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1 **Highly CO₂-supersaturated melts in the Pannonian lithospheric mantle – A**
2 **transient carbon reservoir?**

3

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19

20 Abstract

21

22 The carbon budget of the continental lithospheric mantle is poorly known, although it is
23 believed to be substantially fed by subduction zones and a source of the widespread non
24 volcanic CO₂ degassing occurring in active tectonic intracontinental settings. We propose new
25 constraints on the CO₂ budget of the subcontinental lithospheric mantle below the Pannonian

26 Basin (Central Europe) through present study of a suite of xenoliths from the Bakony-Balaton
27 Highland Volcanic Field. Trails of secondary fluid inclusions, silicate melt inclusions,
28 network of melt veins and melt pockets with large and abundant vesicles, are numerous
29 evidences of mantle metasomatism that affected the lithosphere beneath region. A quantitative
30 estimate of the CO₂ budget of the mantle below the Pannonian Basin was constrained by an
31 innovative approach applied to mantle xenoliths using (1) synchrotron X-ray
32 microtomography, (2) NanoSIMS, Raman spectroscopy and microthermometry, and (3)
33 thermodynamic models (Rhyolite-MELTS). The reconstructed 3D volumes obtained by
34 synchrotron X-ray microtomography allow quantifying the proportions of all petrographic
35 phases in the samples and to visualize their textural relationships. The concentrations of CO₂
36 and H₂O in glass veins and pockets range between 0.27 ± 0.01 (1 σ) and 0.96 ± 0.02 (1 σ), and
37 0.54 ± 0.26 (1 σ) and 4.25 ± 0.27 (1 σ) wt. %, respectively, and have higher CO₂ contents than
38 typical arc magmas (0-0.25 wt. % CO₂ and 1.9-6.3 wt. % H₂O). Trapping vesicle pressures
39 were determined based on silicate melt saturation pressures between 0.69 to 1.78 GPa, and are
40 generally in excess compared to fluid inclusions trapping pressures determined by Raman
41 spectroscopy and microthermometry (0.1-1.1 GPa). The CO₂/silicate melt mass ratios in the
42 metasomatic agent that percolated the lithospheric mantle below the Pannonian Basin are
43 estimated between 9.0 and 25.4 wt. %, indicating that silicate melts were CO₂ supersaturated
44 in their source region. We suggest that slab derived melts initially containing up to 25 wt. %
45 of CO₂ migrated congruently into the lithospheric mantle, and were subsequently fractionated
46 by CO₂ segregation in secondary fluid inclusions by fracturing and creeping of the peridotite
47 mineral matrix. A first order estimation of the bulk minimal CO₂ concentration in the
48 lithospheric mantle below the Pannonian Basin is proposed at 1935 ppm. This transient
49 carbon reservoir is believed to be degassed through the Pannonian Basin due to volcanism and

50 tectonic events, likely drained by the Mid-Hungarian Zone, a regional shear zone of
51 lithospheric scale.

52

53 Keywords: X-ray microtomography; Mantle-xenoliths; CO₂-budget; Lithospheric-mantle

54

55 1 Introduction

56

57 The mantle is thought to be largest carbon reservoir of the Earth (Sleep et al., 2001; Shcheka
58 et al., 2006; Dasgupta and Hirschmann, 2010; Dasgupta, 2013), containing more than 90% of
59 carbon of the Earth (Poinar, 2012). Many geological processes depend on and attest for the
60 existence of deep carbon and its exchange with other major terrestrial reservoirs, such as CO₂
61 degassing at mid-ocean ridges, CO₂-rich magmatism rooted in the mantle (kimberlites,
62 carbonatites), and diamond or carbonate-bearing xenoliths originating from the deep mantle
63 (Hammouda 2003 and references therein). In order to balance the continuous CO₂ degassing
64 at volcanic centers, carbon must have been recycled into the mantle over geological time
65 scales, most likely at convergent margins where carbonate-altered, sediment-bearing oceanic
66 crust returns into the mantle (Zhang & Zindler 1993). Carbon dioxide is known to be a
67 dominant volatile species in magmatic fluids and gases in subduction zone environments
68 (Anderson, 1975; Fyfe et al., 1978) and its mantle origin has clearly been shown by ³He/⁴He
69 and δ¹³C isotopic ratios of fumaroles and hot spring gases (e.g. Sano & Marty, 1995). There
70 have been several recent reviews of the carbon cycle in subduction environments (Jarrard,
71 2003; Wallace, 2005; Gorman et al., 2006; Dasgupta and Hirschmann, 2010; Johnston et al.,
72 2011; Dasgupta, 2013; Burton et al., 2013), which estimate that about half of the subducted
73 carbon is removed from the subducting plate beneath fore-arcs and arcs and returns surface of
74 Earth [40% in Gorman et al. (2006); 20–80% in Dasgupta & Hirschmann (2010); and 18–70%

75 in [Johnston et al. \(2011\)](#)]. A large part of the subducted CO₂ is therefore thought to be stored
76 in the mantle ([Kelemen and Manning, 2015](#)). Kelemen and Manning (2015) recently argued
77 that the carbon input from subduction zones into the overlying plate is larger than output from
78 arc volcanoes plus diffuse venting, and that substantial quantities of carbon are stored in the
79 mantle lithosphere and crust. This recent study suggests that diffuse escape of CO₂ towards
80 the surface and, more importantly, transient storage of CO₂ in the back-arc lithospheric mantle
81 may, when added to the more commonly described fluxes of arc magma and MOR degassing,
82 fully balance the subducted carbon ([Kelemen and Manning, 2015](#)). Therefore, both the diffuse
83 degassing flux in back-arc regions and the lithosphere mantle carbon reservoir need to be
84 further quantified since they have considerable implications for the global carbon cycle.
85 This study aims at constraining the magmatic CO₂ budget of the subcontinental lithospheric
86 mantle below the Pannonian Basin (PB, Central Europe) through the study of a suite of
87 petrographically and geochemically well-studied peridotite xenoliths ([Creon et al., this](#)
88 [volume](#)) from the Balaton-Bakony Highland Volcanic Field (BBHVF, [Szabó et al. 2010](#)). The
89 PB is known for its substantial mantle derived CO₂ accumulations ([Ballentine et al., 1991](#);
90 [Sherwood Lollar et al., 1997](#)), complex geodynamic setting ([Horváth et al., 2015](#)) and
91 volcanic activity ([Harangi and Lenkey, 2007](#)) that have developed within a relatively short
92 time scale of 16 My ([Kovács and Szabó, 2008](#)). The PB represents an ideal setting for
93 studying the fluxing of mantle CO₂ through the lithosphere. This study develops an innovative
94 multi-technique approach in order to investigate the abundance and significance of CO₂ fluid
95 inclusions and vesicles within mantle xenoliths by coupling: (1) three-dimensional
96 synchrotron X-ray microtomography, (2) nanoSIMS, Raman spectroscopic and
97 microthermometric characterization and (3) thermodynamic modeling of the magmatic
98 evolution.
99

100 2 Geological background

101

102 The Pannonian Basin (PB) is located in the Carpathian–Pannonian region of Central Europe.

103 The PB is a typical inter-arc basin ([Embey-Isztin et al, 2001](#)), yet it shows a number of

104 features that are characteristic of rift zones, such as high heat flow, recent alkali volcanism,

105 thin crust and lithosphere, and upwelling asthenosphere ([Embey-Isztin et al., 1990](#); [Praus et](#)

106 [al., 1990](#); [Spakman, 1990](#); [Falus et al., 2007](#)). The Pannonian Basin experienced a complex

107 geodynamic evolution during the Miocene to Pleistocene characterized by the succession of

108 extension-compression-extension events ([Bali et al., 2008](#)). The major driving forces that led

109 to the formation of the Pannonian Basin (~20 Ma) were the continuous subduction and roll-

110 back on its eastern boundaries and the synchronous eastward extrusion of ALCAPA blocks

111 ([Kazmer and Kovacs, 1985](#)) from the Alpine compressional belt ([Horvath, 1993](#); [Fodor et al.,](#)

112 [1999](#)).

113 The Bakony–Balaton Highland Volcanic Field (BBHVF), on the northern shore of Lake

114 Balaton, is situated near the center of the Carpathian-Pannonian region, in the western

115 Pannonian Basin (western Hungary). The volcanic centers of the BBHVF were active

116 between 7.96 Ma and 2.61 Ma ([Balogh and Pécskay, 2001](#); [Balogh and Németh, 2005](#);

117 [Wijbrans et al., 2007](#); [Hidas et al., 2010](#)) and produced mostly alkali basaltic volcanic rocks

118 ([Szabó et al., 1992](#); [Embey-isztin et al., 1993](#)).

119 Ultramafic xenoliths can be found in basanitic lava flows and pyroclastic volcanic products at

120 six locations (Tihany, Bondoró Hill, Füzes-tó, Szentbékállá, Mindszentkállá, Szigliget).

121 Most of the xenoliths are spinel lherzolites, but harzburgite, clinopyroxenite, orthopyroxenite,

122 wehrlite, websterite and, sometimes, composite xenoliths also occur (See [Creon et al. \(this](#)

123 [volume\)](#) for more information).

124

125 3 Petrography of the mantle xenoliths

126

127 More than 150 ultramafic xenoliths from the BBHVF were collected at the Szentbékállá,
128 Szigliget, Füzes-tó and Mindszentkálá localities (see [Creon et al. \(this volume\)](#) for sampling
129 locations). After microscopic observations, four ultramafic xenoliths from Szentbékállá, one
130 from Szigliget and two from Füzes-tó were selected for further study. Samples selected are 7
131 to 12 cm long, rounded spinel peridotites and range from lherzolite to harzburgite
132 compositions. A detailed petrographic description of the samples is given in the companion
133 paper by [Creon et al. \(this volume\)](#), and only a short description will be given here. Further
134 petrographic observations obtained by X-ray tomography will be presented in the *Results*
135 section. Peridotite samples show equigranular to mosaic or protogranular microstructures with
136 minor poikilitic features (based on the classification of [Mercier & Nicolas \(1974\)](#)). Samples
137 are composed of olivine (Ol), orthopyroxene (Opx), clinopyroxene (Cpx) and spinels (Sp).
138 Amphibole (Amp) also occurs in some of the xenoliths (Szigliget and Füzes-tó).
139 Microscopic observations show that the majority of these xenoliths contain veins, primary
140 silicate melt inclusions or melt pockets. Melt pocket sizes are variable (from few hundreds
141 microns to few millimeters in diameter) and are usually connected to each other by thin
142 silicate melt veins. The melt pockets are composed of secondary formed clinopyroxene,
143 olivine, spinel, and interstitial glass with vesicles. Melt pocket shapes can be irregular, but
144 some are euhedral pseudomorphs of protominerals (amphibole). Numerous trails of secondary
145 fluid inclusions are observed in Opx, Cpx and Ol. Fluid inclusions display equilibrium and
146 non-equilibrium features depending on the samples ([Creon et al., this volume](#)).

147

148 3.1 Analytical methods

149

150 3.1.1 Synchrotron X-ray microtomography

151

152 The eleven samples analyzed consist of tiny cylinders 5 to 10 mm in length and 4 mm in
153 diameter (Fig. 1). The internal textures of the mantle xenoliths were characterized through
154 synchrotron radiation computed micro-tomography at the European Synchrotron Radiation
155 Facility (ESRF), Grenoble (France) on the ID19 beamline. ESRF offers access to very high
156 X-ray beam intensity. The detector used is a Fast Readout Low Noise FRELON Camera
157 developed at ESRF, with 2048×2048 pixels and 14 bits resolution ([Baruchel et al., 2000](#);
158 [Labiche et al., 2007](#)).

159 Monochromatic microtomography with 30 keV radiation was done on the 4 mm diameter
160 cylinders, using a multilayer monochromator. The phase contrast effect was obtained with a
161 short propagation distance; the sample-detector distance was of 10 mm for a pixel size equal
162 to 1.27 μm . With this setting (Fig. 1), scanned regions were 4 mm by 2 mm in size when
163 using a half-acquisition mode. Typically, the 3D images obtained are composed of
164 3639×3639×2048 voxels (elementary image volume) with 32-bit grey-scale resolution.
165 Acquisition time was of the order of 45 min for 5000 projections together with around 625
166 reference images (blanks). A primary correction of the ring artifacts was done directly after
167 analysis using specific filtering. Tomographic volume reconstructions (example given in Fig.
168 2) were performed using an algorithm with a single phase retrieval distance in order to
169 optimize the signal/noise ratio ([Paganin et al., 2002](#); [Sanchez et al., 2012](#)). Petrographic data
170 treatments were performed using ImageJ[®] and Avizo[®] softwares. Individual grey levels were
171 identified for each solid (mineral and glass), liquid and gas phase by defining their threshold
172 bracketing on bulk sample grey level histograms calculated using ImageJ[®] (Fig. 3). Then the
173 3-D volumes of each petrographic phase were defined with the Avizo[®] software. Each phase
174 was individualized by segmentation of the volumes corresponding to their density range.

