrium (Fig. 4), although they mostly fall in the least evolved An-richer end (i.e., $An_{82-91}Ab_{9-18}Or_0$, in the mafic-rich scoria, $An_{81-96}Ab_{4-20}Or_{0-1}$ in the more evolved main scoria type, except for two Ab-richer crystal rims with An_{60-72}).

363

364 4.4.2. Clinopyroxene

Clinopyroxene from ol-gabbro xenoliths in the 1902-03 mafic-rich scoria have remarkably Mg-365 366 richer aluminian diopsidic/augitic cores (i.e., W042-49En41-47Fs7-14, Al 0.109-0.210 apfu, Mg# 76-88), covered by Mg-poorer aluminian augitic rims (W042-45En43-44Fs11-16, Al 0.105-0.123 apfu, Mg# 74-367 368 77; Fig. SM4). Similar to the latter are cpx crystals from wehrlite xenoliths (Wo₃₈₋₄₅En₄₁₋₄₉Fs₁₂₋₁₄, Al 369 0.072-0.200 apfu, Mg# 74-81), both being in line with cpx pheno/microphenocrysts from the host 370 scoria (Fig. 5). Clinopyroxene from ol-gabbro (Wo₃₉₋₄₃En₄₁₋₄₆Fs₁₅₋₁₇, Al 0.075-0.137 apfu, Mg# 71-371 77, in 1902-03 products) and gabbro xenoliths (Wo41-42En42-43Fs16-17, Al 0.088-0.109 apfu, Mg# 72-372 73, in 1902-03 products, Wo₃₉₋₄₂En₄₀₋₄₅Fs₁₅₋₁₈, Al 0.057-0.161 apfu, Mg# 71-75, in 1718-1812 373 products) in the historical eruption scoria is also relatively evolved, relatively homogeneous and close 374 to the composition of the corresponding cpx in textural equilibrium.

375 In the prehistoric eruptions scoria clasts, cpx in ol-gabbro xenoliths is only slightly less evolved 376 and Al-richer (Wo₄₂₋₄₃En₄₂₋₄₄Fs₁₂₋₁₆, Al 0.068-0.123 apfu, Mg# 74-79, in 1580 CE products) with 377 respect to that in gabbro xenolith (Wo₄₀₋₄₂En₄₁₋₄₃Fs₁₅₋₁₈, Al 0.071-0.120 apfu, Mg# 70-75, in 1440 CE 378 products), the latter being in line with the composition of the coexisting cpx in textural equilibrium. 379 Similar, relatively evolved and Al-poor compositions have been obtained for cpx in websterite (Wo39-380 42En40-42Fs16-19, Al 0.077-0.114 apfu, Mg# 70-73) and gabbro xenoliths (Wo40-43En40-42Fs16-19, Al 381 0.080-0.160 apfu, Mg# 69-74) from the pre-1440 CE scoria, which are also consistent with the 382 composition of the cpx in textural equilibrium with the host whole-rock sample.

Finally, some anhedral cpx from the 1902-03 mafic-rich scoria display Mg-rich aluminian diopsidic cores ($Wo_{46-50}En_{42-47}Fs_{6-9}$, Al 0.127-0.304 apfu, Mg# 83-88), covered by Mg-poorer aluminian diopsidic/augitic rims ($Wo_{44-45}En_{41-43}Fs_{13-14}$, Al = 0.132-0.195 apfu, Mg# 74-78).

386

387 4.4.3. Olivine

As regards the xenoliths in the 1902-03 mafic-rich scoria, a linear decrease in olivine Mg# can be envisaged moving from the dunite (78-86, with few rims in the range of 73-74) to the wehrlite (73-76) up to ol-gabbro types (68-75). A similar trend is evident for ol in the historical eruption xenoliths, moving from 1718-1812 troctolite (78-80) to 1902-03 ol-gabbro (63-67). The few ol crystals found within ol-gabbro xenoliths in prehistoric 1580 CE scoria have slightly lower Mg# (70-71) with respect to coexisting ol in textural equilibrium.

Anhedral ol crystals generally overlap the composition of the coexisting ol in textural equilibrium. Nevertheless, in the historical eruption samples slightly less evolved Mg-richer ol are present, both in the mafic-rich (Mg# 72-88, typically normally zoned) and in the main scoria type (65-79).

397

398 4.4.4. Orthopyroxene

399 Orthopyroxene is not very common in the La Soufrière xenoliths but is remarkably homogeneous 400 in composition, both in each xenolith type and as a whole, and in line with the composition of the 401 coexisting opx in textural equilibrium. In the historical eruption scoria opx occurs in both ol-gabbro 402 (1902-03 eruption) and gabbro (1902-03 and 1718-1812 eruption) xenoliths, with compositions being 403 almost identical (i.e., Mg# 69-70 vs. 68 and 67-70, respectively). Very similar compositions have 404 been obtained for opx in a gabbro xenolith from the 1440 CE prehistoric eruption scoria (Mg# 68) 405 and in websterite (66-67) and gabbro xenoliths (65-69) from the pre-1440 scoria, whereas a slightly 406 Mg-rich variety has been observed in a norite xenolith (71-73). The latter opx is also slightly higher 407 with respect to the composition of the coexisting opx phenocrysts.

408

12

409 4.4.5. Opaque minerals

- 410 Few opaque minerals were analysed from the xenoliths, mostly covering the range of the crystals 411 in textural equilibrium (Fig. SM5). In the 1902-03 mafic-rich scoria, opaques from a dunite xenolith 412 include a Cr-rich variety (Usp 15.6-17.3 mol.%, Mg# 24-35, Cr# 52-59, Al 0.315-0.462 apfu) and 413 some unusually Al- and Fe-rich crystals (Usp 1.9-2.8 mol.%, Mg# 61-65, Cr# 4-6, Al 1.451-1.547 414 apfu) found as inclusions in olivine. Opaques from wehrlite and ol-gabbro xenoliths are progressively 415 Ti-richer and Mg-Al-poorer (i.e., Usp 25.7-54.3 mol.%, Mg# 19-23, Cr# 32-37, Al 0.154-0.251 apfu and Usp 27.3-34.3 mol.%, Mg# 15.18, Cr# 21.32, Al 0.158-0.207 apfu, respectively). Opaques from 416 417 ol-gabbro and gabbro xenoliths within 1902-03 and 1718-1812 scoria samples are even Mg-Al 418 poorer, i.e., Usp 42.4 and 36.2-42.1 mol.%, Mg# 10 and 10-12, Cr# <5 and Al 0.126 and 0.141-0.167 419 apfu. Similar compositions have been obtained for opaques in gabbro xenoliths from the 1440 CE prehistoric scoria (Usp 36.5-40.3 mol.%, Mg# 9-12, Cr# <7, Al 0.131-0.162 apfu), as well as for pre-420 421 1440 websterite (Usp 37.9-39.9 mol.%, Mg# 9-12, Al 0.121-0.126 apfu) and gabbro xenoliths (Usp 422 36.8-38.6 mol.%, Mg# 9-14, Cr# <5, Al 0.140-0.164 apfu). Slightly Mg-Al-richer opaques have been 423 found in ol-gabbro xenoliths from the 1580 CE scoria, with Usp 27.6-29.6 mol.%, Mg# 16-17, Cr# 424 14-15 and Al 0.213-0.221 apfu.
- In addition, some Ti-rich Al-poor ilmenite opaques (Usp 78.4-79.6 mol.%, Al 0.010-0.029 apfu)
 were analysed in a norite xenolith from the prehistoric 1440 CE scoria.
- 427
- 428 4.4.6. Amphibole

A ~5 mm large single anhedral/subhedral brown amphibole grain was found in a historical scoria
 sample (SVG14; Fig. 2b). This shows a Ca- (1.896-1.944 apfu) Mg-pargasitic composition with high
 ^{IV}Al (2.019-2.108 apfu).

