1	Pre-eruptive reservoir conditions of the peralkaline
2	Rungwe Pumice (Tanzania) Plinian eruption from haüyne-
3	hosted melt inclusions
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22 ABSTRACT

23 The style and explosivity of volcanic eruptions are primarily governed by the interplay between 24 conduit dynamics (e.g., magma ascent rate and degassing efficiency) and pre-eruptive 25 magmatic conditions, both of which influence magma rheology. In highly alkaline magmas (i.e., 26 agaitic index > 1), the depolymerising effect of alkaline elements lowers melt viscosity at a 27 given temperature, theoretically favouring less explosive eruptions-even in silica-rich 28 systems. However, several well-documented eruptions demonstrate that silica-undersaturated 29 peralkaline magmas (e.g., trachytes and phonolites) can produce highly explosive Plinian 30 events, suggesting that additional factors control eruption style. In the East African Rift, 31 volcanoes with peralkaline compositions have exhibited both explosive and effusive behaviour. 32 To better understand the controls on explosivity, we investigated the pre-eruptive magmatic system of the Plinian eruption that produced the Rungwe Pumice (RP) deposit in southern 33 34 Tanzania. The late-stage evolution of the plumbing system was reconstructed through haüyne-35 hosted melt inclusions (MIs), revealing that an evolved magma body was stored at relatively 36 shallow depths shortly before the eruption. Water concentrations in MIs, measured via 37 transmitted Fourier-transform infrared spectroscopy, indicate relatively water-poor conditions (~4.8 wt.%) and shallow water saturation depths (~3.5 km). Our findings suggest that volatile 38 39 concentration alone does not fully explain the explosive behaviour; rather, the degree of water 40 undersaturation and conduit dynamics must also be considered as key factors influencing 41 eruption style.

43 1 INTRODUCTION

44 The eruptive behaviour of magma is governed by the exsolution of volatiles and the efficiency 45 of gas-melt decoupling during ascent in the volcanic conduit (e.g., Gonnermann and Manga, 46 2007; Cassidy et al., 2018). These processes are strongly controlled by several factors, including 47 ascent-and thus decompression-rate, conduit geometry, magma permeability, and the presence of crystals acting as bubble nucleation sites (e.g., Gonnermann, 2015; Shea, 2017). 48 49 However, a crucial role is played by melt composition-particularly major element and 50 dissolved volatile concentrations-which, along with reservoir temperature, pressure, and 51 volatile saturation, directly impacts melt rheology (Sparks, 1978; Dingwell, 1996; Giordano et 52 al., 2008; Degruyter et al., 2012; Gonnermann, 2015 and references therein). A primary control 53 on explosivity is therefore played by water, the most abundant volatile phase dissolved in melts, 54 as it drives decompression-induced supersaturation, bubble nucleation and growth, and 55 ultimately, magma foaming (Gonnermann and Manga, 2007). At the same time, rheology 56 dictates how magma responds to strain and accommodates stress, ultimately influencing 57 whether it undergoes brittle failure and fragmentation (Dingwell and Webb, 1989; Dingwell, 58 1996; Cassidy et al., 2018).

59 The viscous rheology of polymerised, high-silica, calc-alkaline melts, combined with elevated 60 water contents, often leads to violent Plinian-style eruptions (e.g., Walker, 1981; Cioni et al., 2015). In contrast, higher alkali (Na and K) concentrations depolymerise the Si-O bonds in 61 62 phonolitic or peralkaline trachytic and rhyolitic melts resulting in significantly lower melt 63 viscosities-up to two orders of magnitude lower for rhyolite under equivalent temperature and 64 water content conditions—compared to their calc-alkaline counterparts (Giordano et al., 2008; 65 Di Genova et al., 2013). Lower viscosities generally facilitate efficient outgassing and prolonged resistance to strain, potentially resulting in weaker eruptive explosivity. However, 66 several well-documented cases demonstrate that highly energetic, Plinian-style eruptions can 67

68 also originate from such lower-viscosity, alkali-rich magmas (e.g., Mahood, 1984; Cioni et al., 69 1999; White et al., 2012), even under water-undersaturated reservoir conditions (Andújar and Scaillet, 2012a). While high-alkaline series magmas are less abundant on Earth than the calc-70 71 alkaline series, which dominate continental arc settings (e.g., Stern, 2002 and references 72 therein), they occur across all geological environments, from subduction zones to intraplate 73 hotspots, and are particularly common in continental rift settings (e.g., Mahood, 1984; 74 Avanzinelli et al., 2004; Andújar and Scaillet, 2012b; Esposito et al., 2018; Jeffery and 75 Gertisser, 2018), especially along the East African Rift (EAR; e.g., Leat, 1984a; Macdonald and 76 Scaillet, 2006; White et al., 2012; Fontijn et al., 2013; Hutchison et al., 2016; Tadesse et al., 77 2019; Wallace et al., 2025).

Volcanism within the EAR is characterised by extensive, multicentred volcanic systems whose Quaternary activity includes Plinian-style eruptions, sometimes culminating in caldera collapses (e.g., Williams et al., 1984; Macdonald, 1987; Fontijn et al., 2018). Magmatic products range from silica-undersaturated to silica-saturated compositions but are generally enriched in alkali elements. The limited documentation of many of these volcanic centres results in a significant underestimation of volcanic hazards in densely populated regions across the EAR (Biggs et al., 2021).

85 To better understand the factors controlling the explosivity of moderate-viscosity, peralkaline systems widespread along the EAR, we investigated the pre-eruptive conditions of the ~4 ka 86 87 Rungwe Pumice (RP) eruption. These parameters are critical indicators of the current state of 88 magmatic reservoirs and the potential for future unrest. The RP deposit originates from a 89 Plinian-style eruption of Rungwe volcano in the Rungwe Volcanic Province (RVP; Tanzania; 90 (Fontijn et al., 2011). Relevant insights into pre-eruptive physical and chemical conditions and 91 processes are preserved in crystal-hosted melt inclusions (MIs). These melt pockets, trapped in 92 crystal imperfections during growth, can maintain chemical equilibrium with the original melt

and remain largely unaffected by its subsequent evolution during ascent (e.g., Esposito et al.,
2014). Particularly, while the pumice matrix melt undergoes extensive degassing during ascent,
MIs can retain the original volatile concentrations (e.g., Cioni, 2000; Esposito et al., 2014;
Cannatelli et al., 2016; Steele-MacInnis et al., 2017). In this study, the storage conditions, melt
chemistry and water content within the pre-eruptive reservoir are analysed using MIs hosted in
haüyne crystals (feldspathoid of the sodalite group) collected from different stratigraphic
horizons of a type section of the RP deposit (i.e., KF176).

100 2 GEOLOGICAL CONTEXT

101 The EAR is the largest active divergent continental margin and is originated by the divergence 102 of the Somalian and Nubian (African) Plates. It extends over 3000 km from the Afar triple 103 junction to Mozambique (Saria et al., 2014). In its central section, the rift encounters the thick 104 and rigid Tanzanian craton, splitting into the Western and Eastern branches (Saria et al., 2014; 105 Ebinger et al., 2017).

106 Volcanism occurs throughout the EAR but is predominantly concentrated along the more 107 mature Eastern branch (Fig. 1a). However, some of the most active present-day volcanic centres 108 are located along the Western branch, which hosts four of the twenty-one EAR volcanoes 109 known to have erupted in historical times (Wadge et al., 2016). Eruptive activity across the EAR 110 varies from effusive fissure eruptions to caldera-forming Plinian-style events (Macdonald, 111 1987; Fontijn et al., 2011, 2018). Magmatic products range from mafic to silicic compositions, 112 typically following alkaline or peralkaline series, with their petrogenesis extensively studied (e.g., Macdonald and Scaillet, 2006; Hutchison et al., 2018). 113

The Rungwe Volcanic Province (RVP) spans over 1500 km² and is located in the Western branch of the EAR, south of the Tanzanian Craton at the junction of the NW-SE trending Tanganyika-Rukwa rift, the N-S trending North Malawi rift and the NE-SW trending Usangu basin (Fig. 1a; Ebinger et al., 1993). The RVP comprises three main Holocene volcanic
centres—Rungwe, Ngozi and Kyejo—surrounded by hundreds of smaller eruptive centres,
which are scattered throughout the province along two dominant NW-SE and NE-SW structural
alignments (Fontijn et al., 2010a).

121 Volcanism in the RVP began approximately 9 Ma, with an initial phase (lasted until the Early 122 Pleistocene) characterised by alternating effusive and explosive eruptions of alkaline magmas 123 from volcanic centres that are now buried or eroded (Fontijn et al., 2012 and references therein). 124 The most recent phase of activity began with basaltic and trachytic lava flows spreading 125 northward into the Rukwa Rift and Usangu Basin, eventually focusing within a ~30 km-wide 126 NW-SE trending sector encompassing the three main volcanic centres and their satellite cones. 127 This phase transitioned into a predominantly explosive stage, producing large eruptions such as 128 the 10–12 ka Kitulo Pumice eruption and the <1 ka Ngozi Tuff eruption, which formed the 129 present-day Ngozi caldera depression (Fontijn et al., 2010b). In contrast, Kyejo's edifice 130 developed through mildly explosive and effusive activity from smaller centres (Harkin, 1960; 131 Fontijn et al., 2010a), culminating in the RVP's only recorded historical eruption, the 1800 CE 132 Sarabwe lava flows (Fontijn et al., 2012).

133 Rungwe is the largest volcanic edifice in the RVP and is centrally located within the province 134 (Fig. 1b). It is a young $(0.25 \pm 0.01 \text{ Ma}, \text{ whole-rock K-Ar age; Ebinger et al., 1989})$, 135 stratovolcano composed of basaltic, phonolitic, and trachytic lava flows. Its summit was 136 truncated by a sector collapse unrelated to magmatic activity (Fontijn et al., 2012), later infilled 137 by multiple silicic domes and tephra cones. The most prominent deposit associated with the 138 whole RVP is the ~4 ka Rungwe Pumice (RP) deposit (Fontijn et al., 2010b, 2011). More recent 139 eruptions include the Isongole Pumice-Rungwe's second-most explosive event (VEI 4; 140 Newhall and Self, 1982)-and three weaker explosive eruptions dated to <1.5 ka (i.e., 141 Ngumbulu Tephra; Aphyric Pumice, Kizungu Tephra; Fontijn et al., 2010b). Overall, Rungwe

has experienced at least one explosive eruption every 500 years, with activity ranging fromviolent Strombolian to Plinian events (Fontijn et al., 2010b).

144 **2.1 Rungwe Pumice eruption**

145 The RP deposit consists of a massive pumice lapilli fall deposit that blankets the entire RVP. 146 Radiocarbon dating and sedimentary records from Lake Masoko place the eruption at 147 approximately 4 ka (Garcin et al., 2006; Fontijn et al., 2010b). The deposit is lithic-poor and 148 predominantly composed of cream-coloured pumice lapilli of high-alkaline, trachytic 149 composition (whole-rock; Fontijn et al., 2013). A minor fraction (<1wt%) of grey/banded 150 pumice has been identified, interpreted as evidence of magma mingling with a slightly more 151 mafic melt (Fontijn et al., 2011). The RP deposit storage conditions were inferred using the 152 titanomagnetite-ilmenite pair geothermometer (Ghiorso and Evans, 2008) which indicated a 153 temperature of 925 °C and an oxygen fugacity in the NNO+0.25-NNO+0.45 range (nickel-154 nickel oxide buffer; Fontijn et al., 2013). Reservoir ponding depths were estimated at 2.5-13 155 km within the shallow-mid crust with the clinopyroxene-liquid barometer (Putirka, 2008; 156 Fontijn et al., 2013). A maximum pre-eruptive water content of 5.5 wt.% (Fontijn et al., 2013) 157 was estimated using the by-difference method on melt inclusion compositions (Devine et al., 158 1995).

RP's pumice ground mass is microlite-free, phenocrysts poor (<5 vol.%; Fontijn et al., 2013)
and highly vesicular (Fontijn et al., 2013; Cappelli et al., 2025). Alkali feldspar is the dominant
free mineralogical component, with subordinate biotite, clinopyroxene and Fe-Ti oxides.
Plagioclase, though rare, is exclusively present in grey-banded pumices (Fontijn et al., 2013).
Notably, cyan-coloured haüyne occurs in the RP deposit, distinguishing it from most other RVP
deposits and serving as a field marker for identifying RP outcrops (Fontijn et al., 2011).

More than 100 RP outcrops were mapped across the province extending up to 28 km from Rungwe summit, where the deposit maintains a thickness of 30 cm. A ~1 m-thick tephra layer attributed to the RP eruption was found in Lake Masoko's sedimentary record around 25 km SSE of Rungwe (Garcin et al., 2006; Fontijn et al., 2012). While no clear evidence of more distal deposits remains—likely due to pedogenesis—sediment cores from Lake Malawi, ~115 km SE of Rungwe, contain ash horizons chronologically linked to the RP eruption (Fontijn et al., 2011).

172 RP outcrops are radially distributed around the summit, with near-circular isopleths indicating 173 that the eruption originated from the present-day Rungwe summit and likely occurred under 174 wind-still conditions (Fontijn et al., 2011). The maximum lithics method (Carey and Sparks, 175 1986) suggests an eruptive column height of 30-35 km (Fontijn et al., 2011), which was likely 176 sustained throughout the entire duration of the eruption, as no associated large pyroclastic 177 density current deposits have been identified. The peak mass discharge rate, estimated from a range of inferred column heights, falls between 2.8 and 4.8 $\times 10^8$ kg·s⁻¹, while the minimum 178 179 erupted volume is approximately 1.4 km³ dense rock equivalent (Fontijn et al., 2011). Based on 180 these parameters, the RP eruption is classified as Plinian, with a Volcanic Explosivity Index of 181 5 (Newhall and Self, 1982; Fontijn et al., 2011).

A reference section for the RP deposit, designated as KF176 in previous studies (Fontijn et al., 2010b, 2011, 2013), was identified approximately 11.7 km SSE of the Rungwe summit. This ~2.5 m-thick, massive pumice deposit exhibits an unimodal, well-sorted grain-size distribution (sorting $\sigma_{\phi} < 2$) centred around the median at -2.2 ϕ (~4.5 mm)—except for the finer and reversely graded base (lower 50 cm; Fontijn et al., 2011)—and is bounded by overlying and underlying palaeosols. The deposit was previously sampled through its entire thickness by Fontijn et al. (2011) and subdivided into 14 subsamples at 20-25 cm intervals from base to top, sequentially labelled KF176-B to KF176-O, with KF176-A corresponding to the basalpalaeosol.

191 **3 METHODS**

192 **3.1 Componentry analyses**

The different types of components constituting the RP deposit were investigated by counting 3000 particles within the medium (0.35-0.50 mm) and coarse (0.71-1.00 mm) ash fractions for each subsample of the type section (KF176-B to KF176-O). With the aid of a stereomicroscope, the particles were characterised and classified into three categories: juvenile (pumice), lithics and crystals. Detailed methodology and results are provided in **Supplementary Information**.