175 Uncertainties associated with this segmentation were qualified by manually adjusting
176 minimum and maximum thresholds for each phase (Fig. 4). Once the thresholds well defined,
177 the volumes of the phases were segmented and quantified.
178 The densities of the petrographic phases were calculated from their grey levels using model
179 absorbance compiled with a NIST utility (<http://www.nist.gov/pml/data/xcom/>).

180

181 3.1.2 Nano-SIMS microanalyses of C and H

182

183 Carbon and water contents of glass were determined using the NanoSIMS 50 installed at the
184 MNHN (Paris) on gold-coated (10 nm thick) samples. Analyses were performed with a 16
185 keV Cs⁺ primary ion beam rastering the surface of the sample. A primary beam intensity of
186 40 pA was used to perform analyses with a D1-3 (200 μm) aperture diaphragm. Secondary
187 ions of ¹²C, ¹⁶O¹H⁻, ²⁸Si⁻, ²⁷Al¹⁶O⁻ and ⁵⁶Fe were collected using electron multipliers over 3 x
188 3 μm² square using the beam blanking mode (1.5 x 1.5 μm²) to reduce surface contamination
189 (Stephant et al., 2014). A presputtering on a 3 × 3 μm² surface area during 300 s was
190 performed to remove coating and initial surface contamination and to reach the sputtering
191 steady-state. Calibration curves were obtained by measuring tholeiitic glass standards (Fig. 5)
192 provided by the Centre de Recherche en Pétrographie et Géochimie (CRPG), Nancy, France
193 [standard description available in Shishkina et al. (2010)]. Data corrections, using the
194 aforementioned calibration, and error calculations were performed using the R program
195 (Dunn, 2007). Errors combine counting statistics and uncertainty of the calibration curve.
196 However, the errors reported in Table 1 are dominated by the uncertainty of the calibration
197 curve, which corresponds to prediction interval at 68% (1σ) (Bartoli et al., 2014). During the
198 session, the vacuum in the analysis chamber remained at 5×10⁻¹⁰ Torr.

199

200 3.1.3 Raman Spectroscopy and Microthermometry

201

202 3.1.3.1 Raman Spectroscopy

203

204 The fluid inclusions (trapped by order of abundance in Opx, Cpx and Ol) were analyzed (n=
205 99) by Raman spectroscopy at **Faculty of Science Research and Instrument Core Facility**
206 **(ELTE FS-RICF, Hungary)**. A Jobin Yvon confocal Labram Raman instrument was used with
207 a frequency-doubled Nd-YAG laser with an excitation wavelength of 532 nm. Laser energy
208 was 50 mW and 20 mW at the laser source and at the sample surface, respectively. 50× and
209 100× microscope objectives were used to focus the laser onto the sample and to collect the
210 Raman signal. Analytical settings included a 200–500 μm confocal hole, 200 μm spectral slit,
211 600 or 1800 grooves/mm spectrograph gratings, 2–10 accumulations and 2–150 s acquisition
212 time (all depending on the maximum intensity). The spectral resolution of the measurements
213 varied with wave number but was $\leq 2 \text{ cm}^{-1}$ using the grating with 1800 grooves/mm.
214 Following the method of [Berkesi et al. \(2009\)](#), spectra were collected at room and high
215 (150°C) temperatures using a Linkam THMS600 heating/cooling stage. To characterize the
216 bands in the Raman spectra and to identify the phases present, data were compared to the
217 online Raman database of the French Society of Mineralogy and Crystallography
218 (<http://www.obs.univ-bpclermont.fr/sfmc/ramandb2/index.html>) and to that of [Bonelli &](#)
219 [Frezzotti \(2003\)](#), (<http://www.dst.unisi.it/geofluids/raman>). Data were processed using
220 LabSpec v5.25.15 software designed for Jobin-Yvon Horiba LabRam instruments. Minimum
221 and maximum values of CO₂ densities in inclusions were calculated from the two Raman
222 bands of CO₂ due to Fermi resonance ([Fall et al., 2011](#)) (Fig. 6).

223

224 3.1.3.2 Microthermometry

225
226 A Linkam MDS 600 heating-freezing stage (IFP Energies nouvelles, Rueil-Malmaison,
227 France) was used to measure the phase transitions in CO₂-rich fluid inclusions, which
228 homogenized in the liquid phase in our samples. After freezing the fluid at <-75 °C, the
229 inclusions were slowly heated to the triple point temperature check (-56.6 °C for pure CO₂)
230 with a heating rate of 0.5-2 °C.min⁻¹. Final homogenization occurred to the liquid phase
231 (T_hL). The CO₂ density and pCO₂ were calculated following [Span and Wagner, 1996](#) and
232 [Sterner and Pitzer, 1994](#), respectively, for a magma temperature of 1150 ± 15 °C (estimation
233 from [Creon et al. \(this volume\)](#) from the melting conditions.

234

235 3.2 Results

236

237 3.2.1 Synchrotron X-ray tomography

238

239 3.2.1.1 Petrographic observations

240

241 The density resolution of synchrotron X-ray tomography allows clear discrimination among
242 the six major phases of the peridotitic xenoliths: olivine, orthopyroxene, clinopyroxene,
243 spinel, glass and fluid. Figure 3 shows the density spectra of the SZB51 xenolith together with
244 the modal composition corresponding to the pixel count of each phase. In SZB51 (Fig. 3),
245 olivine is the most abundant phase and fluid is the least abundant.

246 Regarding the 3D images obtained by synchrotron X-ray microtomography, we confirm that
247 the rock textures are protogranular, tabular or equigranular-mosaic (according to the
248 classification of [Mercier & Nicolas, 1974](#)) as previously reported by optical microscopy on
249 thin sections of the same samples ([Creon et al., this volume](#)).

250 The glass phase may be located at grain boundaries as veins or melt pockets (Fig. 6). Fluids
251 are observed as trails of secondary fluid inclusions (FI) inside minerals (Ol, Opx, Cpx) or as
252 vesicles inside the glass (Fig. 6e). Glass containing vesicles forms a complex interconnected
253 network along grain boundaries as shown in figure 7. Trails of FI run across pre-existing melt
254 veins and melt veins cross pre-existing trails of FI. Moreover, trails of secondary FI and the
255 glass (+ vesicles) network intersect each other through multiple grains. Small (~100-200 μm ,
256 Fig. 8) newly formed Ol, Cpx and Sp are observed in veins (in low abundance) and in melt
257 pockets (in high abundance).

258 The glass (+ vesicle) network percolates through the whole volume and is in direct contact
259 with the exterior, indicating that vesicles probably lost all the gas they previously contained
260 during their exposure to surface conditions or during sample preparation.

261

262 3.2.1.2 Phase volume quantification

263

264 The volumes of minerals, glass and fluid phases have been quantified within the bulk 3-D
265 volumes. Fluid volumes have been calculated on each sample by direct density threshold
266 segmentation; the obtained volumes are between 0.7 and 4.7 vol% (Fig. 9a & Table 1).

267 Vesicles in melt systematically make the majority of the fluid volume, FIs representing at
268 most 5 vol. %. The FIs can be divided according to which mineral they are hosted in. In
269 sample SZB51, for example, FI represent a volume proportion in opx of 0.6%, in cpx of 0.3%,
270 and in ol of 0.18%. These proportions, weighted by the modal composition of SZB51, imply
271 that opx, ol and cpx trapped respectively 57.8%, 39.0% and 3.3% of the total FI volume (Fig.
272 9b).

273 Glass volume determination was performed in two steps. The first step resulted from the glass
274 volume calculation by direct threshold segmentation (as for fluid volumes) and gave volumes

275 between 0 and 3.8 vol% (best estimate). The second step was performed for some samples
276 (SZB50, SZG23, FT12 and FT01P) that showed evidence of important secondary
277 crystallization of the melt (quenched as glass) such as newly formed Ol, Cpx and Sp (Fig. 8).
278 In order to correct the glass volumes from this crystallization of new phases, and to consider
279 the pre-crystallized bulk volume of melt pockets, we extracted the X-ray images of silicate
280 melt pockets on five slices of each sample and calculated their (melt+newly formed
281 minerals)/melt volume ratios (Fig. 8). The obtained ratios were used to correct the 3D glass
282 volumes from crystallization of new phases. The corrected glass volumes in these samples
283 range between 0 and 23.4 vol. % of the total volume (Fig. 9a, Table 1).
284 The corrections for crystal fractionation were performed only on silicate melt pockets since
285 veins only show very minor crystallization that should have negligible effects on the melt
286 volumes. After correction for crystallization of newly formed phases in the melt pockets, the
287 glass and the fluid volumes display a linear correlation (Fig. 11).

288

289 3.2.1.3 Density determination

290

291 The grey level value of each pixel was quantitatively assigned to a density. Densities
292 determined for Ol (3.37 to 3.41 g.cm⁻³), Opx (3.29 to 3.32 g.cm⁻³) and Cpx (3.42 to 3.43 g.cm⁻³)
293 correspond to those described in the literature using the Mg# of the minerals of these
294 samples [Creon et al. \(this volume\)](#) (Fig. 11). Glass densities were then determined using the
295 average density of glass-assigned pixels in each sample. The densities are distributed between
296 2.45 and 2.74 g.cm⁻³ (Table 1, Figure 11) and are in agreement with andesite to basalt glass
297 densities available in the literature ([Richet et al. 2000 and references therein](#)). Densities
298 calculated with the method of [Dixon et al. \(1995\)](#) (2.41 to 2.59 g.cm⁻³) are also close to those

299 determined in this study (Fig. 11). Glass densities determined by X-ray microtomography are
300 therefore considered to be accurate and will be used in the following steps of this study.
301 The X-ray microtomography density determinations for the fluid vesicles and inclusions were
302 not conclusive due to the poor X-ray absorbance resolution in the lower density range
303 (typically 0 to 1.5 g.cm⁻³), together with the partial-volume-effects (density artifact due to the
304 presence of a phase boundary within a single voxel) on the smaller segmented volumes, which
305 commonly affect the fluid/solid phase boundaries.

306

307 3.2.2 Fluid compositions and densities

308

309 Fluid compositions and densities were measured on FI by Raman spectroscopy and
310 microthermometry. As noted above, fluids originally trapped in vesicles in the melt veins and
311 melt pockets were lost from the sample and could not be analyzed; hence the FI were used as
312 representatives of all the fluids of the samples, even though the vesicles dominated the
313 volume fraction assigned to fluid. Following the method developed in [Berkesi et al. \(2009\)](#),
314 we used Raman spectroscopy at room and elevated temperatures (150 °C) in order to
315 discriminate the potential trace volatiles inside fluid inclusions. With this method, only CO₂
316 was recognized in the fluid inclusions. Independently, the phase transition observed at ~56.6
317 ± 1 °C by microthermometry measurements is in agreement with pure CO₂ inclusions.

318 The comparison of densities obtained with both Raman spectroscopy and microthermometry
319 methods shows a linear correlation, with a correlation factor of 0.89 (R²). This suggests that
320 the fluid density datasets obtained by the two methods are internally consistent (Fig. 14a). The
321 densities of fluids obtained in SZB, SZG and FT are respectively between 0.30 and 1.11 g.cm⁻³
322 ³, 0.48 and 0.82 g.cm⁻³ and 0.30 and 1.06 g.cm⁻³ (Fig.13b).

323

324 3.2.3 Volatiles dissolved in glass

325

326 NanoSIMS analyses have been performed on glass from veins and melt pockets to determine
327 their CO₂ and H₂O contents. H₂O contents vary from 0.54 ± 0.26 (1 σ) to 4.25 ± 0.27 (1 σ) wt.
328 % in glass from veins and from 0.55 ± 0.26 (1 σ) to 2.76 ± 0.28 (1 σ) wt. % in glass from melt
329 pockets. CO₂ contents vary between 0.27 ± 0.01 (1 σ) and 0.48 ± 0.02 (1 σ) wt. % in glass from
330 veins and from 0.38 ± 0.01 (1 σ) to 0.96 ± 0.02 (1 σ) wt. % in glass from melt pockets (Table
331 1).