432

433 **5. Whole-rock and glass geochemistry**

434 **5.1. Whole-rock major elements concentrations**

The investigated La Soufrière scoria samples range from a few basalts (mainly the 1902-03 maficrich scoria) to more abundant basaltic andesites (Fig. 6), covering a relatively small compositional spectrum with respect to the wider range of literature data for the entire La Soufrière activity [mainly focused on the ancient phases; see Cole et al. (2019)]. Scoria clasts from the historical eruptions show a wider variability (i.e., SiO₂ 50.2-55.9 wt.%), whereas those from the prehistoric eruptions are generally more homogeneous and more evolved (SiO₂ 55.2-56.5 wt.% for 1440 and 1580 CE, 55.1-55.5 wt.% for the pre-1440 CE). 442 With the only exception of one historical sample [see Cole et al. (2019)], increasing SiO₂ from the 443 mafic-rich scoria to prehistoric eruption scoria is accompanied by decreasing MgO (from 7.78 to 3.23 444 wt.%) and CaO (10.7 to 8.35 wt.%), increasing Na₂O (2.70 to 3.84 wt.%) and K₂O (0.40 to 0.60 wt.%) 445 and constant TiO₂ (~0.90 wt.%). Aluminium and Fe₂O₃tot both firstly increase slightly (from 17.7 to 446 19.1 and from 9.06 to 9.58 wt.%, respectively), then start to decrease at SiO₂ ~52-53 wt.% (down to 447 17.9 and 8.28 wt.%). The "anomalous" historical sample SVG14 has the lowest SiO₂ (50.2 wt.%) 448 coupled with the highest Al₂O₃ (20.2 wt.%) and CaO (11.5 wt.%) and relatively low MgO (5.47 449 wt.%).

450

451 **5.2.** Groundmass glass and glass inclusions major elements concentrations

Glass compositions are remarkably more heterogeneous and more evolved with respect to the whole-rock ones. In the case of the mafic-rich scoria, the few glass analyses showing comparable SiO₂ contents are richer in TiO₂ (most evidently), Fe₂O₃tot and K₂O and poorer in Al₂O₃, CaO and Na₂O with respect to whole-rock samples.

- 456 Groundmass glass covers a wide compositional range, with an overall increase in the degree of 457 evolution moving from basaltic andesite and andesite in the 1902-03 mafic-rich scoria (SiO₂ 55.8-458 61.6, MgO 1.69-5.81 wt.%), to and esite/dacite in the historical (SiO₂ 62.7-66.4, MgO 0.72-2.10 wt.%) 459 up to dacite in the prehistoric ones (SiO₂ 63.0-67.2, MgO 0.08-1.78 wt.%; Fig. 6). Evolutionary trends are characterized by a clear decline of TiO₂ (0.41-2.85 wt.%), Fe₂O₃tot (0.57-11.6 wt.%), MgO (0.08-460 461 5.81 wt.%) and CaO (3.11-7.52 wt.%), whereas those for Al₂O₃ and Na₂O are more scattered and 462 complex, changing slope or even tendency at SiO₂ ~64 wt.%. The first increases from 11.1 to 17.7 463 wt.%, then it keeps roughly constant (except for one data down to 13.6 wt.%). On the other hand, 464 Na₂O first increases from 2.40 to 5.45 wt.%, then it slightly declines to ~3.6-4.0 wt.%.
- 465 Glass inclusions in the main phenocryst phases (especially in pl and cpx from the historical 466 eruptions) are globally in line with groundmass glasses. The least evolved compositions are found in 467 pl (SiO₂ 52.3-58.2, MgO 2.93-7.54 wt.) and ol (SiO₂ 53.3-56.7, MgO 2.54-3.41 wt.%) from the maficrich scoria (Fig. 7). With respect to groundmass glass of similar degree of evolution, the first are 468 469 strongly enriched in Fe₂O₃tot (9.70-18.7 wt.%), while the latter have higher CaO (6.52-9.85 wt.%). 470 Inclusions in pl from the main scoria type of the historical activity are more homogeneous and 471 systematically more evolved (SiO₂ 62.0-64.3, MgO 1.60-1.82 wt.%), following the above 472 compositional trends. Inclusions in cpx cover a wide compositional spectrum and display some larger 473 scattering, especially evident for Al₂O₃ 12.2-18.0 wt.%; Table SM7). Inclusions in cpx from the 474 mafic-rich scoria display the largest compositional interval (mostly SiO₂ 55.2-61.3, MgO 1.33-2.95 475 wt.%, plus two more evolved compositions with SiO₂ 66.0-67.9 and MgO 1.07-1.14 wt.%), whereas

those from the historical (SiO₂ 62.6-66.1, MgO 0.16-1.33 wt.%) and prehistoric eruptions (SiO₂ 60.1-65.0, MgO 0.04-2.62 wt.%) are generally less variable and more evolved. Glass in cpx from the historical eruption scoria is comparable with that in the coexisting pl. Finally, the few glass inclusions found in opx from both historical and prehistoric eruptions are on the most evolved end of the entire observed compositional spectrum (SiO₂ 61.7-65.9, MgO <1.23 wt.%). All the glass inclusion compositions are in good agreement with those from the existing literature (which also include more evolved compositions for all but glass inclusions in ol).

Only sparse glass occurrences have been recognised in a wehrlite xenolith from a 1902-03 maficrich scoria and a norite xenolith from a 1440 sample (Fig. 6). Compositions are on the whole consistent with the general trends observed for groundmass glass at a given SiO₂, with the first being remarkably less evolved (e.g., SiO₂ 54.7-58.2, CaO 5.02-7.23, MgO 2.44-3.08 wt.%) with respect to the latter (SiO₂ 64.4, CaO 3.27, MgO 1.68 wt.%).

488

489 **5.3. Whole-rock trace element concentrations**

490 Trace element contents and evolutionary trends for the investigated La Soufrière scoria clasts are 491 globally consistent with those for major elements. (Fig. 8 and Fig. SM6). Decreasing Sc (40 to 27 492 ppm), V (266 to 203 ppm), Cr (300 to 30 ppm), Co (33 to 15 ppm) and Ni (110 to 20 ppm) and 493 increasing Ba (51 to 142 ppm), Y (19 to 28 ppm) and Zr (65 to 96 ppm) can be observed moving 494 from the SiO₂-poorer mafic rich to the SiO₂-richest prehistoric scoria. A notable exception is observed 495 for Sr, roughly increasing from 193 to 230 ppm (though with some data scattering) for $SiO_2 > 51.2$ 496 wt.%, whereas the four SiO₂-poorer samples have 215-232 ppm. These latter include not only three 497 mafic-rich scoria types but also the SiO₂-poorer historical sample SVG14, which also displays 498 anomalously low Sc, Cr and Ni (31, 60 and 40 ppm, respectively) and the highest Sr (232 ppm).

499 Chondrite- (Ch) and primitive mantle (PM)-normalised multielemental plots are reported in Fig. 500 9. Patterns for Rare Earth Elements (REE) are almost flat, with $La_N/Yb_N = 1.25-2.13$ (N for 501 normalised abundances), indicating a weak enrichment in light LREE (from La to Nd) with respect 502 to the heavy HREE (Ho-Lu). No significant Eu or Ce enrichment or depletion can be envisaged, with $Eu/Eu^* = Eu_N/(Sm_N * Gd_N)^{1/2}$ ranging from 0.94 to 1.08 and $Ce/Ce^* = Ce_N/(La_N * Nd_N)^{1/2}$ from 0.97 503 504 to 1.07. Basaltic samples have normalised abundances for the most incompatible elements at 16-23 505 xCh, with the lowest values for the SiO₂-poorer sample SGV14. The patterns for basaltic andesite 506 samples have identical shapes, and are displaced to slightly higher REE contents (~19-36 xCh) with 507 lower La_N/Yb_N (1.25-1.64, except for two samples with 1.95 and 2.13) with respect to basalts (1.43-508 1.74). The presented data are broadly consistent with the few available literature data for the least 509 evolved La Soufrière HMB (e.g., most incompatible elements at ~17-26 xCh, La_N/Yb_N 1.42-2.35,

- 510 Ce/Ce* 0.96-1.01) and LMB samples (\sim 20-21 xCh, La_N/Yb_N 1.59-1.73, Ce/Ce* 0.99-1.00, though a 511 LREE-depleted sample with \sim 13 xCh, La_N/Yb_N 0.97, Ce/Ce* 0.86 is also present).
- 512 In PM-normalised plots, the investigated La Soufrière samples display the typically spiked patterns 513 of subduction-related magmas, with HFSE troughs (e.g., Ta, Nb, Ti; High Field Strength Elements), 514 relative enrichment in LILE (e.g., Cs, Rb, Ba; Large Ion Lithophile Elements) and LREE, and high 515 La/Nb (2.33-5.40), Ba/La (13.5-24.1) and Sr/Nd (18.4-26.5, up to 34.6 for sample SVG14). These 516 strongly resemble the typical GLOSS-2 (Global subducting sediment estimate), which is however 517 more enriched in fluid-mobile elements (e.g., LILE and Th-U) and generally in all the displayed 518 incompatible elements (especially REE, mostly between 10 and 40 xPM, whereas La Soufrière 519 samples are constantly <10). All the investigated samples show extremely similar subparallel trends, 520 with more evolved basaltic andesite displaced to slightly higher normalised abundances, and sample 521 SVG14 again featuring the lowest values. The observed patterns are on the whole similar to those for 522 the least evolved La Soufrière HMB (e.g., La/Nb 1.00-2.14, Ba/La 9.03-19.7, Sr/Nd 18.1-23.1) and 523 LMB samples (La/Nb 1.10-1.37, Ba/La 20.0-25.3, Sr/Nd 20.1-23.4).
- 524

525 **6. Discussion**

The petrological data for the last ~1000 years of activity at La Soufrière of St. Vincent can give new insights on the present state of the magmatic system beneath the island and help to evaluate its evolution through time. However, before making inferences on the La Soufrière magmatic system, some preliminary discussion on the nature, and significance, of such data seems necessary. In the following, two main questions are therefore addressed: 1) can whole-rock data on scoria clasts be taken as a representative for true melt compositions? 2) Do xenoliths represent fully-crystallised melt aliquots or cumulate assemblages?