198 **3.2 FTIR analysis**

The total H₂O and CO₂ content dissolved in MI glasses was quantified using transmission Fourier Transform Infrared (FTIR) spectroscopy, by applying the Beer-Lambert Law (§3.5). FTIR spectroscopy measures the absorbance of specific wavelengths in the near-/mid-infrared range caused by the vibrational modes of the target analytes (e.g., von Aulock et al., 2014). A detailed description of the methodology and parameters used is provided in the **Supplementary Information**.

205 FTIR spectra were acquired at the ISTO-CNRS laboratory (Orleans, France) using a Nicolet 206 6700 FTIR spectrometer connected to a Nicolet Continuum Infrared Microscope equipped with 207 an MCT IR Detector Module. The acquiring setup consisted of a Globar light source, an XT-208 KBr beamsplitter and a 32x infinity-corrected Schwarzschild objective matching to a 32x 209 condenser. The square beam aperture was set at 20x20 µm or 40x40 µm for larger MIs, while a 210 15x15 µm aperture was exceptionally used for the smallest MIs. Water concentration was 211 determined from the fundamental stretching vibration of O-H bonds at 3550 cm⁻¹, which retains 212 a strong signal-to-noise ratio. However, at higher water concentrations, this peak can become

saturated unless the sample is extremely thin (von Aulock et al., 2014). In such cases, water content was estimated using the combined absorbance bands of hydroxyl groups (OH⁻; 4500 cm⁻¹) and molecular water (5200 cm⁻¹). Molecular CO₂ yields a vibrational peak at 2351 cm⁻¹ (Brooker et al., 1999), while dissolved carbonate species (CO₃²⁻) produce an absorption doublet between 1670 and 1375 cm⁻¹ (Brooker et al., 1999). Spectra were therefore acquired over a wavenumber range of 1200-6000 cm⁻¹ with a spectral resolution of 4 cm⁻¹ (**Fig. 2**).

219 MIs and embayments (i.e., inclusions open on one side) are hosted in various RP crystalline 220 phases, including alkali feldspars (as tiny lamellar inclusions), pyroxenes, and Fe-Ti oxides 221 (hardly detectable). However, haüyne-hosted MIs were the most suitable for investigation due 222 to their abundance, well-developed morphology, and larger sizes. Additionally, haüyne-the Ca 223 endmember of the haüyne-nosean series $([Na_{5-8}K_{0-1}Ca_{0-2}(Fe,Mg)_{0-1})$ 1]8[Al₆Si₆O₂₄]·(SO₄,S,Cl,OH,CO₃)₁₋₂·*n*H₂O·*m*CO₂)—reportedly forms late in the crystallisation 224 sequence (e.g., Harms and Schmincke, 2000). Notably, RP haüyne never occurs as a mineral 225 226 inclusion, but instead hosts inclusions of apatite, ilmenite and biotite (Fontijn et al., 2013), 227 suggesting it could entrap a melt representative of the late pre-eruptive conditions.

228 Four stratigraphic subsamples representing different horizons of the type section were selected: 229 bottom (KF176-C), middle (KF176-G and KF176-K), and top (KF176-O). More than 100 230 haüyne crystals (0.5–2 mm in size) were handpicked from manually crushed pumice clasts. 231 Crystals were carefully examined to identify potential melt inclusion assemblages (MIAs; 232 Bodnar and Student, 2006; Esposito et al., 2014) and to discard unsuitable MIs (e.g., inclusions 233 smaller than 15 µm, or those with excessive bubbles obstructing the FTIR beam path). Suitable crystals were embedded in a thermosetting resin (CrystalbondTM 509) and doubly polished (von 234 235 Aulock et al., 2014) with diamond pastes to expose MIs on two opposed sides, producing thin 236 crystal wafers (tens of micrometres thick). Due to variations in crystal availability and MI

suitability, the final dataset included a variable number of MIs per horizon, for a total of 86 MIsand embayments.

239 For FTIR analysis, crystals were gently removed from the resin by heating on a hot plate, rinsed 240 in acetone to remove resin residuals, and placed over a CaF₂ window. A background spectrum 241 on the CaF₂ window was collected for calibration before each measurement, and whenever 242 environmental conditions (e.g., temperature, CO₂ concentration, humidity) affected spectral 243 trends. Absorbance values were determined using peak heights rather than peak areas to 244 minimize distortion effects, and a linear baseline was adopted. A minimum of three scans per 245 MI were collected to ensure reproducibility, spectra were then averaged using Omnic[™] 246 software data processing tools to reduce the spectral noise. FTIR spectra were acquired at both 247 the MI centre and closer to rims to evaluate water content consistency, while embayments were 248 scanned in their most internal portions.

249 Since haüyne can host both structural and molecular volatile phases, the same FTIR procedure 250 used for MIs was applied to estimate the H₂O and CO₂ concentrations in MI-hosting haüyne 251 crystals. Haüyne is an isotropic crystal with a cubic symmetry, meaning its absorbance is 252 unaffected by the orientation of optical indicatrix axes, allowing analysis in random orientations 253 (e.g., Bellatreccia et al., 2009). Spectra were acquired close to the MIs adopting the same 254 acquisition settings and a beam aperture of 40x40 µm. Haüyne's water absorbance spectrum consists of three overlapping overtones in the $\sim 2800-3700$ cm⁻¹ range, therefore the integrated 255 256 absorbance was used (Bellatreccia et al., 2009), while the stretching of CO₂ molecules gives a 257 well-defined absorbance peak at ~ 2350 cm⁻¹ (Fig. 2; Bellatreccia et al., 2009).

258 **3.3 EPMA analysis**

259 Major element concentrations of MI previously analysed with FTIR were measured with a 260 JEOL JX-iHP200F field emission electron probe micro-analyser (EPMA) at the IMAP 261 laboratories of the Université Grenoble-Alpes (France). The crystal wafers were embedded in 262 epoxy resin, polished with diamond pastes (down to 1 µm diamond grade), and carbon coated. 263 Analyses were conducted at an acceleration voltage of 15 kV, a probe current of 5 nA, and a 264 defocused beam diameter of 15 µm. The analytes were measured with five wavelength 265 dispersive spectroscopy (WDS) spectrometers and one energy dispersive spectroscopy (EDS) 266 spectrometer (Si only, with StHs6-80 glass as calibration material). Sodium was analysed first 267 to avoid element dispersion and a ZAF correction was applied. The StHs6-80, Atho-G and 268 KE12 reference materials (Jochum et al., 2006) were analysed three times every 30-40 269 measurements to evaluate accuracy. Each MI was measured at least three times for consistency. 270 Simultaneously, major element concentrations of MI-hosting haüyne crystals were analysed 271 under the same conditions, using Cr-Augite, Ortho-Bl3, and Ortho-Bl2 as secondary standards. 272 Groundmass glass chemistry of pumices was also analysed using EPMA. Pumiceous glass 273 shards were obtained from the 125-250 µm grain-size fraction of manually crushed pumice 274 clasts from the four stratigraphic subsamples investigated. The crushed material was wet sieved at 80 µm in demineralised water to remove accretionary dust or clays, then embedded in epoxy 275 276 resin, ground and polished with diamond pastes down to 1 µm grade. The same instrument and 277 analytical conditions used for MI analyses were applied, with a 2 nA probe current and a 5 µm 278 beam diameter. Glass chemistry of each subsample was analysed at least eight times for 279 reproducibility on different glass shards.

Additionally, major element concentrations in alkali feldspar were analysed along core-to-rim transects consisting of five measurement points to assess chemical variations during crystallisation. Crystals were handpicked from the 0.5–2 mm grain-size fraction of crushed pumices, embedded in epoxy resin, and polished following the same preparation steps as other samples. Cr-Augite, Ortho-B13, and Ortho-B12 were used as secondary standards.

285

3.4 Bulk rock analysis

286 Whole-rock major and trace element concentrations were determined for pumices of each 287 deposit subsample (KF176-B to KF176-O) at the Laboratoire G-Time of the Université libre de 288 Bruxelles (Belgium). Samples were manually crushed to a fine powder in an agate mortar and 289 prepared as a solution by alkaline fusion. Loss on ignition was estimated by heating the powders 290 at 800 °C for 2h after drying them completely at 105 °C for over 24h. Major elements were 291 measured with a Thermo Fisher Scientific iCAP inductively coupled plasma-optical emission 292 spectrometer (ICP-OES) using Y as an internal standard. Trace elements were analysed with an 293 Agilent 7700 quadrupole inductive-coupled plasma mass spectrometry (Q-ICP-MS) operated 294 with a He-filled collision cell. The USGS standards BHVO-2, RGM-2, and QLO-2 were 295 analysed as reference materials and one sample duplicate was measured for reproducibility. The 296 total reproducibility based on reference materials was estimated to be better than 4% and 12% 297 relative standard deviation for major and trace elements respectively.

298 3.5 Volatile concentrations

299 The relationship between infrared light absorption and analyte concentration in a sample 300 follows the Beer-Lambert Law (Stolper, 1982), which requires knowledge of thickness, density 301 and molar absorptivity index of the analysed phase. A detailed description of the methodology 302 and parameters is provided in Supplementary Information.

303 To minimize uncertainty introduced by operator bias (e.g., optical focus recognition), MI and 304 host crystal thicknesses were determined using two independent methods and compared. First, 305 a flat-anvil digital micrometre (resolution = 1 μ m; accuracy = $\pm 2 \mu$ m) was used, which reduces 306 the risk of damaging the fragile crystals compared to a pointed micrometre. Then, the thickness 307 was measured using a LEITZ DMR optical microscope equipped with a calibrated and 308 automated focus stage (resolution = $2 \mu m$). Each measurement was repeated two or three times 309 per crystal.

For MI glass, an average density of 2.25 ± 0.01 g·cm⁻³ was estimated as a function of its 310 311 chemical composition (Lange, 1997) which requires glass volume determination. Volume was 312 estimated using the temperature-dependent equations for hydrated iron-bearing silicate glasses 313 (Lange and Carmichael, 1987; Ochs III and Lange, 1999; Richet et al., 2000), with temperature 314 estimated using an alkali feldspar-liquid geothermometer (Eq. (24b) in Putirka, 2008). To assess 315 equilibrium between feldspar rims and pumice groundmass, petrographic observations of thin 316 sections of lapilli-sized clasts from five stratigraphic subsamples (KF176B; KF176E; KF176H; 317 KF176K; KF176N) were conducted using optical petrographic microscopy and scanning 318 electron microscopy (SEM). SEM imaging was performed in backscattered electron mode, at 319 15 kV using a Hirox SH5500P equipped with a Bruker EDS Quantax detector at the Laboratoire 320 G-Time of the Université libre de Bruxelles (Belgium). No disequilibrium features were 321 identified. However, before applying feldspar-based thermometry, textures of feldspars 322 analysed with EPMA were carefully inspected also with SEM. Haüyne density was assumed to be 2.4 g·cm⁻³ (Taylor, 1967). 323

A molar absorptivity index of $66.9 \pm 5.8 \text{ L} \cdot \text{mol}^{-1} \cdot \text{cm}^{-1}$ (Behrens and Hahn, 2009), 324 experimentally determined for high-alkali trachytic glasses, was used for the 3550 cm⁻¹ 325 overtone of MI absorbance spectra. Indices for the 4500 cm⁻¹ and 5200 cm⁻¹ overtones were 326 327 estimated through multiple linear regression (Di Matteo et al., 2004), with the water content calculated from the 3550 cm⁻¹ band as a fixed term. The resulting molar absorptivity indices, 328 with 90% confidence bounds as subscripts and superscripts, were $1.35_{1.01}^{1.71}$ and $1.03_{0.69}^{1.38}$ L·mol⁻ 329 ¹·cm⁻¹ for the 4500 cm⁻¹ and 5200 cm⁻¹ overtones, respectively. These values are similar but 330 slightly different from those previously estimated for trachytic (1.58 and 1.36 L·mol⁻¹·cm⁻¹ 331 respectively; Di Matteo et al., 2004) and phonolitic (1.25 and 1.10 L·mol⁻¹·cm⁻¹ respectively: 332 333 Carroll and Blank, 1997) glass. Integrated molar absorptivity indices for H₂O and CO₂ in haüyne were fixed at 5422 and 1866 $L \cdot mol^{-1} \cdot cm^{-2}$ respectively from haüynes of similar compositions (Bellatreccia et al., 2009).

336 4 RESULTS

337 4.1 Componentry analyses

338 Cream-white, highly vesicular pumice dominates each horizon of the deposit in both the 339 medium (46-81% of the components) and coarse (67-90%) ash fractions but gradually 340 decreases toward the top of the stratigraphic sequence mirroring an opposite increase in crystals 341 content (up to 45 %; Fig. 3). Lithics fragments are a minor but consistent component (~6%) 342 throughout the sequence, except for a peak in the basal 40 cm (Fig. 3). Pumice morphologies 343 are predominantly solid, subrounded, and equidimensional, with a small portion (\sim 5%) of tube 344 pumice. Free crystal phases primarily consist of alkali feldspar (84-99% of the total crystal 345 content), accompanied by subordinate biotite, Fe-Ti oxides, haüyne, pyroxene, and titanite, with 346 accessory olivine (Supplementary Information). Relative crystal abundances remain nearly constant throughout the sequence, except for haüyne, which progressively increases from 347 348 bottom to top, never exceeding 3% of the total crystal population (Supplementary 349 Information).

350 4.2 MI textures

A comprehensive list and description of the haüyne-hosted MIs investigated are provided in Supplementary Table 1. Pale to dark brown MIs are ubiquitous within haüyne crystals of the RP deposit. Their volumes span approximately five orders of magnitude $(10^3-10^7 \ \mu m^3;$ equivalent ellipsoid volume) and sum together can occupy a significant portion of their host crystals.

The vast majority of MIs exhibit blunted, ellipsoidal to sub-spherical morphologies (**Fig. 4a**), though mushroom-shaped inclusions are also common. These often occur in coaxial pairs

connected either to each other or to the crystal exteriors (embayments) through a tapered
bottleneck (Fig. 4b, c). In contrast, sub-planar (Fig. 4d) and fully faceted MIs are rare, with the
latter accounting for only ~2% of the inclusions investigated. Most embayments display
irregular (*complex* as in Ruefer et al., 2021), sub-cylindrical (*simple*), or bulbous morphologies.
MIs of different sizes and shapes can coexist within the same haüyne crystal, but their spatial
distribution appears arbitrary, making the recognition of MIAs challenging.

364 Crystalline phases are absent within MIs, while the number and size of bubbles were the key distinguishing features for MI selection in analyses. MIs can be free of bubbles (Fig. 4a), host 365 366 a single spherical bubble (Fig. 4a), or be partially to entirely filled with a multitude of 367 microscopic bubbles (termed microvesicular MIs; Fig. 4c). Only bubble-free and single-bubble 368 inclusions which do not show any evidence of post-entrapment modification (e.g., leaking 369 through fractures) were considered for further analyses. Bubble-free MIs are the most abundant 370 overall, especially at the lower stratigraphic levels, followed by single-bubble MIs and 371 microvesicular MIs. In rare cases, MIs contain multiple bubbles (usually fewer than 10) of 372 widely variable size (Fig. 4e). Notably, MIs hosted within the same crystal exhibit generally 373 the same bubble contents.