332

333 4 Discussion

334

335 4.1 Magma CO₂-supersaturation conditions

336

337 Several petrographic lines of evidence point to CO₂ supersaturation in the melt trapped within
338 the studied xenoliths at lithospheric depths. Such evidence includes 1) the occurrence of CO₂
339 FI with densities above 1.0 g.cm^{-3} (Table 1) corresponding to pressures higher than the
340 present Moho depth (25-30 km, [Kovacs et al., 2012](#)); 2) the coexistence within veins of
341 vesicles and glass with high dissolved CO₂ and H₂O concentrations in the glass of up to 0.96
342 and 4.25 wt. %, respectively (Table 1). In order to assess the conditions of magma
343 supersaturation, CO₂ saturation isotherms were calculated for the different melt compositions
344 measured in the present samples over a range of assumed temperatures using the Rhyolite-
345 MELTS software (version 1.2.0, [Gualda et al., 2012](#); [Ghiorso and Gualda, 2015](#)). This model
346 is suitable for the calculation of thermodynamic equilibrium of mafic to felsic systems with
347 H₂O and CO₂ over the pressure range 0-3 GPa, corresponding to the conditions of the
348 lithospheric mantle below the BBHVF ([Kovacs et al., 2012](#)).

349 Magma CO₂-saturation shows little temperature dependence between 1100 and 1300 °C and
350 for pressure below 0.5 GPa, as already shown by [Shishkina et al. \(2010\)](#) and [Botcharnikov et](#)
351 [al. \(2006\)](#). The effect of temperature only tends to increase the solubility of CO₂ in the high
352 pressure range of 0.5-3.0 GPa, as also mentioned by [Papale et al. \(2006\)](#). Based on CO₂-
353 saturation isotherms, vesicle-melt equilibrium pressures were calculated from the CO₂
354 contents of melts determined by NanoSIMS (Table 1, Figure 13). These saturation pressures
355 at 1100°C (corresponding to approximated mineral equilibrium temperatures determined in
356 [Creon et al. \(this volume\)](#)) vary between 0.69 and 1.78 GPa and correspond to depths of 26 to
357 62 km (using the geotherm of [Kovacs et al., 2012](#)). Such estimations likely correspond to
358 vesicle-melt equilibrium near the glass transition during rapid cooling of the percolating melt
359 within the melt veins and pockets. Glass transition temperatures (T_g) were estimated between
360 670 to 854°C by [Giordano et al. \(2008\)](#) at asthenospheric pressures, and between 520 to
361 930°C by [Morizet et al. \(2007\)](#) at pressures between 1.0 and 2.5 GPa (phonolite melts). CO₂
362 appears to have almost no effect on the glass transition of silicate melts unlike H₂O, which
363 clearly reduces glass transition temperatures ([Morizet et al. 2007](#)). Furthermore, pressure has
364 a very limited influence on glass transition temperatures as demonstrated by [Bagdassarov et](#)
365 [al. \(2004\)](#). These glass transition temperatures would be intersected along the geotherm of
366 [Kovacs et al. \(2012\)](#) at pressures near ~0.5 GPa i.e. at crustal levels (~20 km depth). Because
367 melt pockets were shown to have been quenched under lithospheric mantle pressures while
368 coexisting with fluid vesicles ([Creon et al., this volume](#)), the crustal saturation pressures
369 recorded for most CO₂ depleted melts are likely to reflect the fast ascent of xenoliths after
370 their extended residence time at lithospheric mantle depths.

371 Melt saturation pressures of 0.69 to 1.78 GPa were converted to CO₂ density using the
372 equations of state of [Span & Wagner \(1996\)](#) and [Sternner & Pitzer \(1994\)](#). The densities
373 obtained vary between 0.93 and 1.33 g.cm⁻³ and are consistent with the density range

374 determined by Raman spectroscopy and microthermometry in the denser FIs (Table 1). This
375 similarity attests to a coeval formation of FI and vesicles at lithospheric mantle conditions.
376 However, no direct correlation is observed between maximum fluid inclusion and vesicle
377 densities, which likely points to the unstable behavior of highly over-pressurized FIs
378 (Roedder, 1984). In most cases (samples SZB16, SZB50, SZG23, SZG44, FT12), fluid
379 inclusion densities are lower than the associated calculated vesicle densities; which can occur
380 due to the decrepitation of the FIs during xenolith ascent (Bodnar, 2003). In the rest of the
381 xenoliths (SZB51, SZB52, FT01P) the densities of the FI are larger than those of the
382 associated vesicles, which reflects FI trapping in equilibrium with vesicles at pre-ascent
383 lithospheric mantle pressures.

384 In figure 14, the CO₂ contents dissolved in glass measured by NanoSIMS (0.27-0.96 wt. %)
385 are compared with the data compilation of Wallace (2005). The values measured in this study
386 are in excess of those reported so far for arc magmas and are within the E-MORB range.
387 Moreover, the CO₂ and H₂O contents measured are larger than those described in the
388 literature in silicate melt pockets of the BBHVF (Bali et al., 2002). Hidas et al. (2010) used
389 silicate melt inclusions (SMI) from Szigliget mantle xenoliths to estimate the volatile content
390 of trapped metasomatic melt. Their calculated melt CO₂ concentrations of ~1 wt. % are in
391 agreement with the maximum CO₂ content measured in glass for this study in sample SZG23
392 from the same xenolith locality (0.8-1.2 wt. % CO₂; Fig. 14). It is thus likely that the SMI
393 analyzed by Hidas et al. (2010) correspond to the trapping of a volatile-rich melt during
394 overgrowth of clinopyroxenes in the mantle lithosphere similar to the most CO₂-rich
395 percolating melts analyzed in this study.

396

397 4.2 CO₂ contents of melts in the subcontinental lithosphere beneath the BBHVF

398

399 The 3-D images clearly reveal interconnections between trails of secondary fluid inclusions,
400 the glass in veins and in melt pockets and the network of vesicles. This is contrary to
401 observations made in similar samples from the same localities by [Demény et al. \(2010\)](#), who
402 described trails of FI in thin sections that did not extend to grain boundaries. The synchrotron
403 X-ray microtomography used in this study allows 3-D reconstructions that show indisputable
404 intersecting relationships of FI trails with grain boundaries (Figure 7). These 3-D observations
405 are clear petrographic evidence for a common fluid being present in the FI and in the
406 metasomatic melt that percolated these mantle xenoliths. As noted above, fluid in vesicles are
407 not preserved anymore and are, therefore, impossible to identify. Hence, we assumed that
408 vesicles have similar compositions as FIs and that they are dominated by CO₂. This
409 assumption is consistent with the high H₂O and CO₂ contents in the glass determined by
410 nanoSIMS.

411 The CO₂/glass mass ratio corresponding to the melt + vesicle system has been calculated
412 using equation (1):

413

$$414 \quad (1) \quad M(\text{CO}_2)/M(\text{Glass}) = V(\text{CO}_2)/V(\text{Glass}) * \rho(\text{CO}_2)/\rho(\text{Glass})$$

415

416 Where: $M(\text{CO}_2)$: mass of CO₂; $M(\text{Glass})$: mass of glass; $V(\text{CO}_2)$: volume of CO₂; $V(\text{Glass})$: volume
417 of glass; $\rho(\text{CO}_2)$: density of CO₂; $\rho(\text{Glass})$: density of glass

418

419 $V(\text{CO}_2)$, $V(\text{glass})$ and $\rho(\text{glass})$ were obtained by synchrotron X-ray microtomography and
420 $\rho(\text{CO}_2)$ was derived from the NanoSIMS analyses and thermodynamic modeling (Rhyolite-
421 MELTS, [Gualda et al. 2012](#); [Ghiorso & Gualda 2015](#)) as described in the previous section
422 (Table 1).

423 The calculated CO₂/melt mass ratios range from 9.0 to 25.4 wt. % (Table 1). These ratios,
424 which can be considered as the CO₂ mass fraction of the entire melt system, would

425 correspond to a single phase system at pressures of 4 to 9 GPa for basaltic melts following the
426 study of [Guillot & Sator \(2011\)](#). The present lithosphere-asthenosphere boundary below the
427 Pannonian Basin is estimated at depths between 60 and 100 km ([Tašárová et al., 2009](#)). The
428 melts characterized here were therefore likely to be supersaturated in the asthenosphere before
429 reaching lithospheric depths. The range of CO₂/melt mass ratios calculated in this study
430 implies that CO₂ contents in melts were variable either due to magmatic processes or to
431 physical fractionation. Below we review the various processes that might affect the CO₂/melt
432 ratio in lithospheric melts.

433

434 4.2.1 Carbonate assimilation and fractionation in the lithospheric mantle

435

436 Carbonate minerals have been described in Pannonian Basin xenoliths from the BBHVF.
437 These carbonates are mainly calcitic droplets ([Demény et al., 2004](#)), rounded ([Bali et al. 2002;](#)
438 [2008](#)) or anhedral ([Chalot-Prat and Arnold, 1999](#)) crystals in silicate veins and/or melt
439 pockets of mantle xenoliths. In the samples of this study, carbonate are also observed as veins
440 ([Creon et al., this volume](#)). [Bali et al. \(2002\)](#) and [Demény et al. \(2004\)](#) suggested the
441 infiltration of a mixed carbonate–silicate melt taking place in these mantle xenoliths, which
442 supports the evidence for CO₂-rich magma percolation beneath the BBHVF documented in
443 this study. Carbonates are, therefore, not the source of the melts but instead precipitate from a
444 CO₂-rich metasomatic agent during melt-rock reactions due to carbon saturation and vapor
445 immiscibility in the percolating silicate melts. Thus, the variability in CO₂ mass fractions in
446 melts calculated in this study is not believed to be caused by variable dissolution of
447 carbonated peridotites in the lithospheric mantle.
448 Reversely, the fractionation of carbonate minerals, or phase separation of carbonate and
449 silicate melts may have potentially played a role in fractionating CO₂ mass fractions in

450 magmas. Since no evidences of carbonate fractionation, nor carbonate melt segregation have
451 been witnessed in our sample set, although they were observed and discussed in comparable
452 samples of xenoliths of Transylvania (Chalot-Prat and Arnold, 1999; Bali et al., 2002;
453 Demény et al., 2004; Bali et al., 2008) we suggest that such processes were not the driving
454 mechanisms of the observed heterogeneity in the CO₂ mass fraction.

455

456 4.2.2. Fluid inclusions and physical fractionation

457

458 The large abundance of CO₂-rich FIs trapped in minerals along fractures and the absence of
459 comparably abundant silicate melt inclusions indicate that CO₂ was preferentially trapped in
460 grains in comparison with the silicate melt.

461 Schiano et al. (2006) suggested that the occurrence of fluid inclusion arrays, commonly
462 observed in mantle xenoliths, could be explained by transcrystalline melt migration, contrary
463 to the commonly accepted idea of simple trapping by "crack and seal" processes of individual
464 or multiple mineral grains of a free fluid phase percolating the deep mantle. However, no
465 textural features suggesting transcrystalline melt migration were observed in our samples.
466 Schiano et al. (2006) explained that only the most primitive droplets formed at fertile grain
467 corners (or edges) have time (if melting does not proceed rapidly to higher degrees) to enter
468 the crystals by thermally driven migration instead of escaping by buoyancy-driven flow along
469 grain edges. Melts analyzed in this study have evolved compositions (55.96-62.71 wt. %
470 SiO₂), consistent with the formation of an interconnected wet-edge network and rapid escape
471 to higher levels, where grain scale channels are relayed by fracture networks to form magma
472 bodies (Schiano et al., 2006). The absence of the high thermal gradient (0.03 to 0.06 K/μm)
473 needed for transcrystalline melt migration (Schiano et al., 2006) could also explain the
474 absence of evidence of transcrystalline melt migration in the mantle xenoliths studied here.

475 The microstructures observed in samples from this study suggest that peridotites are wetted by
476 silicate melt and not by CO₂ as already indicated by several authors (e.g. [Watson & Brenan,](#)
477 [1987](#)). This observation is consistent with the much smaller wetting angles expected for
478 silicate melt in dunite than for CO₂ in dunite ([Watson & Brenan, 1987](#)). Consequently, melt
479 inclusions only rarely trap melts with their primary CO₂ contents (e.g., [Saal et al. 2002](#)).
480 Instead, grain fractures due to mantle dynamic or to local overpressure may much more easily
481 trap volatiles as FI due to preferential viscosity-controlled injection within grain cracks
482 ([Watson and Brenan, 1987](#)). We suggest that cracking and healing processes operating in
483 response to fluid overpressure and creeping of the peridotite matrix induced the preferential
484 entrapment of CO₂ in minerals whereas the silicate melt from the same magma continued its
485 journey at grain boundaries through percolation. Variable amplitudes of this fractionation
486 process may then be expected depending on magma travelling distances, melt/rock ratios and
487 fluid overpressure conditions.