533

534 **6.1. The composition of the La Soufrière magmas**

535 6.1.1. Evidence from the investigated scoria clasts

536 Given their strongly to moderately porphyritic nature, it is not straightforward to assess whether 537 the compositions of the investigated La Soufrière scoria clasts actually reflect those of true melts. 538 Some of the phenocrysts might in fact result from contamination/crystal accumulation, like those 539 showing anhedral shapes suggestive of disequilibrium conditions (i.e., xenocrysts scraped off from 540 crustal rocks during ascent), or antecrysts/protocrysts recycled from a crystal mush. In addition, some 541 of the larger anhedral grains with clast-like appearance (mainly pl, plus one amph in 1902-03 sample 542 SVG14) might also represent dismembered parts of holocrystalline xenoliths, which are fragile when 543 abundant interstitial liquid is present (Wager, 1962). The compositions of such crystals are generally

544 markedly An-rich for pl and Mg-rich for ol and cpx, similarly to some of the phases in textural 545 equilibrium (see section 4). Therefore, it could be argued that some of the phases that apparently 546 record equilibrium conditions actually did not have the time to react with the enclosing magma due 547 to rapid quenching during explosive eruptions (thus explaining also their large compositional ranges).

548 When whole-rock data are taken into account, these mostly display a very coherent behaviour with 549 increasing differentiation. As already reported by Cole et al. (2019), crystal fractionation of mainly 550 ol, cpx and pl (from basalt to basaltic andesite), followed by pl, cpx, opx and opaques (to more 551 evolved basaltic andesites), in line with petrography, mineral chemistry and literature data, 552 satisfactorily explains the observed geochemical trends. Simple major- and trace elements mass 553 balance calculations can efficiently reproduce the evolutionary processes of La Soufrière magmas 554 (see Electronic Appendix 1). In this framework, the "anomalous" historical sample SVG14 is 555 interpreted as representing a melt composition modified by accumulation of An-rich pl (up to An_{25}) 556 and Mg-rich amph, resulting in the lowest SiO₂ and the highest CaO, Al₂O₃ and Sr of the entire 557 dataset. This also explains the lower SiO₂, TiO₂, Na₂O, K₂O, Sc, Ba and REE and higher MgO 558 contents with respect to historical and prehistoric samples with similar petrography (i.e., basaltic 559 andesites with opx+opaques-bearing paragenesis with few or no ol).

560 Notwithstanding the above observations, whole-rock compositions are different from glass data. 561 The latter are not only systematically more evolved (even significantly, as in the mafic-rich scoria), 562 but also display some large variability within each sample grouping and can even reach peculiar compositions that are far from those of the whole-rock samples with similar SiO₂ (e.g., extremely 563 564 high TiO₂ and Fe₂O₃tot and low Al₂O₃). Such discrepancy can be explained assuming that the rapid 565 quenching did not allow total equilibration by elemental diffusion, which is particularly slow for 566 trivalent and tetravalent cations such as Si, Al and Ti (e.g., Zhang et al., 2010). In addition, glass 567 inclusions in mineral phases follow relatively regular compositional variations as a function of the 568 host phase, suggestive of a well-defined crystallisation sequence that is consistent with that inferred 569 from whole-rock data.

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571 6.1.2. Evidence from St. Vincent literature and experimental petrology

As mineral, whole-rock and glass data do not provide univocal response to the question about St. Vincent melt compositions, additional clues can be obtained from comparison with both literature whole-rock data and the results of experimental investigations (Fig. 10). As for the first, lava and juvenile samples from the entire volcanic activity at St. Vincent are considered. Experimental data are those for melts obtained from hydrous HMB parental magma crystallising at high P (1.0-1.3 GPa, with 2.3 and 4.5 wt.% H₂O; Melekhova et al., 2015) and from both HMB and LMB crystallising at 578 low P (0.4 GPa, with 1.7-7.7 and 1.6-5.1 wt.% H₂O, respectively; Pichavant and Macdonald, 2007).
579 Although the latter possibly do not perfectly correspond to true liquid compositions [and were slightly
580 corrected to retrieve two "parental melts" by Pichavant and Macdonald (2007)], these still provide a
581 coherent set of data that can be confidently used to check if the evolution of natural samples is
582 sufficiently well approximated by that of experimental analogues.

583 Present and literature whole-rock samples overall define regular arrays, but some data scattering 584 is also evident, with two groups of samples systematically deviating from the main trends. The first 585 has lower SiO₂, MgO and (less evidently) K₂O and higher CaO and Al₂O₃, which are ascribed to 586 plagioclase accumulation. Modal data for these samples indeed reveal that the total phenocryst load 587 is >20 vol.% (up to 45 vol.%), of which pl is >85 vol.% (up to 99 vol.%; Robertson, 2002). The 588 "anomalous" sample SVG14 generally plots close to this group, thus confirming the role of 589 plagioclase in producing significant compositional heterogeneity in whole-rock data. Interestingly, 590 many samples with higher phenocryst load (i.e., 50-67 vol.%; Heath et al., 1998a; Robertson, 2002) 591 do not depart substantially from the main trend. This is likely because these are not dominated by pl 592 phenocryst, but include also significant proportions of other phases (mostly cpx and ol, plus generally 593 lesser opx and opaques). The second deviating group is represented by five samples from the Yellow 594 Tuff Formation that are known to bear evidence for magma mixing (Heath et al., 1998a), and therefore 595 are taken as representative of a possible mixing trend.

596 As regards the compared experimental melts, low-P crystallisation experiments starting from a 597 LMB satisfactorily reproduce whole-rock differentiation trends for MgO < 8 wt.%. On the other hand, 598 low-P experiments on HMB follow different trends at such MgO contents, whereas they generally 599 seem to better match with the fewer whole-rock data with MgO > 8 wt.%. A major difference is 600 however evident from the CaO vs. MgO plot, where low-P HMB melts are characterised by an evident 601 initial CaO enrichment up to ~12-13 wt.% at ~8 wt.% MgO (likely due to the relatively late onset of 602 cpx crystallisation, especially at $H_2O > 4$ wt.%), not observed for the reported St. Vincent rocks. 603 High-P experimental HMB melts are on the whole similar to low-P ones, but provide a better match 604 with St. Vincent rocks especially in CaO vs. MgO plot, where they roughly simulate the nearly 605 constant or slightly declining CaO that is observed as MgO decreases from 16 to 8 wt.%.

In conclusion, even though crystal accumulation and/or contamination processes were surely active during the evolution of St. Vincent magmas, experimental data suggests that most of the whole rock data can be considered as indicative representatives for melt compositions. Evolutionary processes acted mainly during two different stages. The most primitive HMB melts differentiated at 1.0-1.3 GPa, approximately corresponding to depths of 30-40 km (assuming a conventional geobaric gradient of 0.03 GPa/km). The products of such evolution were LMBs, which successively differentiated at 0.4 GPa (~13 km). It seems reasonable to assume that open-system processes generally did not modify the composition of original liquids substantially, except when pl was significantly involved. This implies that in most cases recycled/accumulated crystals were approximately in the same proportions of equilibrium liquidus phase assemblages, and so assimilated materials were similar in composition to the assimilating magmas [as also suggested by Heath et al. (1998a)]. This is possibly confirmed by the above observation that strongly porphyritic samples with gabbroic (pl+cpx±ol±opx) phenocryst assemblages still follow whole-rock evolutionary trends.