374 The size of single bubbles scales with the volume of their host MIs (Fig. 4g), occupying on 375 average 2.39 ± 1.80 vol.% of the inclusion. In contrast, individual microscopic bubbles within 376 microvesicular clusters contribute at most 0.23 vol.% ($0.04 \pm 0.06 \text{ vol.}\%$ on average; Fig. 4h). 377 The large standard deviation arises from the assumption of an equivalent ellipsoidal volume for 378 MI volume estimation. For multiple-bubble MIs, a total bubble volume is calculated as the sum 379 of all individual bubbles within the same MI-an approach that resulted unfeasible for 380 microvesicular MIs. The total bubble volume in multiple-bubble MIs scales similarly to that of 381 single-bubble MIs (Fig. 4g), resulting in comparable volume occupancies (Fig. 4h).

Embayments frequently contain multiple large bubbles (10.77 ± 0.78 vol.% on average) that tend to concentrate near the outlet where they adopt ellipsoidal to droplet-like shapes (**Fig. 4f**).

384 4.3 Glass geochemistry

385 Melt inclusions have a homogeneous trachytic-phonolitic composition, containing $\sim 61 \pm 0.3$ 386 wt.% SiO₂ and elevated alkaline elements concentrations (Fig. 5). The agpaitic index 387 ([Na+K]/[A1]) ranges between 0.99 and 1.07 with an average at 1.02 ±0.02 while the Mg# ([Mg]/[Mg+Fe]) is 0.22 ±0.01 on average. The full range of sample and reference material 388 389 chemical compositions is provided in Supplementary Table 2. Elemental comparisons reveal 390 no evident trends, except for a positive correlation between CaO and MgO and a negative 391 correlation between Na₂O and K₂O (Fig. 6). Embayments and MIs share similar major element 392 compositions, with the exception of slightly higher K₂O concentrations in MIs. Within 393 individual MIs or embayments, major element compositions remain chemically homogeneous, 394 showing no systematic variation from core to rim (e.g., relative standard deviations of $0.36 \pm$ 395 0.20% for SiO₂ and $1.34 \pm 1.09\%$ for Na₂O), even for mobile elements like Cl and S in open 396 embayments. Major element concentrations in MIs remain largely consistent across 397 stratigraphic horizons (i.e., relative standard deviation range from 0.16% in FeO to 9.42% in 398 P₂O₅), though a slight increase of K₂O and TiO₂, and a decrease in Na₂O are observed from 399 base to top of the sequence (Fig. 7). The chemical composition of pumice groundmass glass 400 closely resembles that of MIs in terms of silica and most major elements (Fig. 6), but it is 401 slightly depleted in K₂O (-7.0 $\pm 1.0\%$ on average) and enriched in Na₂O (+9.4 $\pm 4.4\%$ on 402 average) (Fig. 6). In contrast, tephra glass exhibits a more systematic variation in oxide 403 concentrations from the base to the top of the stratigraphic sequence, with K₂O, MgO, TiO₂, 404 SO₃, and CaO decreasing and Na₂O and Cl increasing (Table 1; Fig. 7).

405 Water concentrations in MIs, determined via FTIR absorption, range from 2.29 wt.% to 5.88 406 wt.%, with an average of 3.63 ± 0.78 wt.% (Fig. 8). No significant difference is observed 407 between MIs with or without a gas bubble $(3.75 \pm 0.75 \text{ wt.}\% \text{ and } 3.63 \pm 0.89 \text{ wt.}\% \text{ respectively})$, 408 or across the stratigraphic sequence. In contrast, embayments contain slightly less water, with 409 an average of 3.14 ± 1.17 wt.%. The uncertainty derived from the error propagation of all 410 parameters in the Beer-Lambert equation is on average $12 \pm 4\%$, which aligns with the typical 411 uncertainty range for such estimations (e.g., 10-20%; Agrinier and Jendrzejewski, 2000; 412 Wysoczanski and Tani, 2006; von Aulock et al., 2014). Among the sources of error, thickness 413 uncertainty is a major contributor, with a $\pm 1 \mu m$ variation yielding an average ~3% error in 414 water content estimation, equivalent to 0.1 wt.% H₂O.

FTIR spectra reveal no absorbance peak at 2351 cm⁻¹ for the molecular CO₂ overtone (Brooker 415 416 et al., 1999), even in the thickest MIs, indicating that molecular CO₂ in the glass is likely below 417 the thickness-dependent detection limit, ranging from tens up to a hundred ppm (von Aulock et 418 al., 2014; Rose-Koga et al., 2021). In lower-silica and depolymerised melts, dissolved carbonate species (CO₃²⁻) dominate over molecular CO₂ in C-O species speciation (Brooker et al., 1999; 419 420 Morizet et al., 2013); however, most of our spectra exhibit significant noise below 1600-1550 421 cm⁻¹. Even in spectra with a high signal-to-noise ratio, no evident carbonate doublet is observed, which may be masked by the H_2O_{mol} peak at 1650 cm⁻¹. As a result, dissolved CO₂ 422 423 concentrations cannot be quantified for MIs, even in bubble-free inclusions, suggesting a 424 generally limited CO₂ uptake in bubbles.

425 **4.4 Crystals geochemistry**

426 *4.4.1 Haüyne*

427 Haüyne crystals are chemically homogenous, exhibiting no evident zonation even under
428 scrupulous SEM investigation. Crystals from different horizons share similar chemical

429 compositions (**Supplementary Table 2**), with the exception of a subtle, progressive increase in 430 SO₃ (wt.%) from the base to the top of the sequence. Water content within haüyne, estimated 431 via FTIR analyses, is on average of 1.53 ± 0.52 wt.%, while CO₂ concentrations range from 0.02 432 wt.% to 0.33 wt.%. The stoichiometric formula of haüyne crystals can be expressed as:

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$$(Na_{5.44-6.15}Ca_{1.61-1.87}K_{0.2-0.26}Fe_{0.04-0.05}Mg_{0.01-0.03})_{\Sigma \cong 8}(Al_{5.78-5.86}Si_{6.14-6.22}O_{24}) \cdot \\ \cdot [(SO_4^{2-})_{1.37-1.73}(Cl^{-})_{0.07-0.14}(OH^{-})_{0.18-0.50}]_{\Sigma = 2} \cdot nH_2O \cdot mCO_2$$

434 where n ranges from 0.51 to 2.68 and m from 0.01 to 0.46. The unit formula was normalised 435 assuming (Si + Al) = 12, while the anionic site sum was set to 2 to account for water occupancy. Since the overtone at ~3500 cm⁻¹ in FTIR spectra, used for water estimation, does not resolve 436 437 water speciation, all OH⁻ was assigned to the anionic site to complete a valence of 2 438 (Bellatreccia et al., 2009), with any remaining water included as molecular H₂O. Sulphur was 439 entirely allocated to sulphate (SO_4^{2-}) consistent with haüvne's stability under oxidising 440 conditions (Baudouin and Parat, 2015 and references therein) and the presence of sharp overtones at ~2200 cm⁻¹ in FTIR spectra (Bellatreccia et al., 2009). The consistent presence of 441 442 Ca in the stoichiometric formula confirms that all analysed crystals are compositionally closer 443 to the haüyne end-member in the haüyne-nosean solid solution. However, a progressive 444 increase in Ca and SO₄, coupled with a decrease in Na, is observed across the stratigraphic 445 sequence from bottom to top, indicating subtle variations along the haüyne-nosean series. 446 Crystal edges are commonly altered to unidentified zeolitic phases enriched in Si and Al, often 447 banded with Ca enrichments (Supplementary Fig. 3).

448 *4.4.2 Alkali feldspars*

The composition of alkali feldspars ranges from Ab₇₁Or₂₈An₁ to Ab₅₄Or₄₄An₂ (Supplementary
Fig. 4a; Supplementary Table 2). Textural observations do not reveal any visible chemical
zonation within the crystals (Supplementary Fig. 4a, b). Across the stratigraphic section, the

anorthite component progressively increases from base to top. Such a trend corresponds to a

gradual increase in CaO at the expense of Na₂O, while K₂O remains essentially stable.

- The bulk composition of pumice closely resembles that of the groundmass glass in terms of major elements (**Fig. 5**) but is slightly depleted in SiO₂ (-3.6 \pm 0.9 wt.% on average) and Na₂O
- 457 (-2.3 ± 0.5 wt.% on average), while being enriched in Al₂O₃ (+1.6 ± 0.5 on average). The 458 significant depletion of alkali in bulk pumices may be partially attributed to weathering, as RP 459 material exhibits noticeable alteration in hand specimens. Across the stratigraphic sequence, 460 SiO₂ content progressively decreases, whereas TiO₂, CaO, MgO, and P₂O₅ concentrations 461 increase from base to top. The other oxides remain relatively stable. Notably, the composition 462 of the basal horizon B differs significantly from the overlying horizons, displaying anomalous 463 trends in Al₂O₃, FeO_{tot}, MnO, K₂O and TiO₂ (**Fig. 5**).

464 Trace element concentrations of bulk pumice are reported in Table 1. The primitive mantle-465 normalised (McDonough and Sun, 1995) concentrations are generally higher than those reported for Rungwe basalts (Castillo et al., 2014) but exhibit similar overall trends (Fig. 9a). 466 467 The highest normalised values are observed for highly incompatible U and Th. REE 468 concentrations, normalised to the primitive mantle (McDonough and Sun, 1995), indicate 469 higher LREE concentrations relative to HREE, along with a pronounced negative Eu anomaly 470 (Fig. 9b). Across the stratigraphy, progressive increasing or decreasing trends are evident (Fig. 471 9c). Specifically, from base to top, most middle REE and LILE (except Rb) tend to increase, 472 while extreme HREE, LREE and HFSE show opposite trends. Fe-Mg substituent metals do not 473 exhibit any clear trend. Notably, several elements in the basal horizon (i.e., sample KF176-B) 474 display anomalous values compared to the rest of the sequence (Fig. 9c).

475 **5 DISCUSSION**

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4.5 Bulk rock geochemistry

To interpret the geochemical data, we considered samples collected at the base of the stratigraphic sequence (e.g., KF176-B), representative of the topmost portions of the magmatic reservoir at the moment of the eruption, while, in contrast, the top deposit horizons (e.g., KF176-O) represent the deepest part of the plumbing system that fed the eruption.

480

5.1 Reservoir Geochemistry

481 Melt inclusions entrapped in RP's haüyne occur as isolated, large inclusions and do not exhibit 482 preferential planar alignments along crystal growth zones, making the identification of distinct 483 MIAs impractical. As a result, the assumption that specific groups of MIs formed at the same 484 reservoir conditions could not be validated through petrographic observations (e.g., Bodnar and 485 Student, 2006; Esposito et al., 2014). However, the major element concentrations of haüyne-486 hosted MIs remain largely consistent across the stratigraphic sequence and between MIs located 487 at different depths within the crystals. This suggests that the magma from which the MIs were 488 entrapped was relatively chemically homogeneous at the time of haüyne crystallisation. A 489 principal component analysis of major elements does not reveal substantial variability apart from a slight increase in the magmatic evolution of MIs entrapped at the top of the reservoir 490 491 (Supplementary Information). Additionally, given the similarity of MI compositions with 492 those of tephra glass and the independence from MI size, we also exclude a boundary layer 493 effect that would have caused the inclusion to entrap a non-representative portion of melt (Faure 494 and Schiano, 2005; Esposito et al., 2014).

The chemical compositions of embayments closely overlap those of MIs, implying that once haüyne began crystallising and trapping inclusions, the magma did not undergo significant compositional changes. This is further validated by the closeness of MIs major element composition to that of the pumice matrix glass than to bulk rock compositions (**Fig. 6**), indicating that MIs likely sampled the very final, most evolved magma before the eruption. However, slight depletions in Na₂O (0.05-1.32 wt.%) and Al₂O₃ (0.19-0.78 wt.%) and

501 enrichment in K₂O (0.01-1.1 wt.%) are observed in MIs in comparison to pumice matrix glass. 502 Haüyne preferentially incorporates Na₂O and Al₂O₃ over K₂O, and a post-entrapment 503 crystallisation (PEC) at MI rims could potentially explain these chemical variations. A mass 504 balance calculation suggests that a PEC of approximately 5 ± 3 vol.% of the original MIs could 505 account for the observed differences. However, this trend is not observed for CaO, another 506 element preferentially incorporated by haüyne. This raises the question of whether PEC slightly 507 influenced MI compositions or if the surrounding melt evolved due to the crystallisation of 508 haüvne together with other phases, such as alkali feldspars. The rims of these feldspars tend 509 toward the orthoclase endmember (Supplementary Information), suggesting progressive 510 feldspar evolution, favouring K uptake over Na, relative to the timing of haüyne MI entrapment, 511 while largely ignoring Ca. However, the chemical homogeneity of MIs glass (core to rim) and 512 the absence of zonation around them in the host suggest that if PEC occurred, it played only a 513 minor role in modifying MI compositions. A complete homogenisation of MI would have 514 required, according to MI size and element diffusion, a timescale long enough to facilitate 515 faceting processes, yet haüyne-hosted MIs in our dataset predominantly exhibit blunt or 516 subrounded shapes, with fully faceted morphologies being rare (<2%).

517 A diffusion-dependent faceting process is observed in quartz-hosted MIs, even in the absence 518 of PEC, with timescales ranging from tens to hundreds of years depending on the inclusion size 519 (Gualda et al., 2012; Pamukcu et al., 2015; Ruefer et al., 2021). However, the MI size-faceting 520 time relationship described by Gualda et al. (2012b) for guartz hosts cannot be directly applied 521 to haüyne, as diffusion for elements other than silica must be considered. Additionally, we note 522 that this speedometer is temperature-dependent and inferred for more viscous rhyolitic melts, 523 which have lower diffusivities than the phonolitic-trachytic melts studied here. Higher 524 elemental diffusivities enhance faceting rates, suggesting that blunt MIs likely had shorter 525 residence times. The occurrence of sub-rounded MIs as small as 15 µm in diameter at the core 526 of crystals therefore suggests that haüyne growth and inclusion entrapment occurred shortly 527 before the eruption on geological timescales, which is further supported by the absence of 528 microlites or growth rims within the MIs.

529 Comparison with chemical concentrations of MIs entrapped in mafic and alkali feldspars hosts 530 (Fig. 6; Fontijn et al., 2013) reveals that haüyne-hosted MIs are more evolved, suggesting that 531 differentiation processes had already occurred before haüyne crystallisation began. This 532 interpretation is supported by trace element concentrations in the bulk pumice, which indicate 533 a certain degree of crystallisation within the magmatic reservoir. The progressive enrichment 534 of the most compatible trace elements with depth in the reservoir suggests advanced fractional 535 crystallisation in its shallowest portions. Elements such as Ba, Sr, and Eu-compatible in alkali 536 feldspars—were progressively sequestered during crystallisation, while incompatible elements 537 accumulated in the residual melt. Considering an extreme scenario where the bulk partition 538 coefficient (D) for highly incompatible elements (e.g., U and Th) approaches zero, the observed 539 enrichment trends from the base to the top of the stratigraphic section correspond to a minimum 540 of 60% Rayleigh fractional crystallisation. This implies that the whole-rock composition of the 541 shallowest portions of the reservoir represents the 40% residual melt of the deeper magma (Fig. 542 9d).