488

489 4.3 Trapped lithospheric melts in mantle xenoliths and arc magmas

490

491 [Creon et al. \(this volume\)](#) interpreted the metasomatic agent that infiltrated these xenoliths to
492 be a CO₂-rich adakite-like magma derived from slab melting that interacted with the
493 lithosphere mantle before being trapped in the peridotites. [Wallace \(2005\)](#) compiled the CO₂
494 and H₂O contents of silicate melt inclusions deduced from arc-magmas worldwide, which
495 range between 0 and 2100 ppm and between 1.9 and 6.3 wt. % respectively (Figure 15). The
496 CO₂ concentrations are much lower than the values determined in this study, which range
497 from 0.29 to 0.96 wt. % and are much higher than that of SMI in arc magmas worldwide. The
498 H₂O contents determined in this study range from 0.54 and 4.25 wt. % and are for the
499 majority, in the same range as those reported by [Wallace \(2005\)](#). We suggest that the CO₂

500 content of arc magmas sampled by SMIs may only represent a negligible fraction of the CO₂
501 associated with slab-derived melts. For instance, in xenoliths from this study as well as in
502 those from the literature ([Bali et al. \(2008\)](#) and [Hidas et al. \(2010\)](#)), only a very small fraction
503 of the CO₂-rich melt may be trapped as SMIs due to the pre-entrapment supersaturation in
504 CO₂ and the preferential accumulation of most of the CO₂ in vesicles and FIs (95-98 % in
505 samples from this study) rather than in the silicate melt. A large compositional gap seems to
506 exist in subduction-related environments between CO₂/magma mass fractions trapped as
507 melts in the lithospheric mantle and melts characterizing erupting subduction-related magmas.
508 This gap may be the result of physical and/or geochemical processes such as 1) intense
509 physical fractionation of CO₂ vesicles and silicate melt during magma ascent to the crust, 2)
510 important dilution of CO₂-rich parental magmas by assimilation of C-poor lithologies during
511 magma-rock interaction. The investigation of these processes is however beyond the scope of
512 this paper and will not be discussed further here.

513 Figure 15 illustrates the different potential sources of H₂O and CO₂ expected in a subduction
514 context where 1) E-MORB, 2) altered oceanic basalts and 3) subducted sediments ([Plank and](#)
515 [Langmuir, 1998](#); [Wallace, 2005](#)) represent rock-type end-members of slab melting. Partial
516 melting of the slab or of the metasomatized mantle wedge would generate melts enriched in
517 incompatible trace elements and with high CO₂ and H₂O contents like those calculated in this
518 study (0.55-4.25 wt. % H₂O and 9.0-25.4 wt. % CO₂; Figure 15 and Table 1). If the CO₂
519 content of the melt is converted into carbonate content at the source (which implies congruent
520 melting of the source), the latter could have contained up to 20-40 wt. % of carbonate.

521 [Dasgupta et al. \(2013\)](#) experimentally showed that carbonated silicate melt with 15-25 wt. %
522 CO₂ and a modest amount of water (1-5 wt. %) can be generated in the upper part of the
523 asthenospheric mantle (1,300-1,750 °C and 2-5 GPa) and is able to percolate upward the
524 lithospheric mantle. We conclude that the high CO₂/melt ratios determined in this study

525 correspond to deep melts derived from slab melting, which have percolated to lithospheric
526 depths without having been substantially fractionated.

527

528 4.4 A lithospheric carbon reservoir?

529

530 The mass ratios of CO₂/peridotite have been calculated for each xenoliths which provide
531 values between 2700 and 24,400 ppm by weight of CO₂ with an average of about 9557 ppm.

532 This is considered representative of the average quantity of carbon stored in the samples
533 examined. However, the present study is based on fluid inclusion-rich samples only, which
534 correspond to ~20 % of an initial sample set of collected mantle xenoliths. If we make a
535 conservative assumption that the remaining 80% of the initial sample set contains on average
536 30 ppm carbon, a typical estimate for the N-MORB mantle (Marty, 2012), then we can
537 deduce a first order estimate of the CO₂ content present in the Pannonian lithospheric mantle.
538 Thus, we estimate that quantity to be 1935 ppm CO₂ ($0.2 \times 9557 \text{ ppm} + 0.8 \times 30 \text{ ppm} = 1935$
539 ppm). We consider this first order CO₂ content as representative of the bulk lithospheric
540 mantle sampled by the alkaline volcanism of the BBHVF. This estimate is within a factor of
541 two of the 1000 ppm estimated by Kelemen & Manning (2015) for back-arc lithospheric
542 mantle based on observed carbon fluxes and reservoirs at subduction zones. For comparison,
543 the carbon content at source regions of MORB is thought to be bound between 50 and 200
544 ppm, whereas the source mantle of OIB is richer and may have be between 1000 and 4000
545 ppm CO₂ (Marty and Tolstikhin, 1998; Saal et al., 2002; Pineau et al., 2004; Cartigny et al.,
546 2008). We suggest that the quantitative peridotite + melt + volatile characteristics described
547 here, and found in many other regions of the Earth, may be representative of the carbon
548 reservoir residing in the lithospheric mantle, at least for back-arc areas.

549 Besides the implications of our findings for the carbon cycle, such a lithospheric carbon
550 reservoir has several consequences for the degassing of mantle volatiles in active geodynamic
551 settings such as the Pannonian Basin, where mantle-derived CO₂ outgassing through the
552 Pannonian Basin sedimentary rocks is observed (Ballentine et al., 1991). This suggests that
553 the carbon reservoir of at least ~2000 ppm quantified in this study is transient, and may be
554 recycled to the atmosphere and to the asthenosphere over geodynamic timescales by passive
555 tectonic degassing, active volcanism or delamination.

556

557 5 Conclusions

558

559 The magmatic-CO₂ budget of the subcontinental lithospheric mantle below the Pannonian
560 Basin has been constrained by coupling synchrotron 3-D X-ray microtomography, nanoSIMS,
561 Raman spectroscopy, and microthermometry in combination with thermodynamic modeling
562 in order to investigate the significance of abundant CO₂-rich fluid inclusions, CO₂-bearing
563 glass and vesicles within lithospheric peridotite xenoliths.

564 3-D volume reconstructions allow quantifying the proportions of all mineral phases in the
565 xenoliths and their textural relationships. Interconnection between trails of secondary fluid
566 inclusions and the network of glass + vesicles demonstrates a common fluid composition
567 between fluid inclusions and melt percolation in these mantle xenoliths. The vapor phase
568 volume (vesicles + fluid inclusions) observed requires CO₂-supersaturation of the silicate
569 melts, which allows to calculate melt saturation pressures based on the dissolved CO₂ content
570 of the glass.

571 The CO₂/glass mass ratio corresponding to the melt + vesicle system is estimated from the
572 microtomography, NanoSIMS analyses and thermodynamic modeling. The CO₂/melt mass
573 ratios obtained range from 9.0 and 25.4 wt. % and are related to a slab-derived melting

574 source. A first estimate of ~1935 ppm CO₂ for the Pannonian Basin lithospheric mantle is
575 proposed and can be considered as representative of the bulk lithospheric mantle sampled by
576 the alkaline basalt volcanism of the BBHVF.

577 This petrologic and geochemical approach is the first direct quantification of the bulk carbon
578 content in the lithospheric mantle, and is in agreement with independent lines of reasoning
579 suggesting a large transient reservoir of carbon in the lithospheric mantle near subduction-arc
580 systems. Further quantification of the volume, residence time, and general significance of this
581 reservoir requires more systematic regional and global studies using similar approach.

582

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592

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803

1 Figures captions

2

3 Figure 1: Principles of synchrotron X-ray microtomography. a- Sample is placed into the
4 beam in different angular positions; the transmitted beam is saved by the detector. b- All
5 radiographs obtained are compiled and a 3D image is reconstructed mathematically. c- Each
6 voxel of the 3D image is assigned to a grey level proportional to the local attenuation
7 coefficient of the incident beam. d- Example of drilled peridotite sample.

8

9 Figure 2: Reconstructed X-ray tomography 3D image of the SZB51 sample (4 mm diameter).
10 The dark grey phase is orthopyroxene, the medium-grey grey phase is olivine and the white is
11 clinopyroxene. We can observe in black the grain boundaries, cracks and some trails of
12 secondary fluid inclusions.

13

14 Figure 3: Density histograms of xenolith SZB51 (A, B) and xenolith FT12 (C, D) acquired by
15 X-ray tomography. Left: full histograms. Right: deconvolved histogram with density peak
16 assignment (B, D).

17

18 Figure 4: Example of phase density threshold defining for fluid vesicle segmentation of
19 xenolith SZB51. Three cases of density threshold assigning were defined for each sample.
20 The “Chosen fluid threshold” is considered the best estimated threshold. The minimum
21 (“Min. fluid threshold”) and maximum (“Max. fluid threshold”) thresholds represent the
22 upper and lower acceptable values near the best estimate. These provide a sense of the
23 uncertainty associated with the user defined density threshold assigning.

24

25 Figure 5: NanoSIMS CO₂ and H₂O calibration curves (dashed curves) obtained with the
26 tholeiitic glass standards (black dots) provided by CRPG, Nancy, France [standard description
27 available in [Shishkina et al. \(2010\)](#)]. CO₂ and H₂O were normalized to SiO₂.

28

29 Figure 6: X-ray microtomography images illustrating the variety of textures and the different
30 evidences of glass and fluid inclusions in mantle xenoliths. a - Protogranular texture. b-
31 Equigranular mosaic texture. c- Equigranular tabular texture. d- Melt pocket with large
32 vesicles (Vs), glass (Gl) and secondary crystallized minerals. e- A melt vein (glass + vesicles)
33 and trails of secondary fluid inclusions (FI). Opx – orthopyroxene, Ol – olivine, Opx –
34 orthopyroxene, SP- spinel, Gl – glass, Vs – vesicle.

35

36 Figure 7: 3D representation of the segmented volumes of glass (blue) and fluid (pink and
37 black) obtained by X-ray tomography in xenolith SZB51 (4 mm diameter). This image clearly
38 illustrates the crosscutting relationship between trails of fluid inclusions and glass+vesicles
39 along grain boundaries. In this sample 5 Vol. % of the fluid phase is in fluid inclusions (black)
40 and 95 Vol. % is in vesicles (pink) trapped in glass.

41

42 Figure 8: Example of melt pocket volume segmentation based on X-ray tomography. Pink,
43 blue and brown phases refer to vesicles, glass and newly formed minerals, respectively.

44

45 Figure 9: a- Abundances of glass, fluid (FI + vesicles) and mineral assemblages for each
46 studied xenoliths ([Creon et al., this volume](#)); b- AbundanceFluid inclusion abundances by
47 host mineral in SZB51.

48

49 Figure 10: Glass and fluid volume correlation. Arrows indicate the crystal fractionation
50 correction of the glass volume. Diamonds represent xenoliths with melt pockets + melt veins;
51 circles represent xenoliths containing only melt veins. Black, dark-grey and light-grey
52 symbols represent respectively SZB (Szentbékállá), SZG (Szigliget) and FT (Füzes-tó)
53 xenoliths.

54

55 Figure 11: Comparison between calculated densities based on EPMA chemical analyses
56 (Dixon et al., 1995) and measured densities by X-ray micro-CT for olivine, orthopyroxene,
57 clinopyroxene and glass. The shaded areas correspond to density range for melt of basalt,
58 andesite and trachyte ([Richet et al. 2000 and references therein](#))

59

60 Figure 12: A- Correlation between density determination methods (Raman spectroscopy vs.
61 microthermometry); B- Histogram of FI density for locations of the studied xenoliths (SZB -
62 Szentbékállá, SZG - Szigliget & FT - Füzes-tó)

63

64 Figure 13: CO₂ saturation curves calculated with rhyolite-MELTS (Gualda et al., 2012;
65 Ghiorso and Gualda, 2015) at 1000 °C using glass compositions obtained by EMPA and H₂O
66 contents obtained by Nano-SIMS measurements. Saturation pressure estimates of the melt
67 prior to quenching are defined by the intercept of the CO₂ contents in glass (NanoSIMS data)
68 and the CO₂ saturation curves (grey lines). Black diamonds: SZB xenoliths), dark-grey
69 diamonds: SZG xenoliths) and light-grey diamonds: FT xenoliths)

70

71 Figure 14: H₂O vs. CO₂ for glass trapped as vein or melt pockets in SZB (Szentbékállá);
72 SZG (Szigliget) and FT (Füzes-tó) mantle xenoliths. For comparison melt inclusions from arc

73 basalts are shown (Grey fields, Modified from [Wallace, 2005](#)). Literature data are shown for
74 Cerro Negro (Roggensack et al., 1997), Fuego (Fg; [Roggensack 2001](#)), Guatemalan volcanoes
75 behind the volcanic front (Guat BVF; [Walker et al. 2003](#)), the Trans-Mexican Volcanic Belt
76 (Luhr, 2001; Cervantes and Wallace, 2003), Stromboli (Metrich et al., 2001) and Cayuse
77 Crater, a cinder cone with OIB-like trace element composition from the Oregon Cascades
78 (Cas OIB; [M. Rowe, unpublished data](#)). For Cerro Negro, only inclusions from the 1992
79 explosive eruption are plotted. For comparison primary CO₂ contents of N-MORB ([Marty and](#)
80 [Tolstikhin, 1998](#)), E-MORB ([Hekinian et al., 2000](#)), Kilauea basalt ([Gerlach et al., 2002](#)) and
81 popping-rock ([Graham and Sarda, 1991](#)) are shown. Also shown is the CO₂ content of basaltic
82 magma from Reunion based on rare high-pressure melt inclusions ([Bureau et al., 1999](#)). SZB,
83 Szentbékállá; SZG, Szigliget; FT, Füzes-tó; Fuego; Guat (BVF), Guatemalan volcanoes
84 behind the volcanic front; Mexico, the Trans-volcanic Belt; Stromb., Stromboli; Cas OIB,
85 Cayuse Crater from the Oregon Cascades.