619

620 6.2. On the nature of the investigated La Soufrière xenoliths

621 Holocrystalline xenoliths hosted in the La Soufrière deposits have been mostly interpreted as 622 cumulitic in origin, according to the detailed works from Wager (1962), Lewis (1973), Arculus and 623 Willis (1980) and Tollan et al. (2012). However, while such authors studied large ejecta blocks, the 624 samples investigated here are much smaller fragments hosted in single scoria clasts, mostly displaying a fine- to medium-grained, nearly equigranular texture, some with interstitial pl (Fig. 3c,e), some 625 626 reminiscent of the typical well-equilibrated adcumulitic texture (Fig. 3b,f). Mineral phase 627 compositions can give additional clues to their origin, but xenoliths must be considered individually, 628 as each lithotype could include xenoliths of both "intrusive" (i.e., melt aliquots that crystallised at 629 depth) and "cumulitic" (i.e., subtractive mineral assemblages fractionated during magma 630 differentiation) origin. To this scope, in figures 11 and 12 mineral data for each of the analysed 631 xenolith are reported separately and compared with their counterparts in textural equilibrium.

632 Gabbro xenoliths were found in all but the mafic-rich samples, typically with homogeneous cpx 633 (usually Mg# 71-75) coexisting with pl displaying a larger compositional spectrum of An₅₈₋₇₃ (plus 634 few An₅₂₋₅₅ and An₇₆₋₇₉, and even An₈₅ in one pre-1440 CE sample). When present, also opx and 635 opaques are generally homogeneous. The less common ol-gabbros have relatively Fo-poor ol with 636 F068-74, F062-66 and F070-71 (respectively for the 1902-03 mafic-rich, the historical and the prehistoric 637 samples), coexisting with Mg-richer cpx (Mg# 72-78), especially in the mafic-rich scoria (up to 84-638 88), and with Mg-richer Ti-poorer opaques. In the mafic-rich scoria these are found with pl that is not 639 particularly An-rich (i.e., An₆₈₋₇₂ and An₇₃₋₇₉), with higher An for crystals coexisting with Mg-richer 640 ol, cpx and opaques. The same is for ol-gabbro xenoliths of the historical and prehistoric samples, 641 which however also include a 1580 CE xenolith with An-rich pl (An₈₂₋₈₉), therefore approaching the 642 Fo₇₈₋₈₀ and An₉₂₋₉₅ association of the 1718-1812 troctolite xenolith that is similar to the typical 643 assemblage for St. Vincent cumulitic xenoliths (Tollan et al., 2012). The 1440 CE norite xenolith has 644 An-poor pl (An₃₈₋₄₇) coexisting with relatively Mg-rich opx (Mg# 71-73) and ilmenite.

645 The various ultramafic xenoliths found mostly in the mafic-rich scoria are uncommon among La 646 Soufrière cumulitic assemblages. The dunites feature ol with Mg-rich cores (Fo₇₇₋₈₆), some of which 647 with inclusions of extremely Al-rich spinels (possibly suggesting a mantle origin, and thus not 648 considered) and Cr-rich spinels similar to those occurring as inclusions in coexisting ol and cpx 649 phenocrysts. In some instances, ol cores are covered by tiny Mg-poorer rims (Fo72-74), likely the 650 product of crystallisation during equilibration with the host melt. The few relatively evolved small pl 651 crystals (An₇₅) found on the borders, likely attached to the xenolith by synneusis (during movement 652 in suspension in the melt). The wehrlite xenoliths have relatively Mg-poor ol (Fo72-76) and cpx (Mg# 653 74-81), in line with the composition of minerals in textural equilibrium, and some have pl of variable 654 compositions on the outer edges, again most likely attached by synneusis. The websterite xenolith in 655 the pre-1440 eruption scoria has homogeneous cpx, opx and opaques compositions similar to those 656 observed in the gabbro xenoliths.

657 The above observations suggest that the investigated xenoliths are of both intrusive and cumulitic 658 origin. The first include gabbros and ol-gabbros (except that from the 1580 CE scoria), as these feature 659 mineral phases with homogeneous compositions in line with coexisting phenocrysts and generally 660 have interstitial pl. Given the very similar composition of coexisting cpx and opx, it seems likely that 661 the websterite xenolith represents a portion of a dismembered gabbro xenolith (i.e., pl had detached). 662 On the other hand, dunites and wehrlites were likely segregated during the initial stages of magma evolution, followed by the troctolite (and the 1580 CE ol-gabbro), when pl became a major 663 664 fractionating phase, and by the norite, in the very latest stages. Additional evidence of these representing cumulate assemblages includes: 1) consistency with crystallisation sequences inferred 665 666 from whole-rock and glass data and with fractionating assemblages from mass balance calculations; 667 2) distribution of the xenolith types (ultramafic in the least evolved samples, gabbroic in the more 668 evolved); 3) variations in phase compositions (e.g., less evolved ol, cpx and opaques in the ultramafic 669 xenoliths, more evolved pl in the norite).

670

671 **6.3.** Constraints on the intensive variables in the La Soufrière magmatic system

Given the large chemical variability of the analysed mineral phases in textural equilibrium, to retrieve reliable data on intensive variables for the La Soufrière magmatic system through geothermobarometry is particularly challenging. The more homogeneous data from the xenoliths could be of some use in this by comparing mineral chemical data with results of experimental investigations, an approach previously undertaken for other Lesser Antilles rocks (e.g., Stamper et al., 2014a; Cooper et al., 2016; Camejo-Harry et al., 2018). To this purpose, selected starting compositions are St. Vincent parental HMB and LMB (Pichavant et al., 2002b; Pichavant and Macdonald, 2007; Melekhova et al., 2015). In addition, Guadeloupe HAB and andesite (GW4O and SG7B-1; Pichavant et al., 2018) and Martinique basaltic andesite (SV1/2 031-22; Pichavant et al., 2002a) were also considered in order to evaluate the possible role of more evolved magmas for which no experimental data on St. Vincent rocks are presently available. Both samples are shown to fall well within the liquid line of descent defined by St. Vincent magmas (Fig. SM7), therefore making reasonable representatives for such magma compositions.

685 The ol+Cr-spinel assemblage of the dunite xenoliths is broadly matched by those obtained from experiments starting from HMB at variable H₂O contents (0.6-5.1 wt.%) and P (0.4-1.0 GPa). In 686 687 these, ol is generally Fo-richer (86-93), suggesting that the dunite xenoliths crystallised from a more 688 evolved melt, consistent also with the lower Cr₂O₃ of Cr-spinel (12.4-24.1 wt.%) with respect to that 689 of experimental counterparts (generally 16.8-21.2 wt.%, but also up to 25.2-29.3 wt.%). The only 690 inference that can be therefore made for such xenoliths is that P was < 1.5 GPa (for 4.5 wt.% H₂O, <691 1.1 for 1.5 wt.% H₂O), as at higher P ol and sp are joined on the liquidus by cpx and cpx+opx (see 692 Pichavant et al., 2002b). The MgO-poorer ol rims were not considered, as clearly related to a later crystallisation stage (see section 6.2). Indeed, if a canonical K_D (Fe-Mg)^{ol/melt} of 0.30 ± 0.03 (Roeder 693 694 and Emslie, 1970; Matzen et al., 2011) is assumed, it results that such ol rims attained equilibrium 695 with melts with Mg# 0.44-0.51, roughly in line with that of the least evolved glass batches found in 696 the groundmass (41-46) or as inclusions (40-47) in the 1902-03 mafic-rich scoria. No experimental 697 counterpart has been found also for the Fo72-76 ol and Mg# 74-81 cpx association of the wehrlite xenolith. However, if K_D (Fe-Mg)^{ol/melt} of 0.30 ± 0.03 and K_D (Fe-Mg)^{cpx/melt} of 0.27 ± 0.03 (Putirka, 698 699 2008) are assumed, equilibrium melts have Mg# of 44-54 and 40-54, broadly corresponding to the 700 range observed for the investigated basaltic andesite samples (43-54).