543 A similar conclusion is reached when comparing trends of compatible versus incompatible trace 544 elements for a range of pumice bulk partition coefficients (Fig. 9c). The only strong deviation 545 from the general trend is observed in sample KF176-B, which exhibits significantly higher 546 concentrations of Ba and other compatible elements like Sr than expected. While the lower SiO₂ 547 and total alkali, and higher FeOtot and compatible trace element concentrations in KF176-B 548 could suggest a less evolved magma, its depletion in MgO and enrichment in incompatible 549 elements relative to the rest of the deposit implies a more advanced stage of differentiation. This suggests that KF176-B may represent an older, separate, shallow mush zone in the 550

551 reservoir, where in-situ equilibrium crystallisation was dominant rather than efficient crystal 552 fractionation. Such a scenario would result in reduced fractionation of compatible elements, 553 similar to processes observed at the uppermost parts of crystalline intrusions (Namur and 554 Humphreys, 2018). Excluding KF176-B from the main trend results in a lower overall degree of fractional crystallisation (Fig. 9c, d). However, the crystal-poor topmost regions of the 555 556 magmatic chamber exhibit the highest degrees of crystallisation, consistent with an effective 557 fractionation process where crystals migrate toward the reservoir walls or settle in the crystal-558 rich lower portions (e.g., horizon KF176-O). This also supports the presence of a positive 559 thermal gradient with depth. The substantial degree of crystallisation inferred from trace 560 element trends contrasts with the major element homogeneity observed in MIs, which have 561 compositions that closely resemble those of the pumice whole rock from the most evolved 562 horizon, KF176-C (Fig. 5). This reinforces the idea that hauyne crystallised relatively late and 563 predominantly trapped a residual melt at the final stages of magma evolution.

564 Experimental studies indicated that well-shaped, euhedral crystals growing at lower cooling 565 rates tend to entrap a representative portion of parental melt in contrast to fast-growing skeletal 566 or dendritic crystals, which often contain highly irregularly shaped melt inclusions (Faure and 567 Schiano, 2005). Alternatively, crystallisation rate variability can create zonation of microscopic 568 MI clusters within the host (Cooper et al., 2015). RP haüyne-hosted MIs are large and 569 subspherical lacking distinctive zonation or clusters of MIs within individual crystals (Cooper 570 et al., 2015). Morphologies such as mushroom-shaped MIs (curvilinear in Faure and Schiano, 571 2005), which were previously observed in olivine under single-stage, low-rate cooling 572 experiments and linked to equilibrium growth (Faure and Schiano, 2005), are also commonly 573 found in haüyne. This suggests that even the most irregular MIs can form at a constant, low 574 undercooling, crystal growth, without the need for dissolution processes induced by local 575 disequilibrium, such as fluid bubble interactions.

576 The stability of haüyne in phonolitic melts from Laacher See (Germany) and Tenerife (Spain) 577 has been experimentally constrained to oxidising conditions ($fO_2 > NNO$ buffer), pressures below 200-300 MPa, and temperatures between 750 and 900 °C (Berndt et al., 2001; Andújar 578 579 et al., 2008; Baudouin and Parat, 2015). Berndt et al. (2001) observed a positive correlation 580 between haüyne stability temperature, sulphur availability, and oxidising conditions in the 581 parental melt. The sulphur accessibility thus likely plays a key role in haüyne stability and 582 growth rate (Harms and Schmincke, 2000; Berndt et al., 2001; Cooper et al., 2015). The RP 583 magma exhibits significantly higher sulphur content than other phonolitic systems, both in haüyne (e.g., ~4 wt.% less in Laacher See phenocrysts; Berndt et al., 2001) and melt (e.g., one 584 585 order of magnitude lower in Laacher See MIs and matrix glasses; Harms and Schmincke, 2000). 586 Berndt et al. (2001) inferred a late-stage differentiation of Laacher See haüyne, with oxidising 587 conditions emerging shortly before eruption due to the injection of fresh, oxidised magma. 588 Similarly, sulphur sparging from depth has been proposed as a mechanism accelerating haüyne 589 growth in Tenerife phonolites (Cooper et al., 2015). In RP haüynes, sulphur concentration 590 decreases toward shallower parts of the reservoir ($\Delta_{SO3} \approx 2600$ ppm on average) mirroring 591 trends in hosted MIs ($\Delta_{SO3} \approx 200$ ppm on average) and pumice groundmass glass ($\Delta_{SO3} \approx 400$ 592 ppm). Among crystalline phases, haüyne likely played a role in sulphur depletion in the residual 593 melt at shallower depths due to preferential uptake of sulphur (Harms and Schmincke, 2000). 594 However, given the low abundance of crystallised haüyne (roughly estimated between 0.0 and 595 0.7 wt.%), it is unlikely to have solely buffered the sulphur content in the magma. Mass balance 596 calculations suggest extremely low sulphur uptake (~38 ppm) in the topmost reservoir, whereas 597 it would be significantly more effective in the deepest portions (600-800 ppm) due to increased 598 haüyne crystallisation. Sulphur enrichment at depth may result from external inputs via gas 599 sparging (Cooper et al., 2015) or mingling with a more oxidised, sulphur-rich magma (Harms 600 and Schmincke, 2000; Berndt et al., 2001), enhancing haüyne stability in the deeper, more 601 oxidised reservoir layers. These deep volatile sources were primarily enriched in sulphur while 602 other volatiles remained constant (e.g., H_2O and CO_2) or showed opposite trends (e.g., CI^-) in 603 haüyne and hosted MIs. Additionally, small grains of pyrrhotite found included in alkali 604 feldspar, biotite, and pyroxene (Fontijn et al., 2013), suggest that initial reducing conditions 605 shifted during haüyne crystallisation, just before the eruption—similar to observations in 606 Laacher See phonolites (Berndt et al., 2001).

607 Under ideal conditions, e.g., excluding any post-entrapment modifications, MIs can retain the 608 volatile content of the original parental melt. However, volatile loss can occur, and as a rule of 609 thumb, the highest volatile concentrations measured in a set of syn-entrapped MIs are 610 considered the best estimate of original melt concentrations (e.g., Esposito et al., 2014). To 611 estimate the water content of parental melt from haüyne-hosted MIs while accounting for 612 uncertainties, we calculated an average based only on values above the third quartile of the 613 dataset distribution, yielding 4.82 ±0.58 wt.% H₂O. Upon crystal cooling, if the rate is not 614 sufficiently rapid, a single post-entrapment bubble may form due to differential contraction 615 between the melt and the host crystal (Danyushevsky et al., 2002; Cannatelli et al., 2016; Rose-616 Koga et al., 2021 and reference therein), inducing volatile exsolution. Alternatively, if a volatile 617 phase was already present in the original magma, a larger, pre-existing bubble could be trapped, 618 typically resulting in greater size variability among co-genetic MIs (e.g., Davidson and 619 Kamenetsky, 2007). The volume percentage occupied by single bubbles in haüyne-hosted MIs 620 remains small and relatively constant across individual crystals, within the same stratigraphic 621 horizon, and even throughout the deposit. This suggests that bubble formation occurred post-622 entrapment, rather than from pre-existing vapour phases in the melt (Danyushevsky et al., 2002; 623 Moore et al., 2015; Cannatelli et al., 2016; Rose-Koga et al., 2021). This interpretation is further 624 supported by the absence of fluid inclusions within haüyne crystals. Notably, bubble-free MIs 625 are the most abundant, particularly in crystals from the shallower levels of the magmatic

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626 reservoir prior to the eruption, implying rapid ascent and cooling rates. In contrast, single 627 bubbles are most common in MIs from the deeper portions of the reservoir, where longer 628 cooling times may have allowed bubble formation during the eruption.

629 Shrinkage bubbles have the potential to induce water exsolution within them (e.g., Steele-630 MacInnis et al., 2017); however, no systematic water depletion trend was observed between 631 bubble-hosting and bubble-free MIs, suggesting minimal impact on water loss. In contrast, 632 studies of shrinkage bubbles indicate that they can contain between 30% and 90% of the 633 originally dissolved CO₂ (Esposito et al., 2011; Moore et al., 2015; Rose-Koga et al., 2021 and 634 references therein; Feignon et al., 2022), highlighting their importance in assessing total CO₂ 635 concentrations in MIs. In the case of RP haüyne-hosted MIs, no molecular CO₂ was detected, 636 even in bubble-free inclusions, suggesting that CO₂ levels were already low at the time of 637 haüvne crystallisation. Nevertheless, significant CO₂ concentrations were measured in haüvne 638 crystals themselves. This discrepancy could potentially be explained by crystal uptake during 639 PEC, which could account for up to 90 ± 50 ppm if ~5 vol.% of the MIs crystallised, while the effect on water content would be minimal. However, CO2^{mol} present below FTIR detection 640 641 limits or structural CO₂ may still represent residual CO₂ within MIs, meaning the estimated PEC contribution—and thus the inferred initial CO2 concentrations for a hypothetical fully 642 643 degassed tephra matrix-represent a maximum scenario.

The formation of multiple bubbles in microvesicular MIs may result from melt second boiling during PEC, volatile leakage upon ascent, heterogeneous nucleation, or the entrapment of an exsolved fluid phase (e.g., Danyushevsky et al., 2002; Cannatelli et al., 2016; Edmonds and Woods, 2018; Jorgenson et al., 2025). Notably, when present, such inclusions share the same texture as all other inclusions within the same crystal, suggesting that they record the full crystal's history rather than individual MI fracturing, decrepitation and volatile exsolution. The presence of a pre-entrapment exsolved fluid phase alone is unlikely to explain the formation of 651 these MIs, given the absence of other clear evidence supporting this. Additionally, their overall abundance is minor and does not appear to be preferentially located at the top of the reservoir, 652 653 as would be expected if a fluid phase had pervasively tapped this region. These bubbles often 654 occupy the entire MI volume, which would imply a fully foamed magma within the reservoir 655 during haüyne crystallisation-an unrealistic scenario, as such a magma would likely be 656 unstable and prone to immediate eruption (Edmonds and Woods, 2018). While PEC could 657 contribute to second boiling, estimated crystallised volume values do not support the degree of 658 vesiculation required for widespread microvesicular MIs. Instead, heterogeneous bubble 659 nucleation upon ascent, with some contribution from PEC, appears to be the most plausible 660 explanation. Since bubble coalescence into a single bubble is a relatively fast process that can 661 take seconds to minutes (e.g., Jorgenson et al., 2025 and references therein), we hypothesize 662 that these MIs experienced a quenching rate intermediate between that of single-bubble and 663 bubble-free MIs.

664 Embayments yield, on average, lower water concentrations than sealed inclusions (Supplementary Table 2) because melt in open embayments progressively equilibrates with 665 666 the surrounding melt during ascent and is more likely to undergo diffusion-controlled re-667 equilibration during syn-eruptive degassing (Liu et al., 2007). The formation of large, tapered 668 bubbles propagating from embayment outlets is associated with syn-eruptive melt 669 supersaturation, which led to volatile degassing and vesiculation just before quenching (Ruefer 670 et al., 2021). Bubbles are present within embayments either intruding from the outlet or forming 671 as discrete bubbles. However, experimental observations have demonstrated that different 672 bubble textures can be generated by the same decompression history (Hosseini et al., 2023). 673 The concentration of bubbles at embayment outlets can be explained by the fact that melt inside 674 the embayment does not respond to decompression disequilibrium in the same manner as the 675 melt outside. Consequently, bubbles tend to nucleate in portions of embayments closer to the

676 free melt and may migrate in and out of the embayment. The preservation of supersaturated 677 conditions within embayments also inhibits decompression-induced microlite formation 678 (Ruefer et al., 2021), as evidenced by the total absence of microlites in the embayments 679 analysed in this study.

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5.2 Thermobarometry

681 To characterise the pre-eruptive conditions of the magmatic reservoir in terms of temperature 682 and pressure, we applied well-established petrological models. The entrapment temperature of 683 haüyne-hosted MIs was estimated using the alkali feldspar-liquid geothermometer from Putirka 684 (calibration error ±23 °C; Putirka, 2008). To assess equilibrium conditions, we calculated an 685 equilibrium constant (Kd; Eq. (2) in Mollo et al., 2015), between each MI glass composition 686 and the core, middle and rim portions of alkali feldspar crystals from the corresponding deposit 687 horizon. The best correlation between the resulting Kd values and the measured equilibrium 688 constant (Supplementary Material in Mollo et al., 2015) was found between MIs and the rims 689 of alkali feldspars. Consequently, entrapment temperatures were estimated by pairing each MI 690 with the rim of each crystal from the corresponding horizon, and the results were averaged to 691 express variance. Temperatures range from ~938 to 1054 °C for MIs, with an average at 975 692 ± 22 °C (Fig. 10). Overall, temperatures estimated with the Putirka (2008) geothermometer are 693 comparable but slightly higher than those previously estimated with a Fe-Ti oxide pair (i.e., 925 °C; Fontijn et al., 2013). While these temperatures are comparable, it remains unclear whether 694 695 the higher values derived from the alkali feldspar-melt thermometer result from the calibration 696 of the equations used or from the fact that the two thermometers recorded different stages of 697 the magmatic history and thermal conditions.

The saturation pressure at the moment of entrapment for a given water concentration was estimated using the solubility model from Di Matteo et al. (average relative error 12%; Di Matteo et al., 2004). To account for a potential CO₂ content in the parental melt up to 90 ppm, 701 we also applied the MagmaSat model (Supplementary Table 3; Ghiorso and Gualda, 2015; 702 Iacovino et al., 2021). Saturation pressures calculated using the Di Matteo et al. (2004) model 703 range between 39 and 127 MPa (Fig. 10). However, considering only the highest recorded water 704 concentrations as representative of the original melt conditions, an average pressure of 92 ± 15 705 MPa is obtained. Including CO₂ in the estimates yields higher saturation pressures, resulting in 706 144 ±28 MPa on average. However, we acknowledge that MagmaSat CO₂-free estimates tend 707 to overestimate saturation pressures compared to the Di Matteo et al. (2004) model by up to 43 708 MPa for water contents ~5 wt.% (Supplementary Table 2), similar to other solubility models 709 preferentially tested for calc-alkaline compositions (e.g., Di Matteo et al., 2004; Allison et al., 710 2019). This suggests that, while CO_2 was present, its influence on saturation pressure estimates 711 was likely minimal.

712 Pressure estimations from solubility models can represent reservoir lithostatic pressures only in 713 case of volatile saturation. The values obtained here are lower than previous estimates from the 714 clinopyroxene-liquid barometer (i.e., 70-350 MPa; Fontijn et al., 2013). This wide range reflects 715 the dependence of pyroxene-hosted MI pressures on water content, with the highest water 716 concentrations yielding pressures considerably greater than those recorded for haüyne-hosted 717 MIs. This could imply that the reservoir was stored at significant depths, at water-718 undersaturated conditions. However, clinopyroxene-hosted MIs likely formed earlier, as 719 indicated by their less evolved compositions, and possibly at deeper crustal levels. No evidence 720 from haüyne-hosted MIs suggests water saturation conditions during haüyne crystallisation, 721 however, the potential occurrence of second boiling due to minor PEC implies conditions were 722 not far from saturation. We interpret our estimates as a minimum storage lithostatic pressure 723 during haüyne crystallisation, corresponding to a depth of 3.4 ± 0.6 km in the crust, assuming 724 an average continental crust density of 2.7 g \cdot cm⁻³.