86

87 Figure 15: H₂O vs. CO₂ diagram illustrating the NanoSIMS data from peridotite hosted glass,
88 and the reconstructed CO₂ contents of the melt+fluid system. For comparison, subducted
89 sediments from [Plank and Langmuir \(1998\)](#), altered MORB from [Staudigel et al. \(1989\)](#), N-
90 MORB from [Marty and Tolstikhin \(1998\)](#), E-MORB from [Hekinian et al. \(2000\)](#) are shown.
91 Green area corresponds to the range of compositions of the subducting crust. Pink area
92 corresponds to plausible compositions for magmas produced from incongruent melting of the
93 subducted crust. Blue area corresponds to melts relatively depleted in volatiles issued from
94 slab melting and modified by subsequent volatile depletion, such as the assimilation of
95 volatile poor material and/or some physical fractionation during magma decompression.

96

Figure 1
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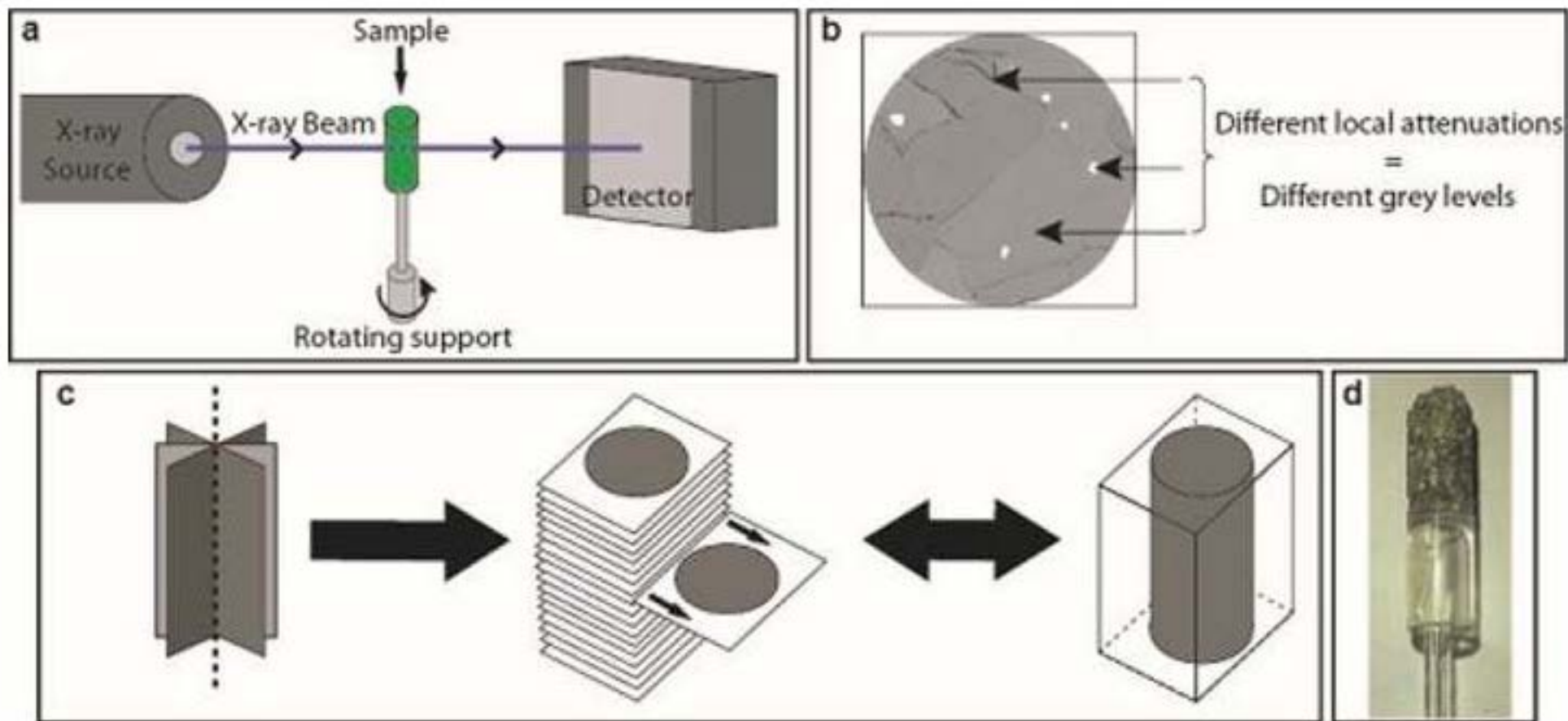


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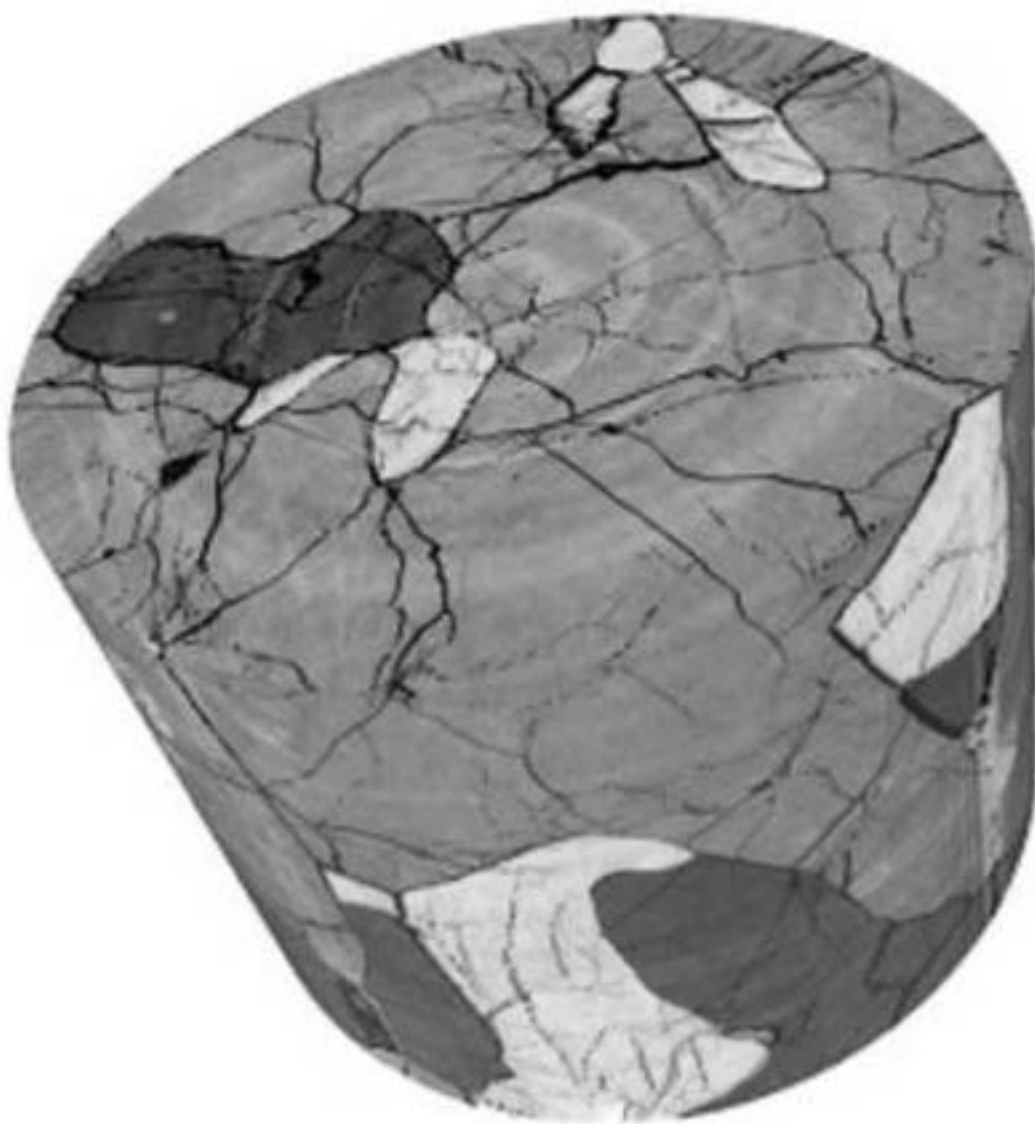


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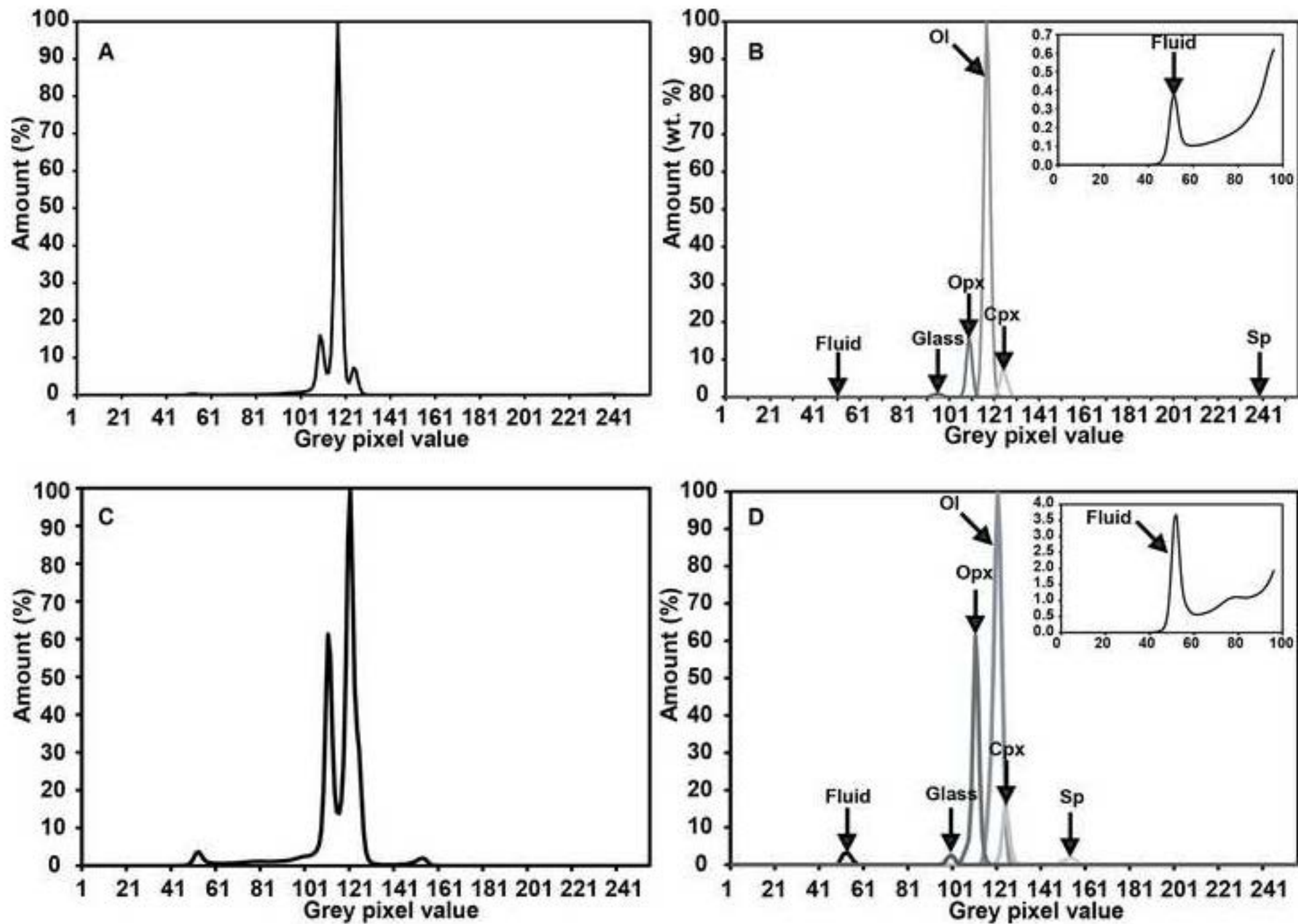