701 The mineral compositions of the troctolite xenolith are roughly matched by those fractionating 702 from a HAB with 5.9 wt.% H₂O at ~0.2 GPa, 1000 °C and Δ NNO +1.0 [run L4-1 of Pichavant et al. 703 (2018)], consisting of Fo₇₄ ol and An₈₈ pl (plus some magnetite with 25 mol.% Usp). Although no 704 experimental counterpart has been found for the 1580 CE cumulitic ol-gabbro xenolith, conditions 705 were likely similar to those for the troctolite xenolith, based on the association of low-Fo ol and high-706 An pl. This is consistent with the estimates of Tollan et al. (2012) for St. Vincent ol-gabbro xenoliths 707 with comparable phase compositions, for which the authors proposed an origin from a HAB or a 708 basaltic andesite with 4.6 wt.% H₂O crystallising at 0.17 GPa. Interestingly, although ol-gabbro 709 assemblages obtained by Pichavant et al. (2018) have similar pl and cpx but rather Mg-richer ol (Fo₈₄₋ 710 85), the system conditions were similar to those for the troctolite assemblage (i.e., HAB with 4.5-5.0 711 wt.% H₂O at ~0.2 GPa, 1000 °C and Δ NNO +2.1-2.2; runs P6-14 and P6-17). The mismatch in ol 712 composition could be a consequence of the very oxidizing experimental conditions, as for lower fO₂ Fo significantly drops (see, e.g., the above run L4-1) and the natural compositions would be thusapproached.

715 No correspondence with experimental assemblages can be envisaged also for the norite xenolith 716 with relatively MgO-rich opx (Mg# 71-73) and An-poor pl (An₃₈₋₄₇). The latter possibly suggests that 717 the parent magmas were highly evolved, reaching compositions that have not yet been experimentally 718 investigated (and/or that explored system conditions were not appropriate). Indeed, few norite 719 assemblages have been obtained starting from andesite melts by Pichavant et al. (2018) for low P and 720 variably hydrous conditions (0.10-0.15 GPa, 1.5-5.7 wt.% H₂O), but these have An-richer pl (49-60) 721 and Mg-poorer opx (Mg# 48-58) and are possibly closer to fully-solidified liquid compositions rather 722 than to cumulates, based on their low glass content (19-44 vol.%). A higher melt fraction (87 vol% 723 glass) norite assemblage obtained by the same authors at 0.21 GPa from an andesite with 6.9 wt.% 724 H₂O has roughly similar opx but significantly An-richer (81) pl. If a K_D (Fe-Mg)^{opx/melt} of 0.28 ± 0.04 725 (Beattie, 1993) is assumed, the equilibrium melt for the norite xenolith opx has Mg# 37-46. Such 726 values are comparable with those of andesitic/dacitic glass samples reported here and of high-SiO₂ 727 (>61 wt.%) and esites from other central/southern Lesser Antilles islands (e.g., Martinique, St. Lucia 728 and Bequia; Smith et al., 1996; Davidson and Wilson, 2011; Bezard et al., 2015).

729 A possible experimental comparative for the "intrusive" xenoliths could be found in the lower T, 730 glass-poorer, more crystallised experimental runs. The ol-gabbro assemblages providing the best 731 matches are from HMB melts with 1.7-3.2 wt.% H₂O crystallising at 0.4 GPa [runs 3-3 and 6-2 of 732 Pichavant and Macdonald (2007), with 39 and 54 vol.% of residual glass]. These have Fo-richer ol 733 (80-82 vs. 68-74) and similar pl (An₇₀₋₇₇ vs. An₆₈₋₇₉) and cpx (En₄₃₋₄₆ vs. En₄₁₋₄₇), suggesting that 734 parental melts for the ol-gabbro xenoliths were more evolved. A good correspondence with gabbro 735 xenoliths assemblages is found in some 0.4 GPa experiments performed by Pichavant et al. (2002a) 736 starting from a basaltic andesite melt. The best match is for 5.9 wt.% H₂O (run HAB8, with 49 vol.% 737 glass), resulting in An₇₀ pl (vs. An₅₈₋₇₃), En₄₂ cpx (En₄₀₋₄₅) and En₆₁ opx (En₆₂₋₆₆). Starting from a 738 HAB with a similar composition (i.e., Al₂O₃ 19.8 vs. 19.0 wt.%, MgO 5.24 vs. 4.24 wt.%, CaO 9.75 739 vs. 9.60 wt.%) and crystallising at similar conditions (run P2, 0.4 GPa, 3.0 wt.% H₂O, 35 vol.% glass), 740 Pichavant et al. (2018) also obtained a mineral assemblage roughly matching that of the gabbro 741 xenoliths (An₆₅ pl, Mg# 64 cpx and En₅₉ opx). Interestingly, numerous experimental analogues for 742 the gabbro xenoliths are found also starting from hydrous (2.0-6.0 wt.% H₂O) and esite melts 743 crystallising at low P (0.10-0.21 GPa; Pichavant et al., 2018, see charges with 24-49 vol.% glass). 744 Websterite assemblages obtained by Pichavant et al. (2002b) and Melekhova et al. (2015) from highly 745 crystallised (53-58 and 34-52 vol.% of glass) HMB experimental melts at moderate H_2O (1.5 and 2.3 746 wt.%, respectively) and high P (1.2-1.8 and 1.0-1.3 GPa), typically produce Mg-richer cpx (En₅₀₋₅₂

- and En_{43-47}) and opx (En_{82-86} and En_{84}) with respect to the investigated xenoliths (En_{40-42} cpx and En_{63-10}) end En_{63-10} (En_{63-10}) and En_{63-10} (En
- ⁷⁴⁸ ₆₄ opx), supporting the hypothesis that these are dismembered gabbro xenoliths (see section 6.2).
- 749 In summary, the comparisons of experimental results with both xenolith and whole-rock data (see 750 section 6.1.2) point to shallow-level processes from variably evolved hydrous melts. Differentiation 751 processes appear to have occurred mainly at ~0.4 GPa, but even to P as low as 0.2-0.1 GPa for the 752 more evolved melts (which were possibly also H₂O-richer). The absence of amph in the cumulitic 753 xenoliths, contrasting with its common occurrence in literature La Soufrière xenoliths, points to 754 relatively low pressures and/or H₂O contents for the magmas of the last ~1000 years of activity. In 755 this regard, worth of note is the xenocrystic anhedral amph found in SVG14 historical sample. The 756 composition of the (hydrous) melt from which this segregated can be roughly estimated assuming a K_D(Fe-Mg)^{amph/melt} of 0.37 (Nandedkar et al., 2014) and using amph Mg#. The latter might be as high 757 as 83-86, if the calculated Fe^{3+} is used, as low as 71-72 if all iron is taken as Fe^{2+} (Table SM8). The 758 759 equilibrium melt would be a LMB with 65-70 Mg# or a basaltic andesite with ~48 Mg#, respectively. Based on Al-Si partitioning, such melt should have 0.202-0.214 Al/Si [using the K_D(Al-Si)^{amph/melt} of 760 761 0.97 from Pichavant et al. (2002a)], thus slightly more consistent with La Soufrière LMBs (0.194-762 0.209), rather than basaltic andesites (0.183-0.199 except for two samples with 0.202 and 0.207). In 763 any of these, the curvilinear margins of the observed amph indicate that it became unstable and 764 destabilised when it was incorporated by the enclosing magma, likely stalling at shallow depths.
- 765

766 **6.4. The evolution of the La Soufrière magmatic system**

767 The present data on scoria clasts and xenoliths suggest that during the last ~1000 years of activity 768 the La Soufrière has been fed by relatively homogeneous magmas, as indicated by the uniform 769 petrography, mineral and glass chemistry and whole-rock data for most of the basaltic andesite scoria 770 prevailing up to the 1718-1812 eruption. Rising magmas likely found favourable conditions to collect 771 in shallow crustal reservoirs (~7 km), where they could evolve and homogenise efficiently before 772 being tapped by eruptions (Fig. 13). Differentiation occurred starting from a less evolved basaltic 773 parental melt (similar in composition to the 1902-03 mafic-rich scoria, i.e., a LMB with ~8 wt.% 774 MgO), producing considerable gabbroic *l.s.* cumulates. Some of these were captured by the ascending 775 magmas as xenoliths, together with batches of fully crystallised magma (intrusive xenoliths) and 776 possibly portions of the basement developed during the ancient activity.

An evident compositional break is recorded in the 1902-03 products, in which less evolved basalts make a discrete juvenile component that reflects a major magma rejuvenation event. The new magma batches likely came from deeper crustal storage levels (~13 km), as testified by the different types of hosted xenoliths. Dunite xenoliths are interpreted as the subtractive assemblages from the 781 differentiation processes linking the 1902-03 basaltic magma batches with near-primitive HMB, 782 similar to (or slightly more evolved than) those sporadically erupted during the ancient activity. 783 Wehrlite xenoliths instead equilibrated with basaltic andesite melts, therefore representing the 784 cumulitic leftover of the differentiation of stagnating basalt magma. The occasional occurrence of 785 amph xenocrysts testifies for the recycling of (likely deeper) cumulates related with H₂O-richer 786 basaltic magmas. Finally, the post-1902-03 activity of 1971-1979 was again fed by relatively 787 homogeneous basaltic andesite magmas (Graham and Thirlwall, 1981), supporting a model of a 788 substantially steady-state behaviour of the La Soufrière magmatic system, episodically perturbed by 789 magma rejuvenation events.