725 5.3 Conceptual model

Based on observations from this and previous studies, we propose a conceptual model todescribe the pre-eruptive conditions and evolution of the RP reservoir.

The RP reservoir likely differentiated from a deeper basaltic parental magma (Furman, 1995; Fontijn et al., 2013) through fractional crystallisation, producing a trachytic/phonolitic melt (**Fig. 11a**). However, as observed in other peralkaline systems along the EAR, where intermediate products are missing (e.g., Macdonald, 1987; Tadesse et al., 2019 and references therein), we do not exclude the possibility of crustal assimilation contributing to magmatic differentiation. This is supported by La/Nb values consistently above 1 (Plank, 2005).

734 Once stored in the upper crust, a temperature gradient promoted crystal fractionation, primarily 735 in the shallower portions of the reservoir, leading to the crystallisation of mafic minerals (e.g., 736 clinopyroxenes and biotite) and early alkali feldspars (Fig. 11b). Magmatic differentiation 737 through fractional crystallisation proceeded over an extended period, as indicated by trace 738 element patterns suggesting up to 50% fractional crystallisation and the presence of alkali 739 feldspar crystals exceeding 2 mm in size. Crystals that formed near the reservoir roof were 740 likely transported toward its walls and deeper regions. However, given the low crystal content 741 in ash components and lapilli clasts, it is plausible that only a small fraction of these crystals 742 was retained in the eruptible portion of the reservoir. Most crystals may have segregated from 743 the magma, similar to the crystal-poor rhyolitic magmas that leave behind batholithic crystalline 744 mushes (Bachmann and Bergantz, 2004). This is consistent with observations from other peralkaline systems in the EAR, where mush segregation rates can reach 10⁻¹-10⁻⁴ km³·yr⁻¹ 745 746 (Iddon et al., 2019).

Haüyne crystallisation likely occurred at shallower crustal depths than mafic mineral
crystallisation, during a stage when the reservoir's major element chemistry had largely

homogenised (**Fig. 11c**). Residence time at these depths must have been relatively short, as evidenced by the unfaceted shapes of MIs and the absence of breakdown or reaction rims around biotite crystals (Fontijn et al., 2013), which are known to be unstable to depressurisation-induced water loss (e.g., Fougnot et al., 1996).

753 A minimum storage depth of approximately 3.4 km is inferred under water-undersaturated 754 conditions. Water content in RP melt inclusions is relatively low compared to the estimated 755 volatile content at saturation for phonolitic magmas at crustal pressures (e.g., ~8 wt.% at 200 756 MPa; Carroll and Blank, 1997; Berndt et al., 2001) or values recorded in other EAR peralkaline 757 magmas (e.g., Wilding et al., 1993; Iddon and Edmonds, 2020). However, it aligns with 758 undersaturated values observed in other phonolitic magmas containing modal haüyne, such as 759 the El Abrigo ignimbrite in Tenerife, Spain (Andújar et al., 2008). While shallow magma 760 storage is often considered unusual, petrological evidence and direct observations (e.g., 761 boreholes encountering magma pockets at Krafla, Iceland, and Menengai, Kenya; Papale and 762 Garg, 2022), suggest that it may be more common than previously thought. Similar shallow 763 storage depths have also been inferred for other phonolitic systems in divergent, convergent, 764 and intraplate settings (e.g., Teide, Andújar et al., 2008; Andújar and Scaillet, 2012b; Vesuvius, 765 Italy, Scaillet et al., 2008).

766 Haüyne stability in the deeper portions of the reservoir may have been influenced by more 767 oxidising conditions. This could result from either (1) the input of fresh, oxidised magma or (2) 768 the upward migration of a SO₃-rich volatile phase. In the first scenario, the fresh magma must 769 have had a major element composition similar to that of the resident melt and a short residence 770 time (according to crystallisation kinetics), given the lack of detectable changes in groundmass 771 glass and alkali feldspar (and other phases) growth rims. The rare (<1% modal components) 772 grey/banded pumice found in the lapilli fraction (Fontijn et al., 2011), which is crystal-rich and 773 hosts plagioclase, has been inferred as evidence of mingling with intruding magma (Supplementary Table 2). In the second scenario, evidence for a volatile phase sparging from depth includes sulphur enrichment in both haüyne crystals and the melt at the reservoir bottom. The volatile phase must have been sulphur-rich but depleted in other volatiles, as no enrichment trends are observed for other species. Whether driven by rapid magma accumulation or external volatile input, either process could have contributed to destabilising the system and initiating the unrest that culminated in the eruption (Fig. 11c,d; e.g., Caricchi et al., 2018; Jorgenson et al., 2024).

781 Pumice clasts in the finer-grained KF176-B horizon appear darker, softer, and more altered than 782 those in overlying deposits. Reverse grading at the base of RP deposits is observed primarily at 783 outcrops southeast of Rungwe and near the vent, where isopach thickness is also slightly greater 784 (Fontijn et al., 2010b). This suggests an initial eruption phase of lower explosivity, producing a 785 fine-grained lapilli fallout deposit primarily in the south, followed by a hiatus long enough to 786 allow early alteration processes to affect the pumice glass. Major and trace element 787 concentrations indicate that these products experienced a distinct evolution compared to the 788 main RP magma, possibly tapping a shallow residual from previous activity. Subsequently, the 789 peak in lithics in KF176-C may correspond to a conduit-clearing event in the eruption's initial 790 stages, consistent with the observed reverse grading (Fontijn et al., 2011), before transitioning 791 into the main explosive phase.

Comparative studies of phonolitic-trachytic systems suggest water-saturation alone is not definitive in triggering explosivity, while its proximity and relation with reservoir depth appear to be relevant factors in influencing eruptive explosivity (Andújar and Scaillet (2012a). In contrast, rheology at storage conditions does not appear to directly control eruption style, as peralkaline systems with similar viscosities have produced both effusive and highly explosive eruptions (Andújar and Scaillet, 2012a). At a given temperature high-alkali silicic magmas yield viscosities more than one order of magnitude lower than their sub-alkaline counterparts (Di 799 Genova et al., 2013). Low viscosities help decouple volatiles from magma and theoretically 800 promote efficient outgassing and mildly explosive eruptions (Cassidy et al., 2018). For RP magmas at storage conditions, an average viscosity of $10^{3.45 \pm 0.33}$ Pa·s using the Giordano et al. 801 (2008) model. At equivalent temperatures, these viscosities are comparable to those of other 802 803 explosive phonolitic/trachytic melts (Andújar and Scaillet, 2012a) but also to those of lathitic 804 or shoshonitic magmas that have produced Strombolian or lava-fountaining activity (Di Genova 805 et al., 2013). Volatile loss during ascent significantly impacts magma viscosity. For RP magmas, 806 decreasing water content to anhydrous conditions increases viscosity by up to four orders of magnitude (up to 10^6 Pa·s). Despite this increase, viscosities remain below general values for 807 808 strain-induced brittle fragmentation (> 10^8 Pa·s; Papale, 1999). Additionally, the aphyric nature 809 of RP pumice clasts and their low phenocryst content suggest that crystallinity had little 810 influence on magma rheology (Dingwell et al., 1993).

Water content in the RP reservoir was relatively low but possibly close to saturation. The high bubble number density in pumice clasts (Cappelli et al., 2025) suggests that rapid ascent and syn-eruptive bubble nucleation significantly influenced magma rheology. Thus, conduit dynamics likely played a key role in preventing outgassing and driving brittle magma fragmentation, as observed in other relatively low-viscosity explosive eruptions (e.g., Houghton et al., 2004; Hughes et al., 2017).

817 6 CONCLUSIONS

We propose that the RP magma was stored at relatively shallow depths under conditions close to saturation. The injection of an S-rich gaseous phase into the reservoir likely induced haüyne crystallisation and contributed to system destabilisation. Shortly after, as inferred from the unfaceted nature of haüyne-hosted melt inclusions, the eruption commenced. The interplay between ponding depth and saturation conditions may have played a key role in driving explosivity. However, further investigations of conduit dynamics are needed to fully unravel 824 the degassing and fragmentation processes that ultimately dictated the magma's eruptive 825 behaviour beyond the conditions initially acquired at the reservoir level.

826 Across the East African Rift, several peralkaline silicic systems have produced violent 827 eruptions, sometimes culminating in caldera-forming events while others have exhibited less 828 violent effusive activity despite similar magmatic compositions (e.g., Leat, 1984b; Macdonald 829 and Scaillet, 2006; White et al., 2012). Although the eruptive volumes of these explosive events are generally modest (~1 km³ DRE max) compared to arc magmatism, the transitional nature 830 831 of such systems still poses a significant hazard to densely populated regions. We emphasize the 832 importance of a comprehensive assessment of pre-eruptive conditions, as individual 833 parameters—such as water content or magma viscosity—may be insufficient to predict eruption 834 style. Relying solely on these factors could lead to an underestimation of the explosive potential 835 of a given volcanic system.

836 DATA AVAILABILITY STATEMENT

The data underlying this article are available in the article and in its online supplementary material. Additionally, the data underlying this article are available in EarthChem Library, at <u>https://dx.doi.org/[doi]</u>

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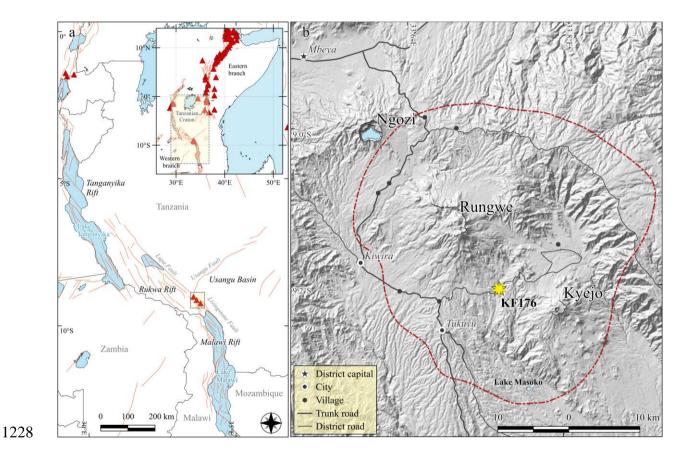
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1221 FIGURE CAPTIONS

Fig. 1: a) Location of the Rungwe Volcanic Province (*yellow box*) at the junction between the Eastern and Western branches of the EAR. Major rift faults (*red lines*) and volcanoes (*red triangles*) are indicated; **b)** shaded relief map of the RVP. The extension of the 25 cm isopach of the Rungwe Pumice fallout deposit is indicated by the red, dashed line (modified after Fontijn et al., 2011) while the location of the type section KF176 is indicated with the yellow star (12 m resolution DEM data: ASF DAAC, ALOS PALSAR RTC High res 2010; © JAXA/METI).



1229Fig. 2: Typical appearance of FTIR spectra for haüyne crystals and MIs volcanic glasses from1230our sample set. The main peaks for total water, molecular water, CO_2 and SO_4 are reported,1231while the inset zooms into the Near-IR region where the small molecular water and OH^- peaks1232are found.

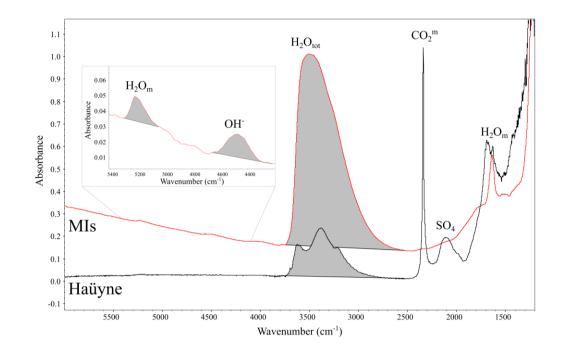
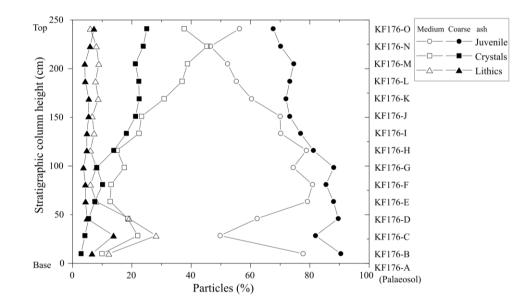


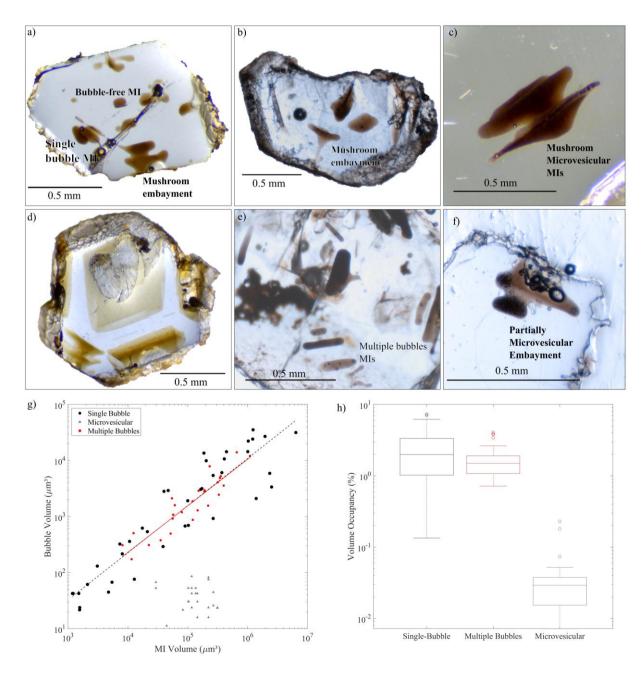
Fig. 3: Variations of componentry abundance across the Rungwe Pumice deposit for themedium and coarse ash grain sizes.



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Fig. 4: Panels a) to f) show haüyne crystals hosting various MI types, including a) ellipsoidal single-bubble or bubble-free MIs; a) and b) mushroom-shaped embayment; c) microvesicular MI; d) large, sub-planar MIs; e) elongated MIs hosting multiple bubbles of variable size; f) embayment hosting several large bubbles connected to the outside and microvesicular in its innermost portions. Panels g) and h) illustrate the relationship between MI and bubble volumes

for different bubble-hosting MI types. For each multiple-bubble MIs, the total bubble volume is considered as the sum of all individual bubble volumes within it, while for microvesicular MIs, individual bubble volumes are shown. **g**) Scatter plot showing the linear relationship between MI and bubble volumes. Linear trends are represented by dashed black and solid red lines for single- and multiple-bubble MIs, respectively; **h**) Box-and-whisker plot displaying the statistical distribution of bubble volume occupancy for different MI types. The horizontal line represents the median, while whiskers enclose 90% of the dataset, and circles indicate outliers.