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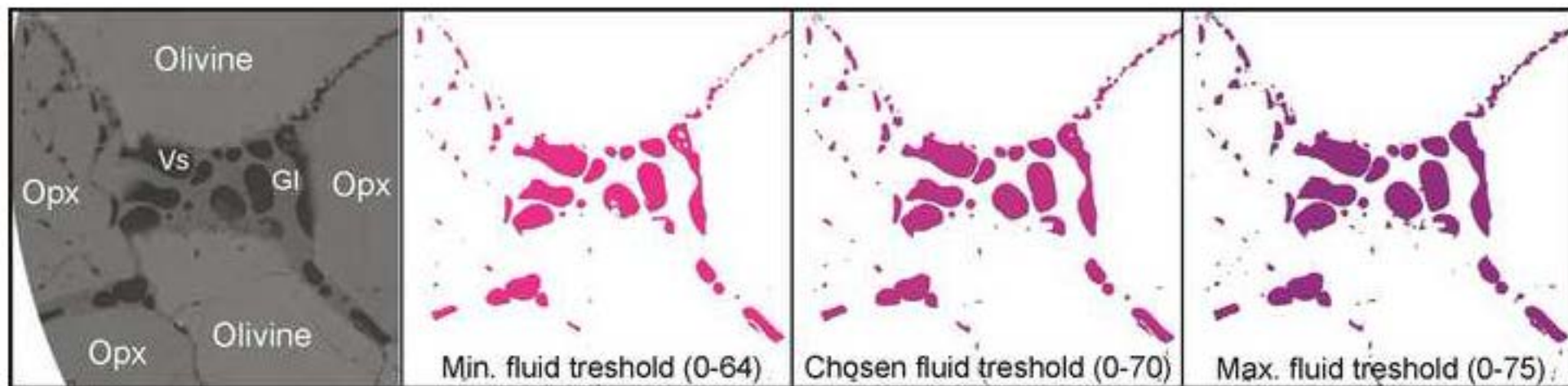


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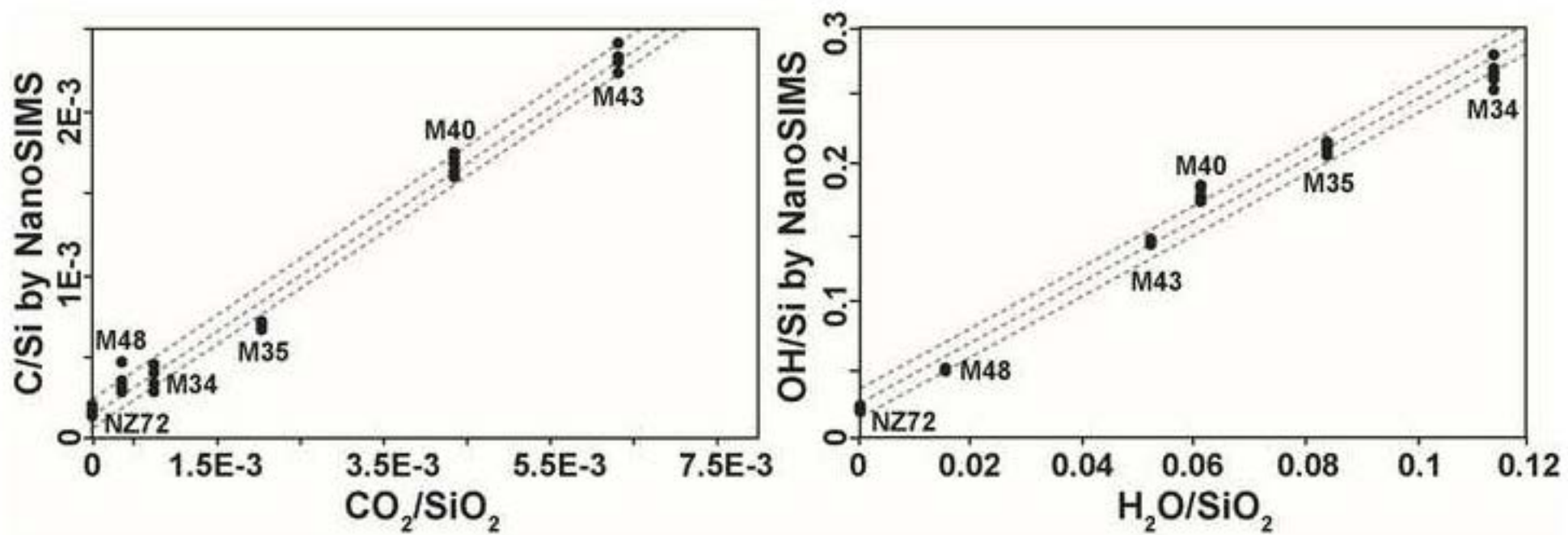


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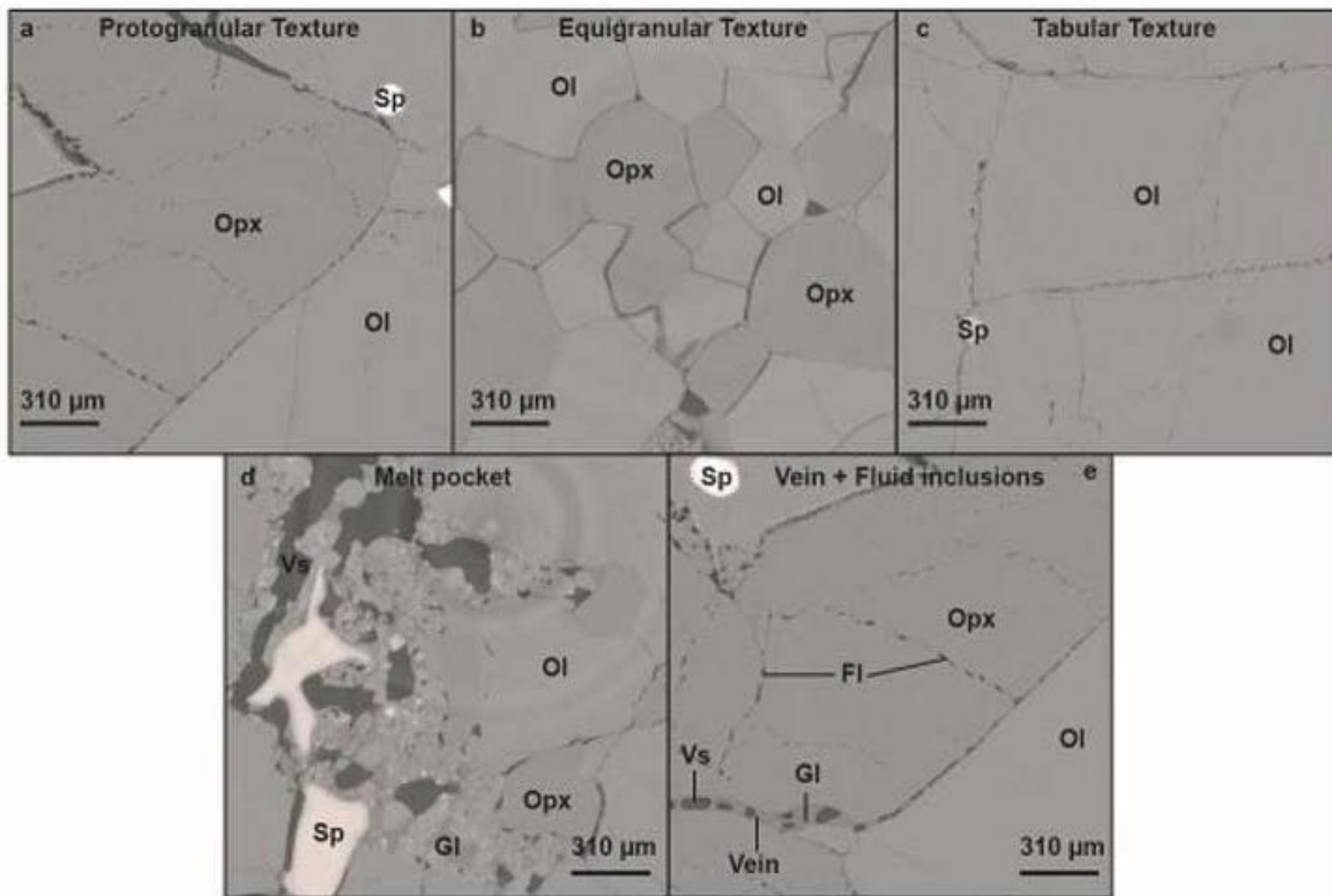


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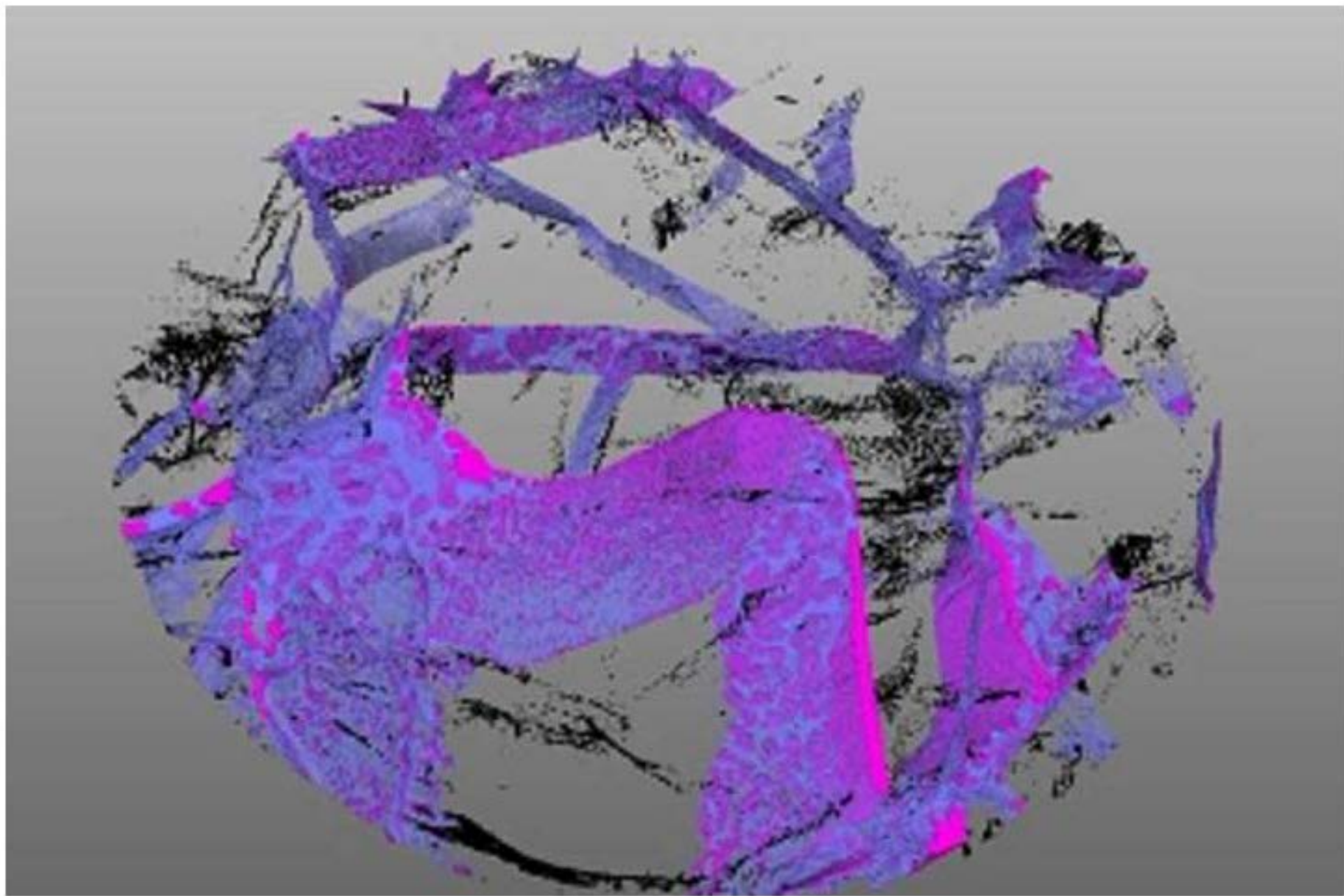