790 Volcanic styles were likely affected by such evolution, with eruptions being related to the building-791 up of the volatile pressures, eventually leading to particularly violent explosive events (as in the 1580 792 CE case; Cole et al., 2019) when basaltic andesite magmas accumulated at shallow depths. On the 793 other hand, explosive activity associated with magma rejuvenation, such as the 1902-03 eruption, was 794 possibly driven by the development of chemical instabilities as the newly-arrived magma interacted 795 with the basaltic andesite magmas still residing in the reservoirs. Although the overall chemical 796 similarity between the two magmas (as well as between magmas and entrained gabbroic xenoliths) 797 strongly clouds the effects of such interactions, the larger variability in phase and glass compositions 798 in the mafic-rich scoria can be taken at least as a partial evidence.

799 As regards the older phases of activity, notwithstanding the significant amount of literature data, 800 no attempt has been done up to date to put all the petrological data in a chronostratigraphic context, 801 and juvenile products can be only crudely said to display much larger compositional spectrum with 802 respect to the recent activity (e.g., Cole et al., 2019). Similarly, the differences in the intensive 803 parameters for the feeding magmas (see section 2.2.1), have not been linked to specific periods of the 804 volcano history. Moreover, no precise stratigraphic control exists also for the xenolith data, as those 805 from Tollan et al. (2012) were collected *ex situ* concentrated in stream beds, whereas those from the 806 earliest literature are from the deposits of the 1902 eruption (Wager, 1962) and of some undefined 807 historical and prehistoric events (Lewis et al., 1973; Arculus and Willis, 1980). Therefore, only some 808 general inferences can be presently made on such stage of activity, the larger chemical variability of 809 emplaced magmas possibly speaking for a less efficient reservoir system and/or for higher magma 810 production rates. This resulted in more common (mainly effusive) eruptions of both near-primitive 811 HMBs (totally lacking in the latest activity), and relatively evolved basalts similar to those of the 812 1902-03 event. While mantle source compositions [i.e., a spinel-lherzolite, as indicated by low 813 LREE/HREE enrichment; see also Heath et al. (1998a)] and melting processes should have remained 814 substantially the same throughout the entire St. Vincent volcanic history, some changes might have

occurred in the crustal storage/plumbing system, influencing the paths of magma ascent *en route* to
the surface. This could simply reflect ongoing magmatism building a progressively more complex
and mature plumbing system (Heath et al., 1998a) through the emplacement of significant volumes
of mainly gabbroic *l.s.* intrusions and cumulates.

819 The structure of the crust beneath La Soufrière should be therefore that of a vertically-extended 820 crystal mush, capped by a layer of volcanic and volcanoclastic deposits (Fig. 13). The shallower 821 portions consist of various types of gabbroic assemblages (ol-gabbro, gabbro, troctolite and norite) 822 of cumulitic and intrusive origin, related to the differentiation/crystallisation of relatively evolved 823 melts (mostly at ~7 km, but possibly up to ~3.5 km for the most evolved). The lowermost portions, 824 i.e., below ~13 km, consist of ol+cpx (±Cr-spinel) cumulates, the liquidus assemblages for the less 825 evolved basalt precursors, and ol-gabbro intrusives. The absence of pl in the cumulitic xenoliths of 826 this level, in apparent contrast with the results of the proposed magma evolution models, could 827 suggest that this is mostly carried to shallower levels by ascending magmas due to its relatively low 828 density. Both portions of the mush pile should include also amph-rich domains where appropriate P-829 H₂O-melt composition conditions were attained (see Tollan et al., 2012). The distinction between the 830 two is in agreement with the results by Melekhova et al. (2019), who recognised a distinct increase 831 in P-wave velocity (v_P) at ~12 km depth. Below ~20 km a new increase of v_P is interpreted to reflect 832 a mainly (ultramafic) cumulitic layer extending down to the Moho (~30 km).

833 Similar crustal models were proposed for the nearby islands of Bequia of the Grenadines (Camejo-834 Harry et al., 2018) and Grenada (Stamper et al., 2014a) based on igneous xenolith data. However, the 835 latter feature more abundant amph and lesser pl (especially at Grenada) and indicate a larger range of 836 depths (8-17 km, possibly up to ~34 km for Bequia, 7-17 km for Grenada). Such differences might 837 be explained by the southward increase of the oceanic lithosphere thickness that according to Stamper 838 et al. (2014a) was also responsible for the increasing depths of magma generation (i.e., in the garnet-839 bearing peridotite facies). This is possibly related to the segmentation of the Benioff zone (see section 840 2.1), with the subducting slab becoming near vertical beneath Grenada. The higher mush pile below 841 Bequia and Grenada might have forced hydrous primitive melts to stall at higher depths where amph 842 crystallisation (and significant suppression of pl stability) could occur.

843

844 **7. Conclusions**

The present study on the juvenile products and entrained xenoliths has shed some light on magmatic processes during the last ~1000 years of activity of the La Soufrière volcano of St. Vincent. Prehistoric eruptions (pre-1440, 1440 and 1580 CE) and the historical eruption of 1718-1812 CE were fed by relatively homogeneous basaltic andesite magmas that host gabbroic xenoliths of intrusive (i.e., holocrystalline equivalents of magmas) and cumulitic (assemblages of fractionating minerals)
origin. Based on experimental petrology constraints, and especially on the composition of mineral
phases (association of An-rich plagioclase and relatively Fo-poor olivine), it was concluded that these
latter were segregated in a shallow level upper crustal reservoir (~7 km depth) from a hydrous highAl₂O₃ basalt/basaltic andesite magma.

854 Conversely, the historical eruption of 1902-03 CE was fed not only by basaltic andesite magmas 855 similar to those of the previous activity, but also by less evolved basaltic magmas, reflected in the presence of a mafic-rich scoria component (with pl+cpx+ol, instead of pl+cpx+opx+opaques) hosting 856 857 also ultramafic cumulitic xenoliths. Evolutionary processes likely occurred at higher levels (~13 km 858 depth) starting from a (possibly less) hydrous basalt magma that was similar in composition to the 859 parental melts of the basaltic andesite of the previous period of activity. This suggests that a 860 significant magma rejuvenation event likely had a role in triggering the 1902-03 eruption, in contrast 861 with the previous stages of activity where the building-up of volatile pressures related to the shallow-862 level stalling of relatively evolved basaltic andesite magmas was probably the main driving force. 863 The return to a condition dominated by relatively homogeneous basaltic andesite feeding magmas during the successive 1971-1979 activity, suggests that magma rejuvenation was just an episode in 864 865 the framework of the steady-state evolution of the La Soufrière magmatic system.

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1040 Figure captions

1041 Fig. 1. Geological sketch maps of a) the Lesser Antilles arc, showing the active volcanoes, the 1042 position of the volcanic front for the Old (Limestone Caribbes chain, dashed grey line, islands in 1043 white) and Recent arc (Volcanic Caribbees chain, black line, islands in dark grey) and of the 1044 subduction hinge (after Bouysse et al., 1990), and b) the island of St. Vincent (after Robertson, 2005). 1045 Fig. 2. Representative photomicrographs of the investigated historical and prehistoric scoria samples 1046 from the La Soufrière volcano (plane polarised view, except a and e, crossed polarisers view). a) 1047 1902-03 mafic-rich scoria (SVG44) with phenocrysts of pl (grey, twinned), ol (e.g., lower left, zoned) 1048 and cpx (top centre) in a vesicular glassy groundmass; b) main scoria type from the 1902-03 historical 1049 eruption (SVG14) showing a large anhedral/subhedral brown amph grain; c) scoria from the 1718-1050 1812 historical eruption (SVG16) with pl phenocrysts (colourless) and a glomerocryst made of cpx 1051 (pale green), opx (pale yellow) and opaques set into a vesicular glassy groundmass; **d**) scoria from 1052 the prehistoric 1580 CE eruption (SVG4) with phenocrysts of pl, cpx, opx and opaques (in a 1053 glomerocryst at the lower centre) set into a moderately vesicular glassy groundmass; e) scoria from 1054 the prehistoric 1480 CE eruption (SVG61) with phenocrysts of pl, cpx and opx, a ol microphenocryst 1055 (centre) and a large anhedral ol grain (upper right) set into a weakly vesicular glassy groundmass; f) 1056 scoria from pre-1440 CE prehistoric eruption (SVG21) with phenocrysts of pl, cpx, opx and opaques 1057 set into a weakly vesicular groundmass.