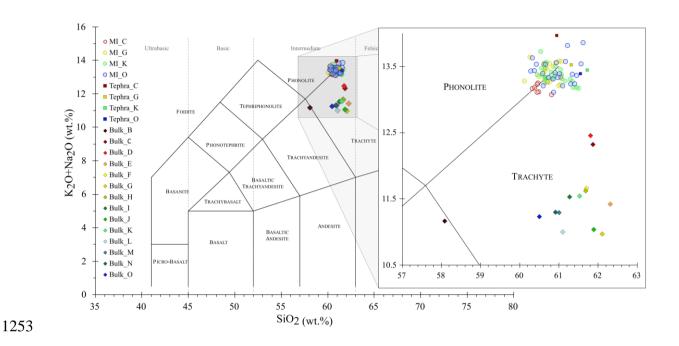


Fig. 6: Comparison of major element oxide concentrations of bulk rock, pumice matrix glass
and haüyne-hosted melt inclusions. For reference, compositions of MIs hosted in various
mineralogical phases after Fontijn et al. (2013) are also reported.

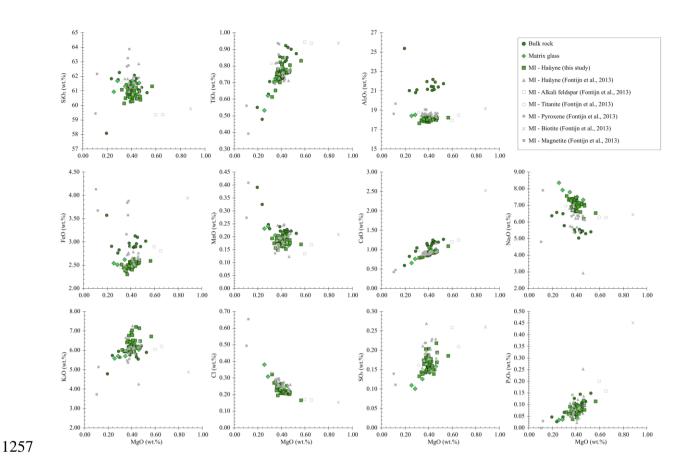


Fig. 7: a) and b) major element binary plots for melt inclusions and embayments; c) changes of major element concentrations across the deposit for melt inclusions, embayments and tephra matrix glass. The *red* and *blue* curves show mean values for each horizon for melt inclusions and embayments respectively.

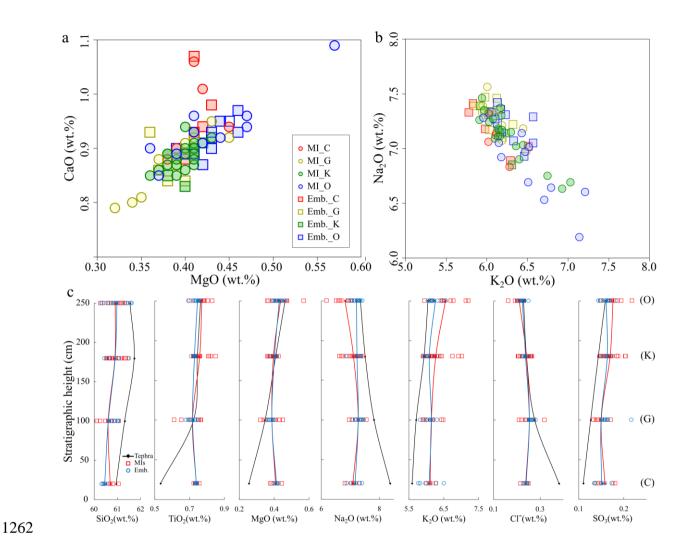
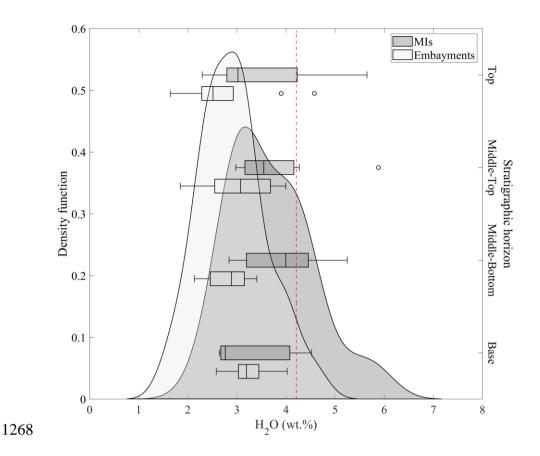
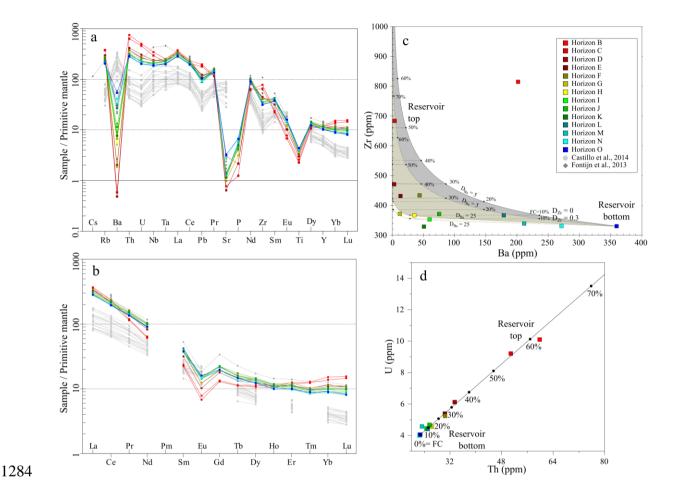


Fig. 8: *left axis*: Kernel density estimations of MIs and embayments water concentrations. The red dashed line indicates the uppermost quartile limit of H_2O distributions for MIs; *right axis*: Box-and-whisker plots of H_2O concentrations within MIs and embayments for the different horizons of the RP type section. The *inner line* indicates the median, w*hiskers* delimit the lower and upper quartiles, while *circles* indicate outliers.

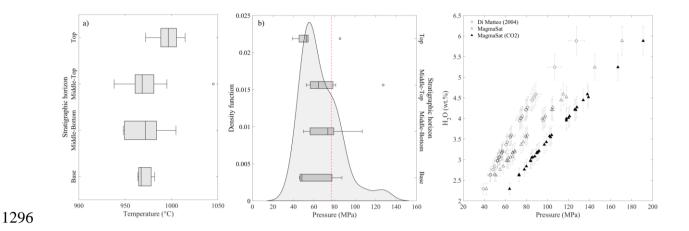


1269 Fig. 9: a) Spider diagram of primitive mantle-normalised (McDonough and Sun, 1995) trace 1270 elements and **b**) REE for pumice bulk rock of the different stratigraphic horizons, compared to values available in the literature for RP pumice bulk rock (Fontijn et al., 2013) and for older 1271 1272 Rungwe basalts (Castillo et al., 2014); c) comparison of the variation of concentration across 1273 the stratigraphic horizons for incompatible (i.e., increases from bottom to top of the magmatic 1274 reservoir; e.g., Zr) and compatible (i.e., decreases from bottom to top of the magmatic 1275 reservoir; e.g., Ba) trace elements. Hypothetical variations of element concentration as a 1276 function of fractional crystallisation (FC%) were computed assuming the horizon O as the less 1277 evolved (initial) liquid composition. Two cases were considered: the maximum (bulk partition coefficient, $D_{Zr} = 0$; grey area) and a lesser ($D_{Zr} = 0.3$; yellow area) incompatibility of Zr while 1278 1279 the compatibility of Ba was ranged between $D_{Ba}=5$ and $D_{Ba}=25$. The bulk partition coefficient 1280 ranges were selected as the best areas enclosing data points after iterative attempts; d) 1281 comparison of the concentration variation across the stratigraphic horizons for two

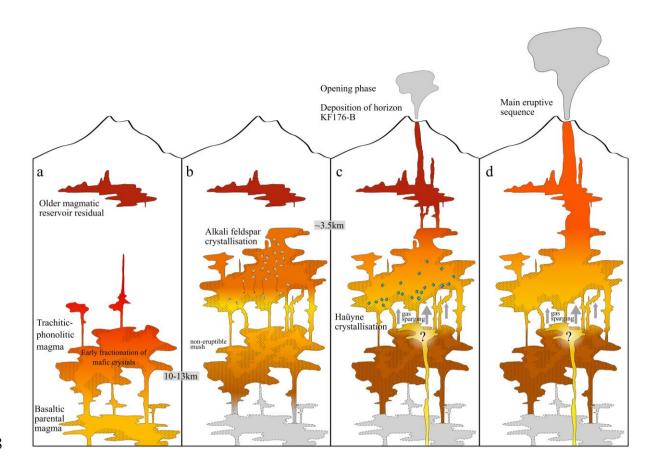
- 1282 incompatible elements. A hypothetical FC trend was traced assuming a total incompatibility (D
- 1283 = 0) for both elements. The legend shared by all diagrams is shown in panel c).



1285 Fig. 10: a) Box-and-whisker plots of temperatures estimated for MIs using the alkali feldspar-1286 liquid geothermometer (Putirka, 2008) across the stratigraphic sequence. The inner line represents the median, whiskers delimit the upper and lower quartiles and circles indicate 1287 1288 outliers; b) left axis: Kernel density estimations of saturation pressures calculated using the Di 1289 Matteo et al. (2004) model. The red dashed line marks the lower pressure limit estimated from 1290 the upper quartile of H₂O distributions in MIs analyses; right axis: Box-and-whisker plots of 1291 saturation pressures across the RP type section. The inner line indicates the median, whiskers 1292 delimit the lower and upper quartiles, while *circles* indicate outliers; c) comparison of Di Matteo 1293 et al. (2004) and MagmaSat solubility models. MagmaSat results including 90 ppm of CO₂ are



1297 Fig.13: Schematic conceptual model of the proposed plumbing system dynamics preceding the 1298 Rungwe Pumice eruption. a) A magmatic batch evolves at depth from a presumably basaltic 1299 primitive magma forming a peralkaline trachytic melt. Early fractionation of biotite and 1300 pyroxenes occurs at a depth of up to 13 km (Fontijn et al., 2013); b) as magma ascends through 1301 the crust, alkali feldspars fractionation promotes the formation of a crystalline mush at the badse 1302 of the reservoir; c) gaseous phases sparging from depth induce haüyne crystallisation at shallow 1303 depths, likely destabilising the system and triggering the eruption. The role of a deep-rising 1304 magmatic plume remains uncertain. The eruption begins with a lower-intensity opening phase 1305 involving a shallower, older reservoir that underwent a distinct magmatic evolution; d) 1306 Eventually, prolonged gas sparging and/or magma input leads to the onset of a highly explosive 1307 eruption.



TABLES

 Table 1

 Major element compositions obtained with EPMA of pumice matrix glass of five different horizons of the Rungwe Pumice tephra deposit. Major and trace element composition of pumice bulk rock for the whole deposit were analysed with ICP-OES and Q-ICP-MS respectively. Bulk rock major element oxides are normalized to 100%.

Sample	Matrix glass					Bulk rock													
. 0/	KF176-C F	F176-D F	F176-G F	KF176-K I	KF176-O	KF176-B 1	KF176-C 1	KF176-D	KF176-E 1	KF176-F 1	KF176-G 1	KF176-H	KF176-I	KF176-J I	KF176-K	KF176-L I	KF176-M 1	KF176-N 1	KF176-
vt.% 5iO2	(1.0	(17	61.3	(17	(1.6	58.08	C1 04	61.77	62.27	(1.67	62.08	(1.(((1.25	61.86	61.50	61.08	60.98	60.88	60.4
	61.0	61.7		61.7	61.6		61.84			61.67		61.66	61.25						
iO2	0.534	0.623	0.723	0.745	0.764	0.551	0.479	0.630	0.707	0.770	0.776	0.827	0.901	0.834	0.852	0.913	0.857	0.874	0.9
12O3	18.45	18.56	18.42	18.53	18.46	25.39	21.03	20.84	21.11	21.17	21.45	21.07	21.40	21.45	21.14	21.91	21.97	21.76	22.
eO	2.55	2.51	2.63	2.51	2.58	3.57	2.91	2.76	2.83	2.91	2.88	2.90	2.90	2.89	2.96	3.10	2.97	3.02	3
4nO	0.233	0.242	0.207	0.163	0.168	0.392	0.326	0.248	0.231	0.222	0.224	0.219	0.222	0.213	0.228	0.221	0.240	0.214	0.2
1gO	0.26	0.29	0.35	0.41	0.46	0.20	0.24	0.29	0.30	0.39	0.42	0.43	0.48	0.44	0.45	0.46	0.39	0.53	0
aO	0.66	0.77	0.84	0.94	1.01	0.59	0.83	0.96	1.05	1.11	1.11	1.17	1.20	1.17	1.21	1.20	1.16	1.27	1
la2O	8.37	7.94	7.82	7.51	7.34	6.38	6.58	6.50	5.78	5.39	5.05	5.55	5.30	5.39	5.45	5.46	5.54	5.40	5
2O	5.59	5.71	5.71	5.93	6.05	4.80	5.74	5.96	5.65	6.27	5.94	6.08	6.24	5.66	6.10	5.56	5.76	5.90	5
21	0.382	0.310	0.277	0.242	0.229														
O3	0.11	0.10	0.13	0.14	0.16														
2O5	0.035	0.037	0.061	0.085	0.096	0.05	0.03	0.05	0.07	0.09	0.07	0.09	0.11	0.09	0.11	0.11	0.13	0.15	0.
l'otal	98.12	98.78	98.47	98.92	98.86	norm.	norm.	norm.	norm.	norm.	norm.	norm.	norm.	norm.	norm.	norm.	norm.	norm.	nor
H2O (b.d.)	1.88	1.22	1.53	1.08	1.14														
.OI						6.97	6.09	5.79	6.43	6.87	7.70	6.77	7.87	7.23	6.91	8.06	8.00	7.77	8
pm																			
ic						13.50	10.49	8.82	8.30	8.89	7.14	7.10	7.05	6.89	7.43	7.34	6.99	7.11	7
,						6.32	22.92	10.22	20.65	16.94	21.70	12.01	23.55	16.59	33.38	12.75	25.08	14.15	18
'n						bdl	0.17	1.08	bdl	3.36	bdl	1.81	0.94	0.54	0.18	0.91	bdl	0.33	(
o						0.33	0.15	0.26	0.19	0.65	0.29	0.30	0.69	0.40	0.59	0.65	0.37	0.68	(
i						bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	
u						15.33	9.11	29.80	8.60	10.94	5.41	8.98	9.29	9.24	5.03	7.52	4.90	5.52	4
'n						175.71	115.40	100.73	79.74	86.25	68.83	104.11	74.57	79.63	81.91	79.25	73.83	71.48	70
la						57.03	49.20	51.30	50.22	53.68	47.38	48.66	49.71	50.58	47.14	51.37	46.82	46.95	47
tь						179.08	225.89	177.70	163.26	166.19	145.02	145.69	141.47	148.61	133.17	140.89	123.87	124.83	122
r						60.79	15.25	12.65	19.92	28.27	20.05	21.41	26.79	25.77	22.64	27.42	25.29	55.25	64
						51.64	49.32	47.88	48.49	52.58	46.35	47.93	47.65	49.37	44.05	48.52	45.30	43.68	43
r						814.60	683.68	471.29	431.77	434.09	371.99	366.71	353.08	370.99	328.65	366.56	338.92	331.12	330
īЬ						227.58	194.45	157.28	150.21	152.95	135.22	136.57	131.89	137.79	121.98	136.36	124.80	121.78	123
a						201.52	3.83	3.17	13.35	43.91	12.21	35.37	59.77	75.27	50.64	178.75	211.14	271.03	359
a						240.50	234.77	217.15	214.12	221.15	193.89	197.66	194.37	203.17	181.44	201.92	185.51	183.67	185
e						395.08	345.25	364.91	368.51	392.78	347.20	361.03	349.06	368.15	330.03	365.79	337.09	322.46	327
r						30.19	29.16	34.75	36.94	40.62	35.97	37.77	37.47	38.83	34.87	38.50	35.38	34.06	34
id.						80.15	77.00	103.37	114.27	129.81	113.25	122.11	121.44	125.67	111.73	125.23	112.36	110.11	113
m						9.57	9.08	12.81	15.42	17.05	15.08	16.46	16.49	16.95	15.46	17.17	15.01	14.89	15
u						1.20	1.04	1.57	1.88	2.20	1.86	2.29	2.27	2.38	2.21	2.35	2.23	2.30	2
ld						7.30	7.02	9.70	10.55	12.07	10.44	11.80	11.77	11.88	10.47	12.16	10.76	10.65	10
b						1.12	1.11	1.32	10.55	12.07	1.49	1.55	1.58	1.58	1.44	12.10	1.44	1.34	1
b)y						7.61	7.20	8.38	9.06	9.84	8.84	9.21	8.97	9.61	8.29	9.34	8.67	8.18	8
0						1.64	1.54	1.65	1.59	1.76	1.67	1.72	1.65	1.71	1.59	1.72	1.56	1.47	1
r						5.49	5.16	4.83	4.96	5.41	4.69	4.78	4.60	5.01	4.31	4.82	4.49	4.20	2
m						0.87	0.84	4.85	4.96 0.66	0.69	4.69	4.78	4.60 0.64	0.64	4.51 0.59	4.82	4.49 0.57	4.20	(
m b						0.87 6.69	0.84 6.05	5.10	0.66 4.77	5.02	4.33	0.65 4.39	0.64 4.30	0.64 4.55	4.08	0.65 4.42	4.05	0.57 3.80	1
u c						1.06	0.99	0.72	0.73	0.75	0.65	0.64	0.61	0.70	0.56	0.67	0.59	0.54	
if a						15.16	13.10	9.41	8.59	8.71	7.55	7.36	7.19	7.58	6.51	7.43	6.79	6.41	(
						9.35	8.09	8.84	8.93	9.38	8.42	8.53	8.27	8.61	7.58	8.45	7.70	7.39	
b						29.29	26.79	18.09	17.03	17.21	14.69	14.90	14.37	15.17	13.43	14.49	14.01	13.96	15
ĥ						59.84	50.87	33.45	30.40	30.46	26.31	25.69	24.64	25.59	22.61	25.07	23.28	22.50	2
U						10.10	9.21	6.12	5.38	5.24	4.56	4.67	4.45	4.67	4.04	4.41	4.58	3	.99