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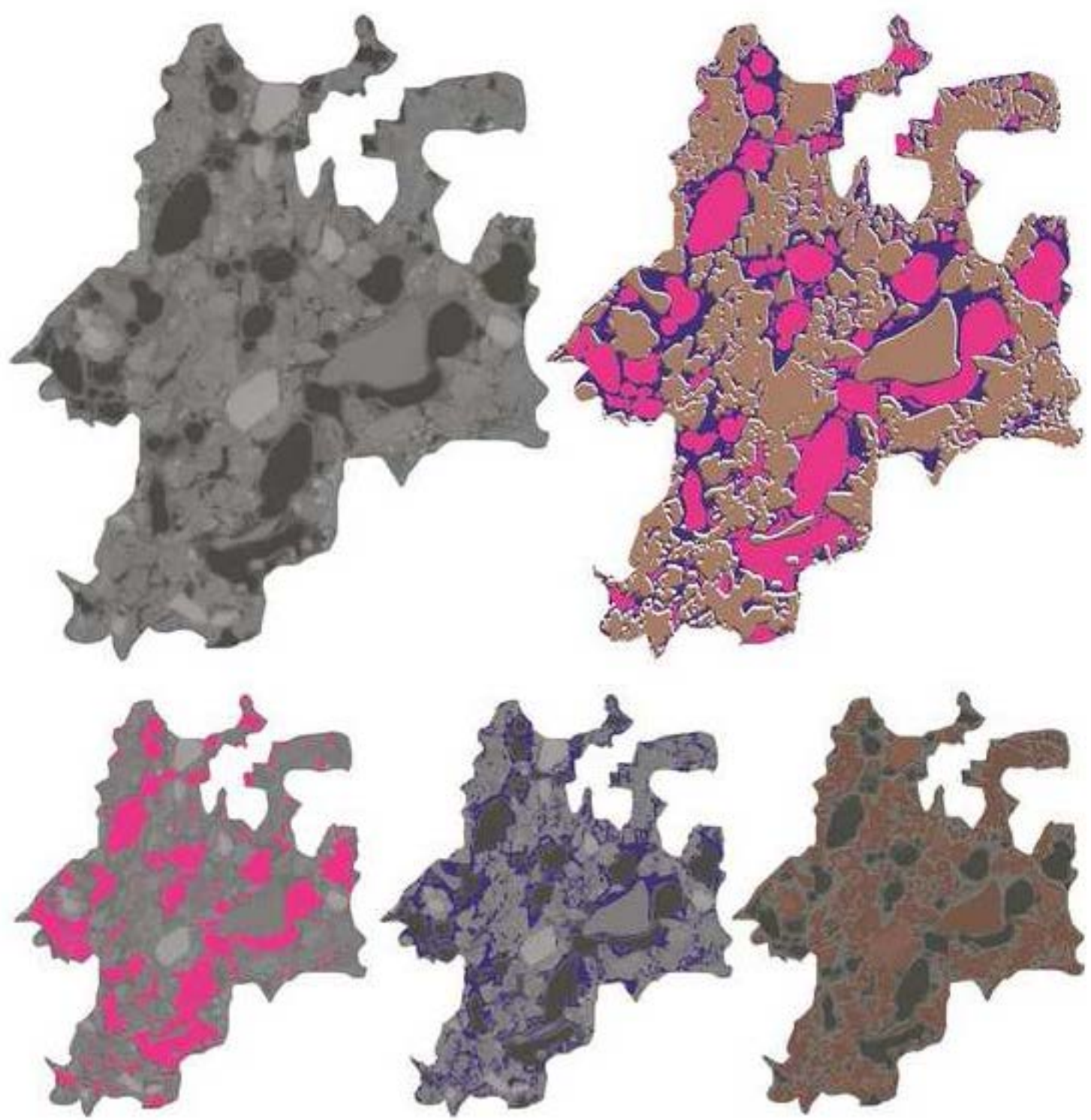


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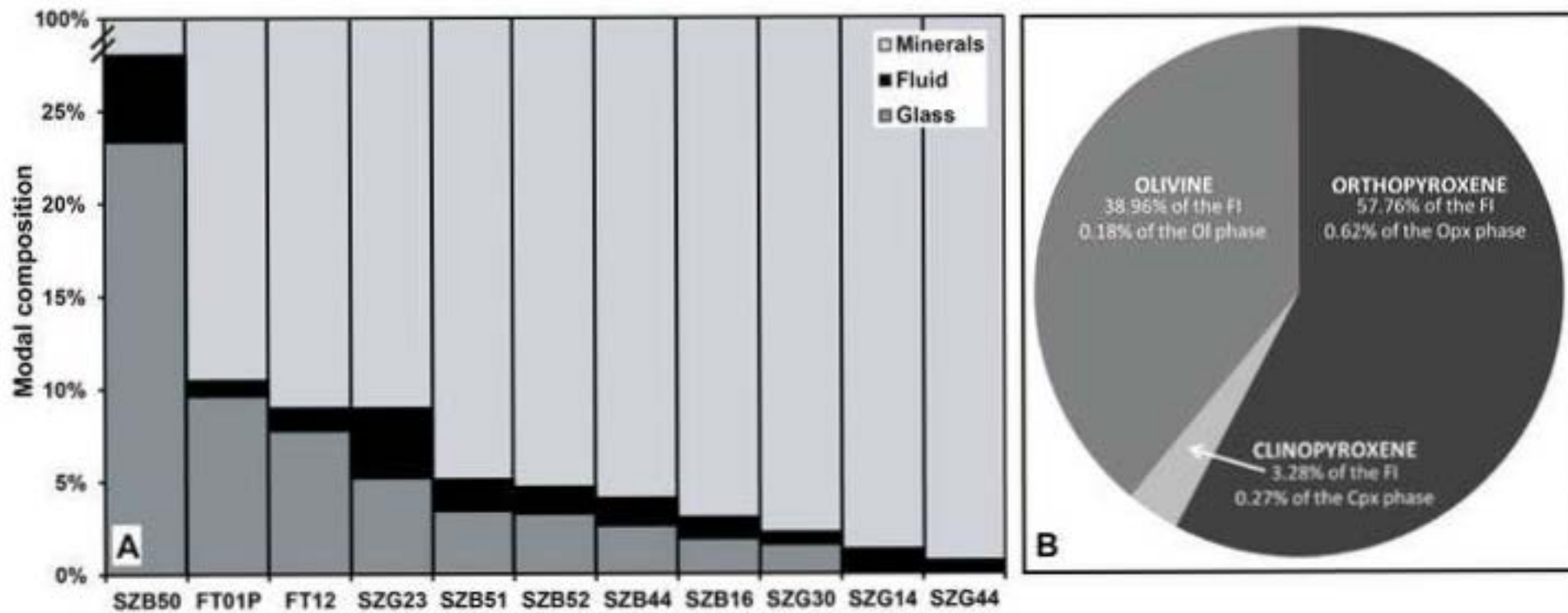


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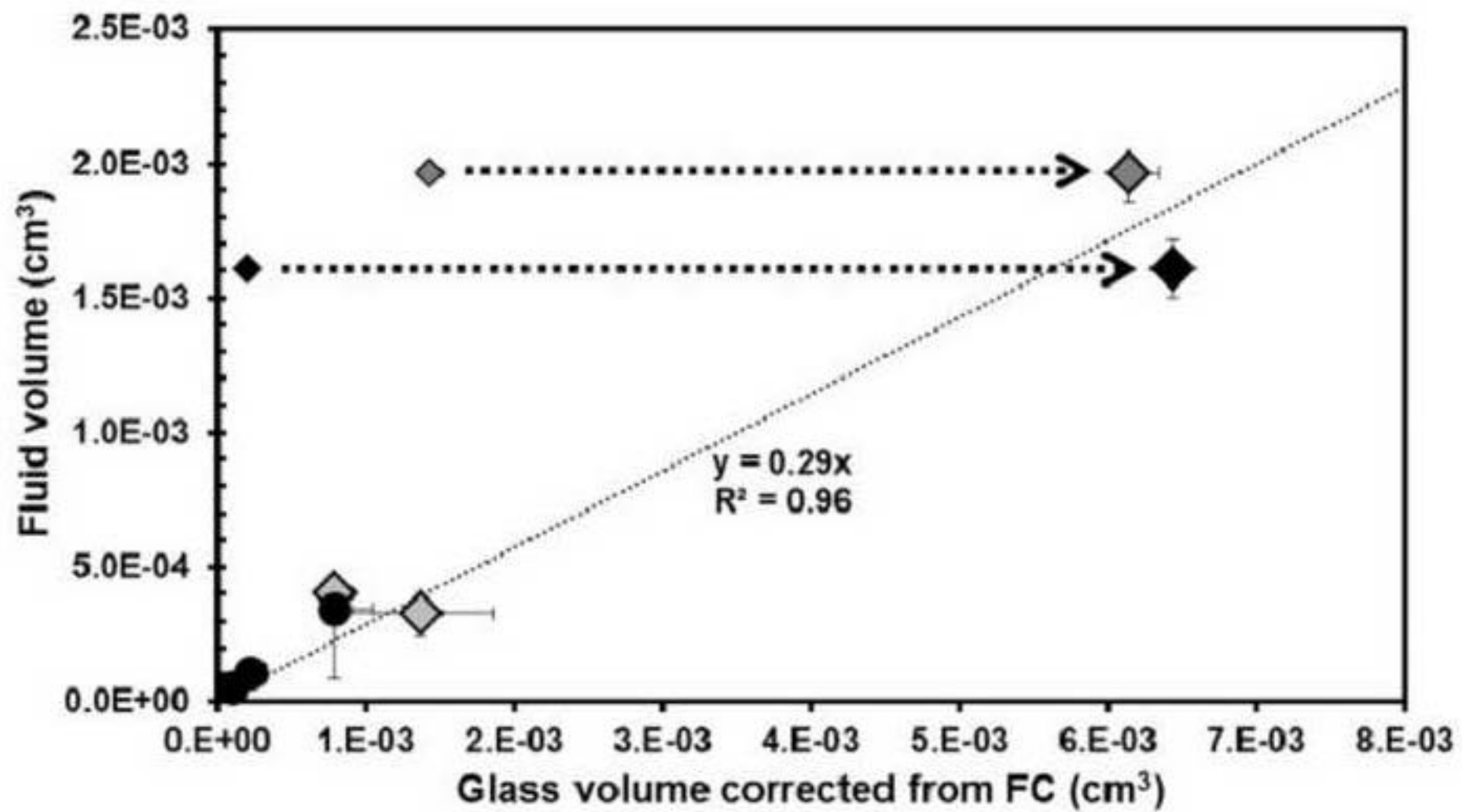