- 1058 Fig. 3. Representative photomicrographs of the xenoliths embedded within the investigated historical 1059 and prehistoric scoria samples from the La Soufrière volcano (crossed polarisers view, except b, plane polarised view). a) dunite xenolith in 1902-03 mafic-rich scoria (SVG44); b) wehrlite xenolith in 1060 1061 1902-03 mafic-rich scoria (SVG72); c) ol-microgabbro xenolith in 1902-03 mafic-rich scoria 1062 (SVG72) consisting of cpx and ol (e.g., crystals with light blue interference colours at the left and 1063 lower right edges) with interstitial pl; d) troctolite xenolith in 1718-1812 historical eruption scoria 1064 (SVG16); e) gabbro xenolith in 1440 CE prehistoric eruption scoria (SVG61), with larger cpx and 1065 interstitial pl and opaques; f) micro-norite xenolith in 1440 CE eruption scoria (SVG60).
- Fig. 4. Composition of the analysed plagioclase crystals in textural equilibrium (dashed fields for
 crystals in glomerocrysts) and with anhedral shapes from the investigated historical and prehistoric
 scoria samples from the La Soufrière volcano.
- **Fig. 5.** Ca vs. Mg# and Al vs. Mg# for the analysed clinopyroxene crystals in textural equilibrium (core, rim and microphenocrysts, dashed fields for crystals in glomerocrysts), with anhedral shapes and in xenoliths (ultramafic and gabbroic) from the investigated **a**) and **b**) historical (1902-03 maficrich scoria and main scoria type, respectively) and **c**) prehistoric scoria samples from the La Soufrière
- 1073 volcano.

- **Fig. 6.** Major element binary variation diagrams for whole-rock (normalised to 100% on a LOI-free basis) and glass data (in the groundmass and in the xenoliths, not normalizsed to 100%) of the investigated historical and prehistoric scoria samples from the La Soufrière volcano. The SiO₂ intervals for rock classification based on the TAS diagram (Le Maitre, 2002) are reported as dashed
- 1078 lines (B for basalt, bA basaltic andesite, A andesite, D dacite). A highlight on the "anomalous" 1902-
- 1079 03 sample SVG14 is also provided (see text for further details).
- Fig. 7. MgO vs. SiO₂ and CaO vs. SiO₂ diagrams for the analysed glass occurring as inclusions in a)
 plagioclase, b) olivine, c) clinopyroxene and d) orthopyroxene crystals in the investigated historical
 and prehistoric scoria samples from the La Soufrière volcano. Literature data from Heath et al.
 (1998a) are also reported for comparison.
- **Fig. 8.** Selected trace element binary variation diagrams for whole-rock (normalised to 100% on a LOI-free basis) data of the investigated historical and prehistoric scoria samples from the La Soufrière volcano. A highlight on the "anomalous" 1902-03 sample SVG14 is also provided (see text for further details).
- **Fig. 9.** Chondrite- and primitive mantle-normalised multielemental diagrams (after Sun and McDonough, 1989) for the investigated historical and prehistoric scoria samples from the La Soufrière volcano with **a**) and **b**) basalt and **c**) and **d**) basaltic andesite composition. The patterns for La Soufrière HMB and LMB literature samples (Heath et al., 1998a) and for the Global subducting sediment estimate (GLOSS-2; Plank, 2014) are also reported for comparison.
- 1093 **Fig. 10. a)** SiO₂, **b)** Al₂O₃, **c)** CaO and **d)** K₂O vs. MgO binary variation diagrams comparing the 1094 investigated historical and prehistoric scoria samples from the La Soufrière volcano with the existing 1095 literature for the entire volcanic activity at St. Vincent (Graham and Thirlwall, 1981; Heath et al., 1096 1998a; Robertson, 2002) and with experimental melts obtained starting from La Soufrière HMB and 1097 LMB at 0.4 GPa (Pichavant and Macdonald, 2007) and at 1.0-1.3 GPa with 2.3 and 4.5 wt.% H₂O 1098 (Melekhova et al., 2015). In c) the general trends defined by experimental melt data for 1.0 GPa and 1099 1.3 GPa are evidenced with the brown and purple arrows. St. Vincent literature data possibly defining a mixing trend and bearing evidence for some significant plagioclase accumulation are also 1100
- highlighted, along with the "anomalous" 1902-03 sample SVG14. See text for further explanations.
- Fig. 11. Synoptic view of the compositional variations for the analysed mineral phases in the xenoliths
 and in textural equilibrium with the investigated a) 1902-03 mafic-rich scoria and b) the historical
- 1104 1902-03 eruption and 1718-1812 eruption scoria samples from the La Soufrière volcano.
- 1105 **Fig. 12.** Synoptic view of the compositional variations for the analysed mineral phases in the xenoliths
- and in textural equilibrium with the investigated 1580, 1440 and pre-1440 CE eruption scoria samples
- 1107 from the La Soufrière volcano.

- Fig. 13. Schematic conceptual model for the magmatic system feeding the La Soufrière volcano
 during the last ~1000 years of activity. bA = basaltic andesite; HAB = high-Al2O3 basalt; HMB =
 high-MgO basalt.
- 1111

1112 **Table captions**

- 1113 **Table 1.** Synoptic table summarizing the main petrographic features of the observed xenoliths 1114 embedded within the analysed La Soufrière volcano scoria samples from the historical eruptions of
- 1115 1902-03 (* for the mafic-rich scoria) and 1718-1812, and from the prehistoric eruptions of 1580 CE,
- 1116 1440 CE and pre-1440 CE.
- 1117 Question marks indicate uncertain attribution due to small size
- 1118 Mineral phases are listed in order of decreasing abundance
- 1119 amph = amphibole; pl = plagioclase; cpx = clinopyroxene; ol = olivine; opx = orthopyroxene; opaque
- 1120 = opaque minerals
- 1121

1122 Electronic Supplementary Material

- Electronic Appendix 1. Major- and trace elements mass balance calculations for the investigated La
 Soufrière scoria clasts from the last ~1000 years of activity.
- 1125 **Table SM1.** Major- and trace element concentrations (respectively in wt.%, including values
- recalculated to 100% on a LOI-free basis, and in ppm) and LOI values (Loss On Ignition, wt.%) for
- the analysed La Soufrière volcano scoria samples (historical eruptions: 1902-03 and 1718-1812;
- 1128 prehistoric eruptions: 1580 CE, 1440 CE and pre-1440 CE). Data from Cole et al. (2019) are in italics.
- 1129 * = mafic-rich scoria
- 1130 bdl = below detection limits
- 1131 Mg# = $100*Mg/(Mg+Mn+Fe^{2+})$ on molar basis
- 1132 Table SM2. Major element concentrations (wt.%) and calculated structural formulae (apfu, atoms
- 1133 per formula unit, on 8 oxygens and 5 cations) for plagioclase crystals (pl) from the analysed La
- 1134 Soufrière volcano scoria samples (historical eruptions: 1902-03 and 1718-1812; prehistoric eruptions:
- 1135 1580 CE, 1440 CE and pre-1440 CE). Data from Cole et al. (2019) are in italics.
- 1136 * = mafic-rich scoria
- 1137 bdl = below detection limits
- 1138 Ab = albite mol.%; Or = orthoclase mol.%; An = anorthite mol.%
- 1139 mpc = microphenocryst; in ol = in olivine; in cpx = in clinopyroxene; in opx = in orthopyroxene; in
- 1140 opaque = in opaque mineral; glom = in glomerocrysts