1315 Supplementary Information

- a. Supplementary Table 1: Melt inclusions and embayment catalogue comprehensive of detailed descriptions of qualitative and quantitative features (available upon request to lorenzo.cappelli@ulb.be).
- b. Supplementary Table 2: Chemistry data for melt inclusions, embayments, mineralogical phases and grey pumice glass. Results for the standards used as a reference for EPMA and mass spectrometry are also reported as well as the instrument conditions used for EPMA (available upon request to lorenzo.cappelli@ulb.be).
- c. Supplementary Table 3: Results of thermobarometry models used to estimate the reservoir conditions. A summary of each barometry method used is reported together with the estimated storage depth for each (available upon request to lorenzo.cappelli@ulb.be).
- 1327
- 1328 d. Componentry Analyses:

1329 **Detailed methodology:**

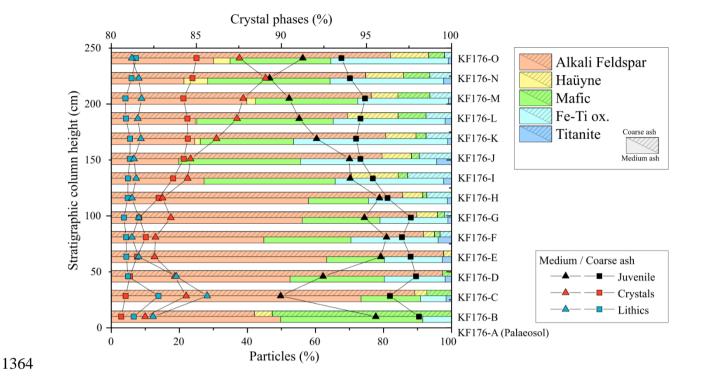
1330 The different types of components that constitute the coarse ash portion of the Runge Pumice 1331 deposit were investigated for each subsample of the type section (KF176-B to KF176-O). The 1332 $0.355-0.500 \text{ mm} (1.5\varphi-1\varphi)$ and the $0.710-1.000 \text{ mm} (0.5\varphi-0\varphi)$ grain-size ranges were selected 1333 for the observations (hereafter termed medium and coarse ash fraction respectively). The 1334 fractions were partitioned up to six times using a sample splitter to reduce the sample size and 1335 still maintain a representative and functional amount. With the aid of a stereomicroscope, the 1336 clasts were characterised and allocated to 3 classes, i.e. juvenile, lithics and crystals (further 1337 subdivided into feldspars, mafic minerals, Fe-Ti oxides, haüyne and titanite), until a total of 1338 3000 clasts per sample were categorised.

1339 **Results:**

The coarse ash fraction of the deposit analysed is composed of three broad clast types: i) juvenile, ii) lithics, and iii) crystals. Juvenile clasts are cream-white highly vesicular pumices. Clast shapes are mostly solid, equidimensional and subrounded but, according to the size and shape of vesicles, can be more elongated or irregular. Lithic clasts include rock fragments of

1344 different nature extracted from the volcanic edifice, older lava deposits, and holocrystalline 1345 rocks. Free crystal phases include alkali feldspar, haüvne, pyroxene, biotite, Fe-Ti oxides, 1346 titanite, and accessory olivine. In the coarser ash fraction, the juvenile class is dominant in each 1347 layer of the deposit (always > 65% of the components), but it is substantially less dominant in the medium ash fraction (between 45% and 80% of the components) where the crystal content 1348 1349 reaches up to the 45% of the components (Fig. 1SI). A gradual increase of the crystal content 1350 is observed from the bottom to the top of the stratigraphic section, particularly for the medium 1351 ash fraction, associated with an opposite trend of the juvenile content (Fig. 1SI). The lithic 1352 component represents a small fraction of both analysed grain sizes (3 - 12%) and it is almost 1353 constant along the stratigraphic section, except for a peak around 25 cm from the bottom 1354 (samples KF176C-KF176D) where it reaches up to 28% of the components (Fig. 1SI).

1355 On average, $88\% \pm 3\%$ and $96\% \pm 2\%$ of the crystal component within the medium and coarse 1356 ash fractions respectively are represented by alkali feldspars (Fig. 1SI). Mafic minerals 1357 (pyroxenes and biotite) are the second most abundant crystalline component (between 3.4% 1358 and 10.5%) followed by Fe-Ti oxides (between 1.5% and 9.0%). Relative crystal abundances 1359 are almost constant throughout the stratigraphic column, apart from haüyne which shows a 1360 progressive increase from bottom to top, even though it never represents more than 3% of the 1361 crystal population (Fig. 1SI). The medium ash contains a smaller amount of haüvne, which is 1362 absent in the bottom 150 cm of the deposit (Fig. 1SI). Titanite crystals are found only in the 1363 medium ash and in small numbers (always < 0.9%).



Supplementary Figure 1: Variations of componentry occurrence across the Rungwe Pumice deposit (*markers*). The top x-axis illustrates the cumulative distribution of the different crystalline phases (*bars*) within the crystal component. As the alkali feldspars always represent at least 80% of crystals, the axis origin was set at this value. For both, the medium and coarse ash grain sizes are reported separately.

1371 e. MI water content analyses:

1372 **Detailed methodology:**

To quantify the total amount of water dissolved within MI glasses we used the transmission Fourier Transformed Infrared spectroscopy technique (FTIR; §3.2.2). The FTIR method allows quantifying the sample absorbance for specific wavelengths in the near-/mid-infrared light range (usually between 1400 and 8000 cm⁻¹; von Aulock et al., 2014), which is caused by the vibration of specific molecules (analytes). The relationship between light absorption and the

1378 analyte concentration in the sample is described by the *Beer-Lambert Law*, which can be1379 rewritten to express the analyte mass fraction (Stolper, 1982):

67

$$w = \frac{A \cdot M}{\epsilon \cdot l \cdot \rho} \times 100 \tag{1}$$

1380 where *w* is the analyte concentration in wt.%, *A* (dimensionless) the absorbance of the sample, 1381 corresponding to the height (or area) of specific peaks in the FTIR spectrum (§ 3.2.2), *M* the 1382 molecular weight of the analyte (e.g., 18.015 g·mol⁻¹ for H₂O), ϵ (1·mol⁻¹·cm⁻¹) the composition-1383 dependant absorptivity index of the sample for the analyte of interest, *l* (cm) the thickness of 1384 the sample crossed by the beam, and ρ the density of the sample (in g·1⁻¹).

1385 *Sample preparation*

1386 Alkali feldspar crystals in the RP deposit host MIs and embayments (i.e., inclusions open on 1387 one side), however, their lamellar shapes and tiny volumes made them impractical to be used 1388 for this study. MIs hosted in pyroxenes and Fe-Ti oxides were hard to identify due to the dark and opaque colour of the crystals. Haüyne crystals instead tend to contain visibly more MIs 1389 1390 than the other mineral phases and therefore represented the best target for this investigation. 1391 Five stratigraphic horizons representative of the bottom (KF176-C), middle (KF176-G and 1392 KF176-K), and top (KF176-O) sequences of the deposit type section were selected. For each 1393 layer, several pumice clasts in the 11.3-16 mm $(-3.5\varphi/-4\varphi)$ grain-size range were manually 1394 crushed. Over 100 haüyne crystals were handpicked from the crushed material in the 0.5-2 mm 1395 $(1\varphi/-1\varphi)$ grain-size range and subjected to careful observation and sample selection. The 1396 crystals were placed on a glass slide and embedded in a thermosetting resin (Crystalbond™ 1397 509). An exploratory first polishing of the crystal surfaces allowed a detailed description of MI 1398 features and recognition of potential melt inclusion assemblages (MIAs, i.e., groups of coeval 1399 MIs originated from the same magma at the same reservoir conditions; Bodnar and Student, 1400 2006; Esposito et al., 2014) which helps in describing the history of both the MI and MI's host.

1401 Moreover, FTIR measurements require a clear path large enough to let the infrared beam pass 1402 through the sample to avoid any contamination of the spectrum. Therefore, small MIs (i.e., 1403 smaller than 15 μ m, the minimum infrared beam aperture) or MIs hosting too many bubbles 1404 were discarded (§ 4.2).

1405 To ensure that only the MI was crossed by the infrared beam, the crystals hosting MIs or 1406 embayments suitable for analyses were polished on two sides in a way that two parallel surfaces 1407 of the selected MIs were exposed (doubly exposed; von Aulock et al., 2014). Firstly, the crystals 1408 were grounded on one side with a 6 µm diamond paste until one or more MIs were properly 1409 exposed. Final polishing was achieved with progressively finer-grade diamond pastes (3 µm 1410 and 1 µm). Finally, the crystal was gently removed from the resin and flipped over to expose 1411 and polish the opposite side of the MI following the same process. This way we obtained fragile 1412 crystal wafers tens of micrometres thick. Eventually, a variable number of MIs per layer was 1413 successfully prepared because of crystal availability and MIs' suitability for analyses, for a total 1414 of 86 MIs and embayments.

1415 Infrared absorption

1416 The transmission FTIR spectra of MIs were acquired at the ISTO-CNRS laboratory (Orleans, 1417 France) using a Nicolet 6700 FTIR spectrometer connected to a Nicolet Continu μ m Infrared 1418 Microscope equipped with a MCT IR Detector Module. The acquiring setup consisted of a 1419 Globar light source, an XT-KBr beamsplitter and a 32x infinity-corrected Schwarzschild 1420 objective matching to a 32x condenser. The crystals were gently removed from the resin after 1421 heating on a hot plate, then rinsed in acetone to remove any resin residuals and placed over a 1422 CaF₂ window.

1423 The fundamental stretching vibration of O-H bonds in H_2O and OH^- absorbs infrared light at 1424 the 3550 cm⁻¹ wavenumber (**Fig. 2**). The height (or area) of this characteristic overtone peak in

1425 IR spectra is directly proportional to the total concentration of water in the sample, and usually, 1426 it retains a good signal-to-noise ratio. However, this overtone can be easily saturated at 1427 relatively high water concentrations unless the thickness of the sample is extremely thin (von 1428 Aulock et al., 2014). In that case, water concentration can be measured by the combination of 1429 the absorbance band of the hydroxyl group (OH⁻; 4500 cm⁻¹) with the band of molecular water (5200 cm⁻¹). Therefore, we acquired spectra in the 1200-6000 cm⁻¹ wavenumbers unit range 1430 with a spectral resolution of 4 cm⁻¹ (Fig. 2). The aperture of the square beam was set at 20x20 1431 1432 µm or 40x40 µm when the size of the MI allowed it, while a 15x15 µm beam aperture was 1433 exceptionally used for the smallest MIs. For each spectrum, 256 (512 for the 15x15 µm beam 1434 aperture) acquisitions were collected. A background spectrum on the CaF₂ window was 1435 collected for calibration before each sample analysis, and every time the spectra showed 1436 abnormal trends due to environmental changes in temperature, CO₂ concentration or humidity. 1437 We chose to use peak heights instead of areas as they are less subjected to peak distortions, especially for the combination bands. Finally, we adopted a linear baseline to measure peak 1438 1439 intensity. A minimum of three scans were collected for each melt inclusion for reproducibility 1440 which were then averaged through the data processing tools of Omnic[™] software to reduce the 1441 spectral noise. FTIR spectra were acquired at the centre of MIs but also closer to rims to 1442 evaluate the consistency of the estimated water content, while embayments were scanned in 1443 their most internal portions.