- 1141 pl-anh = anhedral pl; olGb-xeno = in ol-gabbro xenolith; Dun-xeno = in dunite xenolith; Wehr-xeno
- 1142 = in wehrlite xenolith; Tro-xeno = in troctolite xenolith; Gb-xeno = in gabbro xenolith; Nor-xeno =
- 1143 in norite xenolith
- 1144 **Table SM3.** Major element concentrations (wt.%) and calculated structural formulae (apfu, atoms
- 1145 per formula unit, on 6 oxygens and 4 cations) for clinopyroxene crystals (cpx) from the analysed La
- 1146 Soufrière volcano scoria samples (historical eruptions: 1902-03 and 1718-1812; prehistoric eruptions:
- 1147 1580 CE, 1440 CE and pre-1440 CE). Data from Cole et al. (2019) are in italics.
- 1148 * = mafic-rich scoria
- 1149 bdl = below detection limits
- 1150 $Mg\# = 100*Mg/(Mg+Mn+Fe^{2+})$; En = enstatite mol.%; Wo = wollastonite mol.%; Fs = ferrosilite 1151 mol.%
- 1152 mpc = microphenocryst; in pl = in plagioclase; in opx = in orthopyroxene; glom = in glomerocrysts
- 1153 cpx-anh = anhedral cpx; in Wehr-xeno = in wehrlite xenolith; olGb-xeno = in ol-gabbro xenolith; Gb-
- 1154 xeno = in gabbro xenolith; Web-xeno = in websterite xenolith
- 1155 **Table SM4.** Major element concentrations (wt.%) and calculated structural formulae (apfu, atoms
- 1156 per formula unit) for olivine crystals (ol, on 4 oxygens and 3 cations) from the analysed La Soufrière
- volcano scoria samples (historical eruptions: 1902-03 and 1718-1812; prehistoric eruptions: 1580 CE,
- 1158 1440 CE and pre-1440 CE). Data from Cole et al. (2019) are in italics.
- 1159 * = mafic-rich scoria
- 1160 bdl = below detection limits
- 1161 $Mg\# = 100*Mg/(Mg+Mn+Fe^{2+})$; Fo = forsterite mol.%; Fa = fayalite mol.%; Teph = tephroite mol.%
- 1162 mpc = microphenocryst; in cpx = in clinopyroxene; glom = in glomerocrysts
- 1163 ol-anh = anhedral ol; Dun-xeno = in dunite xenolith; Wehr-xeno = in wehrlite xenolith; olGb-xeno =
- 1164 in ol-gabbro xenolith; Tro-xeno = in troctolite xenolith
- 1165 **Table SM5.** Major element concentrations (wt.%) and calculated structural formulae (apfu, atoms
- 1166 per formula unit, on 6 oxygens and 4 cations) for orthopyroxene crystals (opx) from the analysed La
- 1167 Soufrière volcano scoria samples (historical eruptions: 1902-03 and 1718-1812; prehistoric eruptions:
- 1168 1580 CE, 1440 CE and pre-1440 CE). Data from Cole et al. (2019) are in italics.
- 1169 * = mafic-rich scoria
- 1170 bdl = below detection limits
- 1171 $Mg\# = 100*Mg/(Mg+Mn+Fe^{2+})$; En = enstatite mol.%; Wo = wollastonite mol.%; Fs = ferrosilite
- 1172 mol.%
- 1173 mpc = microphenocryst; in cpx = in clinopyroxene; in pl = in plagioclase; glom = in glomerocryst

- 1174 olGb-xeno = in ol-gabbro xenolith; Gb-xeno = in gabbro xenolith; Nor-xeno = in norite xenolith;
- 1175 Web-xeno = in websterite xenolith
- 1176 Table SM6. Major element concentrations (wt.%) and calculated structural formulae (apfu, atoms
- 1177 per formula unit) for Ti-magnetite (mt, on 4 oxygens and 3 cations) and ilmenite-hematite (ilm, on 3
- 1178 oxygens and 2 cations) crystals from the analysed La Soufrière volcano scoria samples (historical
- eruptions: 1902-03 and 1718-1812; prehistoric eruptions: 1580 CE, 1440 CE and pre-1440 CE). Data
- 1180 from Cole et al. (2019) are in italics.
- 1181 * = mafic-rich scoria
- 1182 bdl = below detection limits
- 1183 Ulvöspinel mol% (Usp%), and FeO and Fe₂O₃ were calculated following Carmichael (1967)
- 1184 $Mg\# = 100*Mg/(Mg+Mn+Fe^{2+}); Cr\# = 100*Cr/(Cr+Al)$
- 1185 mpc = microphenocryst; in ol = in olivine; in cpx = in clinopyroxene; in pl = in plagioclase; in opx =
- 1186 in orthopyroxene, glom in glomerocryst
- 1187 Dun-xeno = in dunite xenolith; Wehr-xeno = in wehrlite xenolith; olGb-xeno = in ol-gabbro xenolith;
- 1188 Gb-xeno = in gabbro xenolith Nor-xeno = in norite xenolith; Web-xeno = in websterite xenolith
- 1189 **Table SM7.** Major element concentrations (wt.%, including values recalculated to 100%) for glass
- 1190 from the analysed La Soufrière volcano scoria samples (historical eruptions: 1902-03 and 1718-1812;
- 1191 prehistoric eruptions: 1580 CE, 1440 CE and pre-1440 CE). Data from Cole et al. (2019) are in italics.
- 1192 * = mafic-rich scoria
- 1193 bdl = below detection limits
- 1194 $Mg\# = 100*Mg/(Mg+Mn+Fe^{2+})$ on molar basis
- 1195 gm = groundmass; in ol = in olivine; in cpx = in clinopyroxene; in pl = in plagioclase; in opx = in
- 1196 orthopyroxene; glom = in glomerocryst
- 1197 Wehr-xeno = in wehrlite xenolith; Nor-xeno = in norite xenolith
- 1198 Table SM8. Major element concentrations (wt.%) and calculated structural formulae (apfu, atoms
- 1199 per formula unit, on 24 oxygens and 13 cations; Leake et al., 1997) for amphibole crystals (amph)
- 1200 from the analysed La Soufrière volcano scoria samples (historical eruptions: 1902-03; prehistoric
- 1201 eruptions: 1580 CE).
- 1202 bdl = below detection limits
- 1203 Mg# = $100*Mg/(Mg+Mn+Fe_{tot})$
- 1204 $Mg\#* = 100*Mg/(Mg+Mn+Fe^{2+})$
- 1205 amph-anh = anhedral amph
- 1206 **Table SM9.** Quality control data for the ICP-OES/ICP-MS whole-rock analyses.

- 1207 meas and cert are respectively measured and certified values for international standards; dup is for
- 1208 duplicate analyses of investigated samples
- 1209 **Table SM10.** Quality control data for the EDS mineral phases and glass microanalyses.
- 1210 cert = certified values; meas = measured values; meas av = average of measured values
- 1211 cert 100% = certified values recalculated to 100%; meas av 100% = measured values recalculated to
- 1212 100%
- 1213 USNM are Smithsonian standards, now labelled "NMNH"; MAC are standards from Micro-Analysis
- 1214 Consultant Ltd. (St. Ives, UK)
- 1215

1216 Supplementary figures

1217 Fig. SM1. Composition of the analysed plagioclase crystals occurring as inclusions in olivine and

- 1218 clinopyroxene crystals from the investigated historical and prehistoric scoria samples from the La1219 Soufrière volcano.
- **Fig. SM2.** Composition of the analysed plagioclase crystals occurring in xenoliths (ultramafic and gabbroic, with dashed fields when a highlight on each sub-types is needed) from the investigated historical and prehistoric scoria samples from the La Soufrière volcano.
- Fig. SM3. Classification of the analysed clinopyroxene and orthopyroxene crystals in textural equilibrium (core, rim and microphenocrysts, dashed fields for crystals in glomerocrysts) and with anhedral shapes from the investigated historical and prehistoric scoria samples from the La Soufrière volcano.
- Fig. SM4. Classification of the analysed clinopyroxene and orthopyroxene crystals in xenoliths
 (ultramafic and gabbroic) from the investigated historical and prehistoric scoria samples from the La
 Soufrière volcano.
- Fig. SM5. Al (apfu) vs. Mg# and Ti (apfu) vs. Mg# for the analysed opaque crystals in groundmass (dashed fields for crystals in glomerocrysts), occurring as inclusions and in xenoliths (ultramafic and gabbroic) from the investigated a) 1902-03 mafic-rich scoria, b) historical 1902-03 and 1718-1812 eruptions and c) 1580, 1440 and pre-1440 CE prehistoric eruptions samples from the La Soufrière volcano.
- Fig. SM6. Additional trace element binary variation diagrams for whole-rock (normalised to 100%
 on a LOI-free basis) data of the investigated historical and prehistoric scoria samples from the La
 Soufrière volcano. A highlight on the "anomalous" 1902-03 sample SVG14 is also provided (see text
- 1238 for further details).
- **Fig. SM7.** Major element binary variation diagrams comparing the investigated historical and prehistoric scoria samples from the La Soufrière volcano with the existing literature for the entire

- 1241 volcanic activity at St. Vincent (symbols and data sources as in Fig. 10) and with the starting
- 1242 compositions for the experimental investigations performed by Pichavant et al. (2002a) and Pichavant
- 1243 et al. (2018) for Martinique basaltic andesite (SV1/2 031-22b) and Guadeloupe HAB (GW4O) and
- 1244 andesite (SG7B-1), respectively.