1444 The molar absorptivity index in Eq. (1) is a composition-dependent constant that can be 1445 determined for each glass composition once the water content is constrained with other 1446 analytical methods (e.g., Karl Fisher titration or secondary ion mass spectrometry). However, 1447 absorptivity indices present in the literature for glasses with a chemical composition similar to 1448 that of the sample can be applied with good approximation (von Aulock et al., 2014). For high-1449 alkali trachytic glasses a value of $66.9 \pm 5.8 \ 1 \cdot mol^{-1} \cdot cm^{-1}$ was experimentally determined for

the 3550 cm⁻¹ overtone (Behrens and Hahn, 2009) while absorptivity indices for the 4500 cm⁻¹ and 5200 cm⁻¹ overtones (ϵ_{4500} and ϵ_{5200}) were estimated through a multiple linear regression based on the absorbance values of the 4500 cm⁻¹ and 5200 cm⁻¹ peaks (A_{4500} and A_{5200} respectively), according to the following relationship (Di Matteo et al., 2004):

70

$$\frac{w_{3550}}{100} = \frac{1}{\epsilon_{4500}} \cdot \frac{A_{4500}M}{l\rho} + \frac{1}{\epsilon_{5200}} \frac{A_{5200}M}{l\rho} \quad (2)$$

where w_{3550} is the water content (in wt.%) calculated with the 3550 cm⁻¹ band and used as a 1454 1455 fixed term for the regression. The regression was performed including those spectra that did not 1456 reach saturation and at the same time showed well-shaped combination bands. It was iteratively 1457 reproduced by systematically excluding those spectra values that deranged the interpolation until a satisfying goodness was achieved ($r^2 \approx 0.9$). The regressed molar absorptivity indices 1458 and their 90% confidence bounds (subscript and superscript values) measured $1.35_{1.01}^{1.71}$ l·mol⁻ 1459 1 cm⁻¹ and $1.03_{0.69}^{1.38}$ l·mol⁻¹ cm⁻¹ for the 4500 cm⁻¹ and 5200 cm⁻¹ overtones respectively. These 1460 1461 indices are similar but slightly different from those previously estimated for trachytic (1.58 1·mol⁻¹·cm⁻¹ and 1.36 1·mol⁻¹·cm⁻¹ respectively; Di Matteo et al., 2004) and phonolitic (1.25 1462 1·mol⁻¹·cm⁻¹ and 1.10 1·mol⁻¹·cm⁻¹ respectively; Carroll and Blank, 1997) glass. 1463

1464 Thickness

1465 The MI thickness is a fundamental parameter that must be carefully determined as it represents 1466 one of the major sources of error in Eq. (1), especially for the thinner crystal wafers. For example, in our dataset, a difference of 1 μ m produces on average a ~3% percentage error on 1467 the water content estimation, equivalent to 0.1 wt.% H₂O. Moreover, during sample preparation, 1468 1469 it is important to monitor the thickness of the samples as this could determine which absorbance 1470 band can be used (von Aulock et al., 2014). To minimise the uncertainty introduced by 1471 operator's biases (e.g., optical focus recognition) two different methods were applied, and the 1472 results compared. Firstly, the thickness was measured with a flat-anvil digital micrometre

(resolution = 1 μ m; accuracy = $\pm 2 \mu$ m). A flat micrometre was chosen over a pointed 1473 1474 micrometre to minimise the risk of damaging or disrupting the extremely fragile crystals. Once 1475 we ensured that crystals were adhering properly to their glass slides, each glass slide was 1476 measured together with the hosted crystal and on its own. Crystal thickness was then obtained 1477 by difference. The measurements were repeated two or three times per crystal. Subsequently, 1478 we used an optical microscope with calibrated focus stages. A LEITZ DMR microscope 1479 equipped with an automated stage (resolution = $2 \mu m$) was used, and each crystal was measured 1480 two times for consistency.

To verify the consistency of the thickness measurements, we applied the spectral fringes counting method for those transmission spectra that showed interference fringe patterns (Nichols and Wysoczanski, 2007; von Aulock et al., 2014). This method consists of counting the spectral fringes in a specific spectral region where no absorption peaks are present (usually in the 2000-2700 cm⁻¹ range; Wysoczanski and Tani, 2006), and which relate to the thickness (*l*) following the equation (Nichols and Wysoczanski, 2007):

$$l = \frac{m}{2n(v_1 - v_2)}$$
(3)

1487 where *m* is the number of waves, *n* is the refractive index of the sample (dimensionless), and v_1 and v_2 are the highest and lowest wavenumber (in cm⁻¹) respectively of the selected spectral 1488 interval. Interference fringes were detected on both haüyne crystals and MIs, and refractive 1489 1490 indices of 1.50 and 1.52 were used respectively (Taylor, 1967; Keller et al., 1978). While a good 1491 correlation was found between thicknesses measured with the optical focus stages method and 1492 the digital micrometre, a systematic underestimation ($35.4\% \pm 18\%$ percentage error on 1493 average) was observed between the results obtained with these methods and the thicknesses 1494 estimated by the interference fringes method.

1495 Glass density

1496 The density of MI glass has been estimated as a function of its chemical composition at room1497 temperature following the equation (Lange, 1997):

72

$$\rho_{298K} = \frac{\sum X_i M_i}{V_{298K}}$$
(4)

1498 where X_i represents the mole fraction of each major oxide component (i), and M_i their molecular 1499 weights. The volume of the glass at room temperature (V_{298K}) is considered equivalent to the 1500 volume of the glass at the quenching temperature (V_{liq}) as no contraction after solidification is 1501 expected because of the volume-constrained nature of MIs and the fast cooling in the 1502 atmosphere of pumiceous tephra. Therefore, the contribution of the thermal expansion 1503 coefficient (Eq. (4) in Lange, 1997) was not considered and the glass volume at quenching 1504 temperature was estimated using the equation for iron-bearing silicate glasses, modified after 1505 Lange and Carmichael (1987):

$$V_{liq}(T) = \sum X_i(T)V_i(T) + X_{Na_2O}X_{TiO_2}V_{Na_2O-TiO_2} + X_{H_2O}V_{H_2O}(T)$$
(5)

1506 where V_i is the volume regression of major oxide components as a function of the liquid 1507 temperature with respect to the volume at the modelled temperature of 1300 °C (Lange and 1508 Carmichael, 1987):

$$V_i(T) = V_{i,1573K} + \frac{dV_i}{dT}(T - 1573K)$$
(6)

and $V_{H_2O}(T)$ is the temperature-dependent molar volume of water modelled starting from the volume at ~1000 °C (Ochs III and Lange, 1999; Richet et al., 2000). The mole fractions of the MI's major oxides (X_i) were derived from chemical analyses measured with a JEOL JXiHP200F field emission electron probe micro-analyser (EPMA) at the IMAP laboratories of the Université Grenoble-Alpes (France). The crystal wafers were embedded in epoxy resin, polished (down to 1 µm diamond grade), and carbon coated. We used an acceleration voltage of 15 kV, a probe current of 5 nA, and a defocused beam diameter of 15 µm to analyse the MIs. The analytes were measured with four WDS (wavelength dispersive spectroscopy) and one EDS (energy dispersive spectroscopy; Si only, with StHs6-80 glass as calibration material) spectrometers. Sodium was analysed first to avoid element dispersion and a ZAF correction was applied. The StHs6-80, Atho-G and KE12 reference materials (Jochum et al., 2006) were analysed three times each, after every 30-40 measurements to evaluate accuracy. Each MI was measured at least three times for consistency. Water concentrations were roughly estimated using the by-difference method (e.g., Devine et al., 1995).

1523 To constrain the MI's temperature T, equivalent to the fictive temperature of the pre-eruption 1524 silicate liquid, we applied the alkali feldspar-liquid geothermometer (Eq. (24b) in Putirka, 1525 2008). This thermometer describes the chemical equilibrium between magmatic melts and 1526 alkali feldspars as a function of temperature. Alkali feldspar crystals were handpicked from the 1527 0.5-2 mm grain-size fraction of crushed pumices and then embedded in epoxy resin. Pumiceous 1528 glass shards were obtained from the 125-250 µm grain-size fraction of manually crushed 1529 pumice clasts. The crushed material was rinsed and wet sieved at 80 µm in demineralised water 1530 to remove accretionary dust or clays and embedded in epoxy resin. Grain mounts of glass shards 1531 and alkali feldspar crystals were grounded and polished with diamond paste down to a grade of 1 µm. Alkali feldspar composition was analysed by EPMA, using the same instrument and 1532 1533 analytical conditions as those used to analyse the haüyne-hosted MIs, and with Cr-Augite, 1534 Ortho-Bl3 and Ortho-Bl2 as secondary standards. For the glass shards, a 2 nA probe current 1535 and a 5 µm beam diameter were used. We analysed glass from each layer at least eight times 1536 for reproducibility, while core-to-rim transects of 5 measurements were performed on the alkali 1537 feldspars. As the alkali feldspar-liquid geothermometer does not foresee an equilibrium 1538 constant to constrain the equilibrium between melt and alkali feldspars (Putirka, 2008), to assess 1539 equilibrium between feldspar rims and pumice groundmass, petrographic observations of thin 1540 sections from five stratigraphic subsamples (KF176B; KF176E; KF176H; KF176K; KF176N)

were conducted using optical petrographic microscopy and scanning electron microscopy (SEM). SEM imaging was performed in backscattered electron mode, at 15 kV using a Hirox SH5500P equipped with a Bruker EDS Quantax detector at the Laboratoire G-Time of the Université libre de Bruxelles (Belgium). No disequilibrium features were identified. However, before applying feldspar-based thermometry, textures of feldspars analysed with EPMA were further assessed via SEM. Haüyne density was assumed to be 2.4 g·cm⁻³ (Taylor, 1967).

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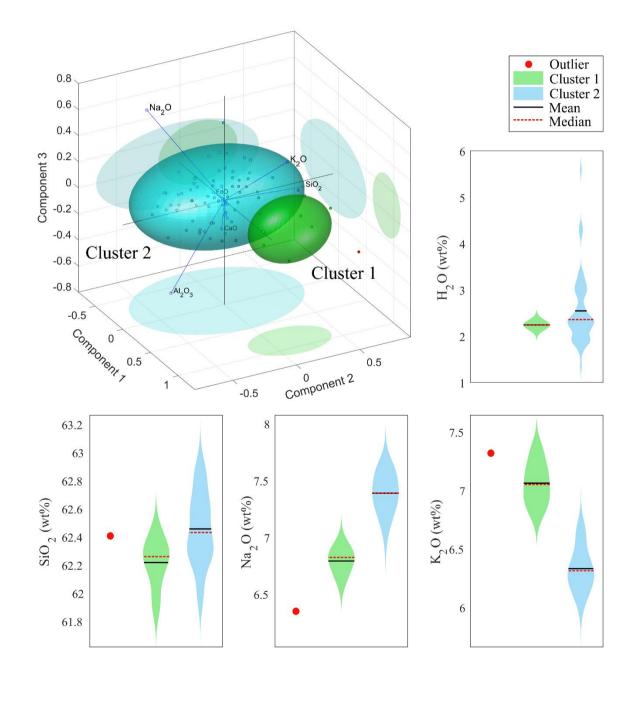
f. Principal component analysis

A principal component analysis of MI major element concentrations allowed to identify the most relevant oxides affecting the chemical variability of the melt, in an attempt to identify chemically differentiated magmas co-occurring in the magmatic reservoir. The three most representative principal components were used to identify chemically distinct clusters of MIs through hierarchical clustering.

1553 Results:

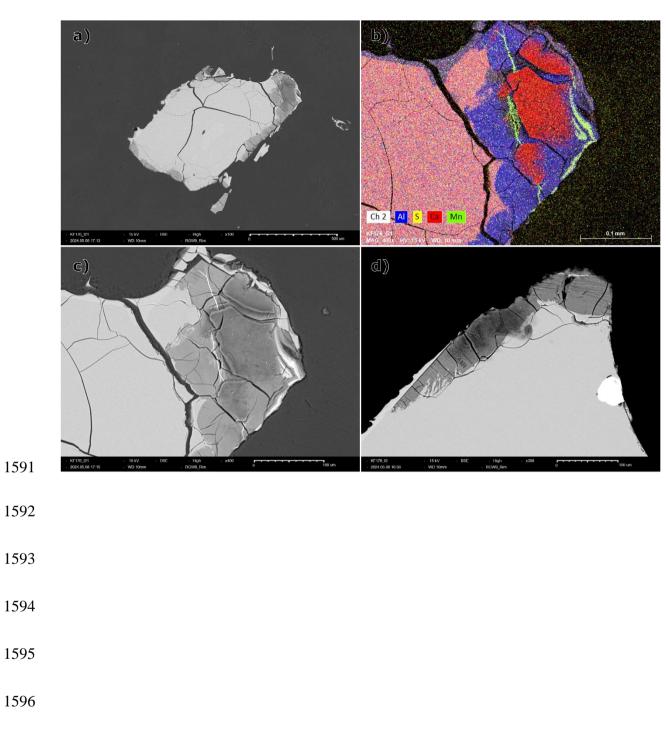
1554 The principal component analysis of MI glass composition suggests that the chemical variance 1555 is dominated by K₂O, Na₂O and SiO₂, which constitute the majority of the first three principal 1556 components and represent a total variance of ~26%, ~25% and ~23% respectively. The attempt 1557 to cluster the MIs according to their stratigraphic horizon was unsuccessful. Instead, the first three principal components, which represent ~96% of the variance, were used to group the data 1558 1559 into two major chemical clusters plus an outlier sample (Fig. 2SI) through a hierarchical 1560 clustering. While the first cluster includes only six MIs, the second cluster includes most of the MI dataset and represents a slightly more silicic magma, enriched in Na₂O and depleted in K₂O 1561 (Fig. 2SI). The incorporation of MIs in one or the other cluster is not related to the distribution 1562 1563 within crystals or to whether it hosts a shrinkage bubble. However, all the MIs belonging to the 1564 more evolved first cluster are found in the topmost stratigraphic horizons.

Supplementary Figure 2: Principal component analysis of major element concentrations: for each melt inclusion (including embayments) the values of the three major principal components obtained are shown together with the variance contribution of each element (blue lines). The samples were divided into two major clusters (green and cyan ellipses) plus an outlier (red dot) through hierarchical clustering. Statistical distribution of SiO2, Na2O, K2O and H2O concentrations for each cluster are shown in violin plots.



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1582	g.	Supplementary Figure 3SI: Backscattered electron images of haüyne crystals showing
1583	-	zeolitic alterations tens to hundreds µm thick around crystal surfaces; b) The energy
1584		dispersive X-ray spectroscopy (EDS) mapping of relative chemical concentrations
1585		shows in false colours the enrichment in Al and Ca in the zeolite and the depletion of
1586		sulfur. Mn veins are also present. Images were collected with an accelerating voltage of
1587		15 kV using a Hirox SH5500P scanning electronic microscope (SEM) equipped with a
1588		Bruker EDS Quantax detector at the Laboratoire G-Time of the Université libre de
1589		Bruxelles (Belgium).



- a. Supplementary Figure 4: a) Feldspar ternary diagram reporting core and rim analyses of alkali feldspars. Phyton3 tool Thermobar (v.0.07; Wieser et al., 2022) was used for data plotting; b) SEM backscattered electron image of an alkali feldspar crystal showing no evident textural zonation. The *red line* indicates the core-to-rim transect of analyses. (*Wieser, P.E., Petrelli, M., Lubbers, J., Wieser, E., Özaydın, S., Kent, A.J.R., Till, C.B.,*
- 1607 2022. Thermobar: An open-source Python3 tool for thermobarometry and hygrometry.
- 1608 Volcanica 5, 349–384. https://doi.org/10.30909/vol.05.02.349384).