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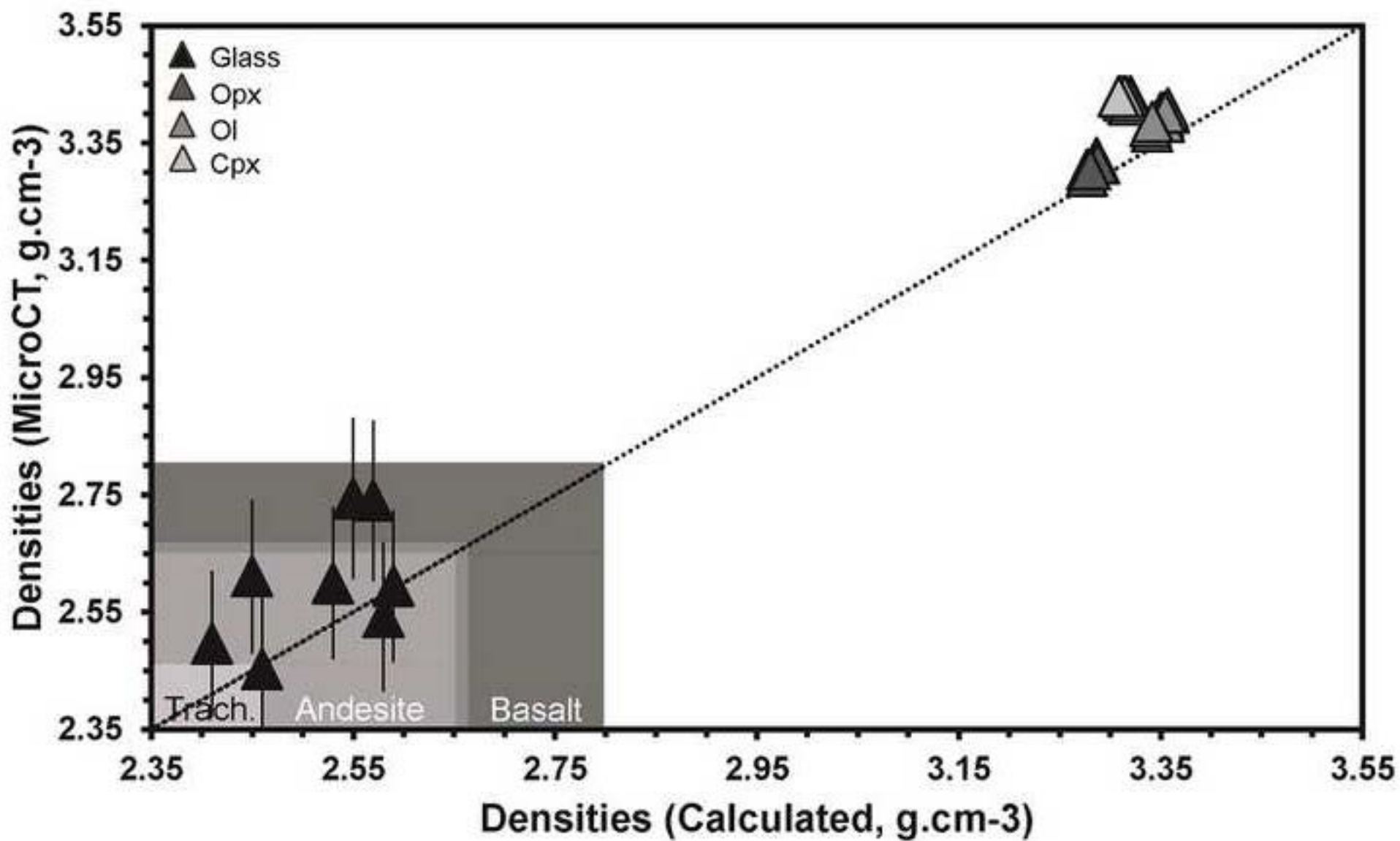


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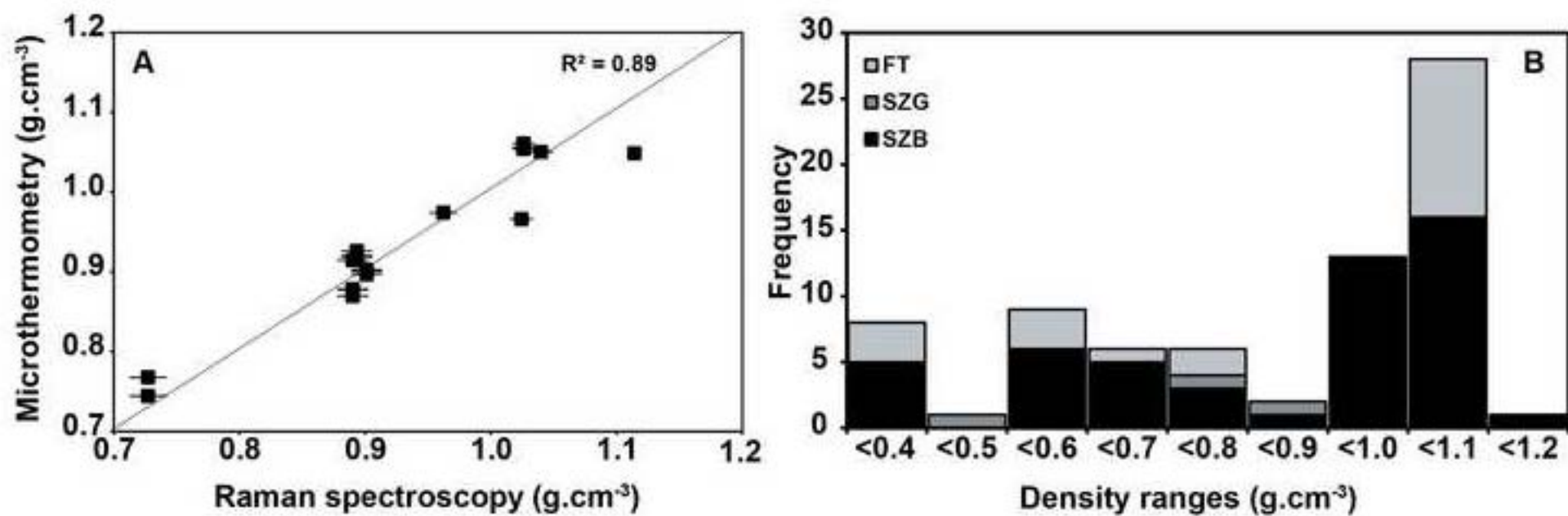


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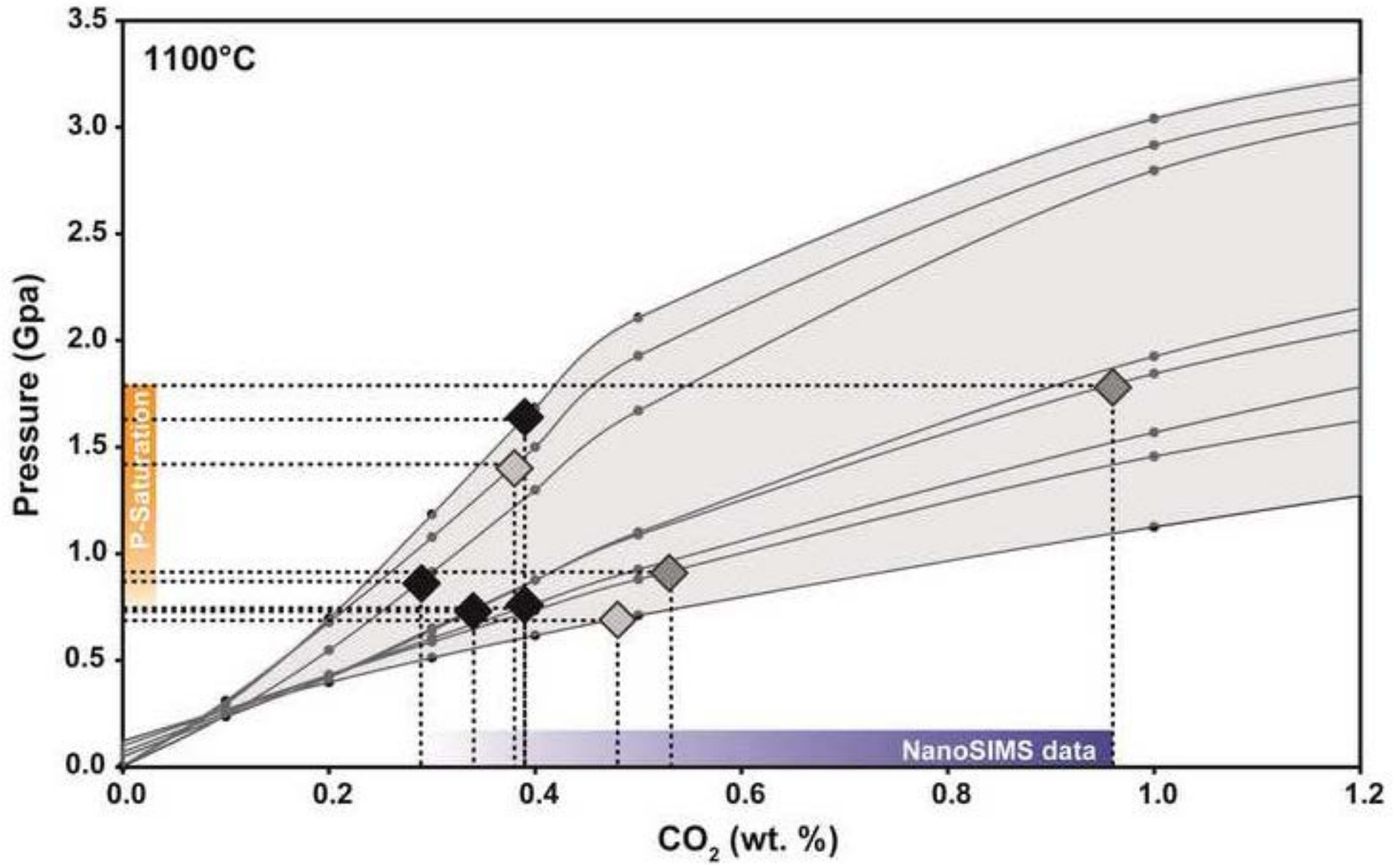
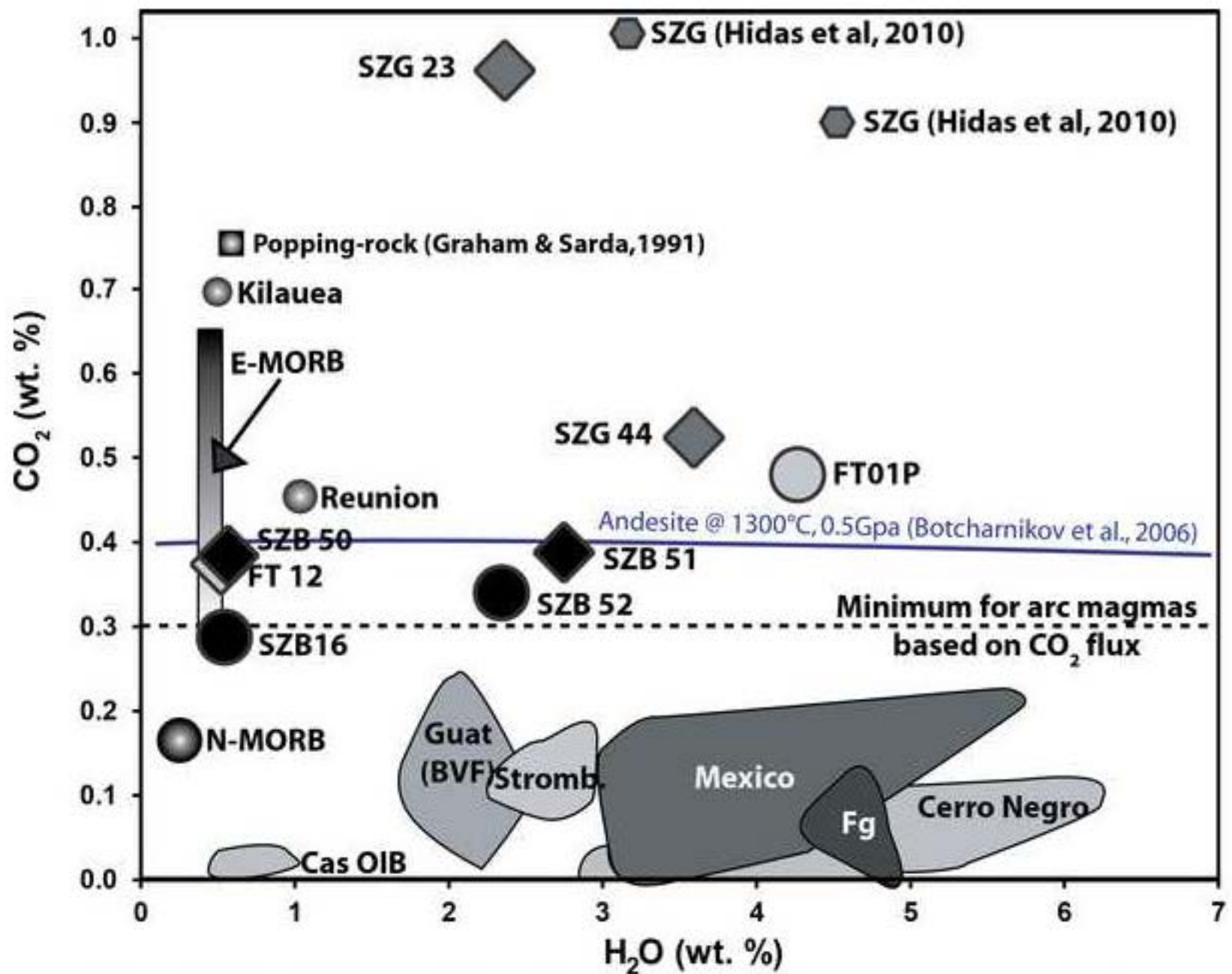


Figure 14

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SZB, Szentbékállá; SZG, Szigliget; FT, Füzes-Tó; Fg, Fuego; Guat (BVF), Guatemalan volcanoes behind the volcanic front, Mexico, the Trans-Mexican Volcanic Belt; Stromb., Stromboli; Cas OIB, Cayuse Crater from the Oregon Cascades

Figure 15

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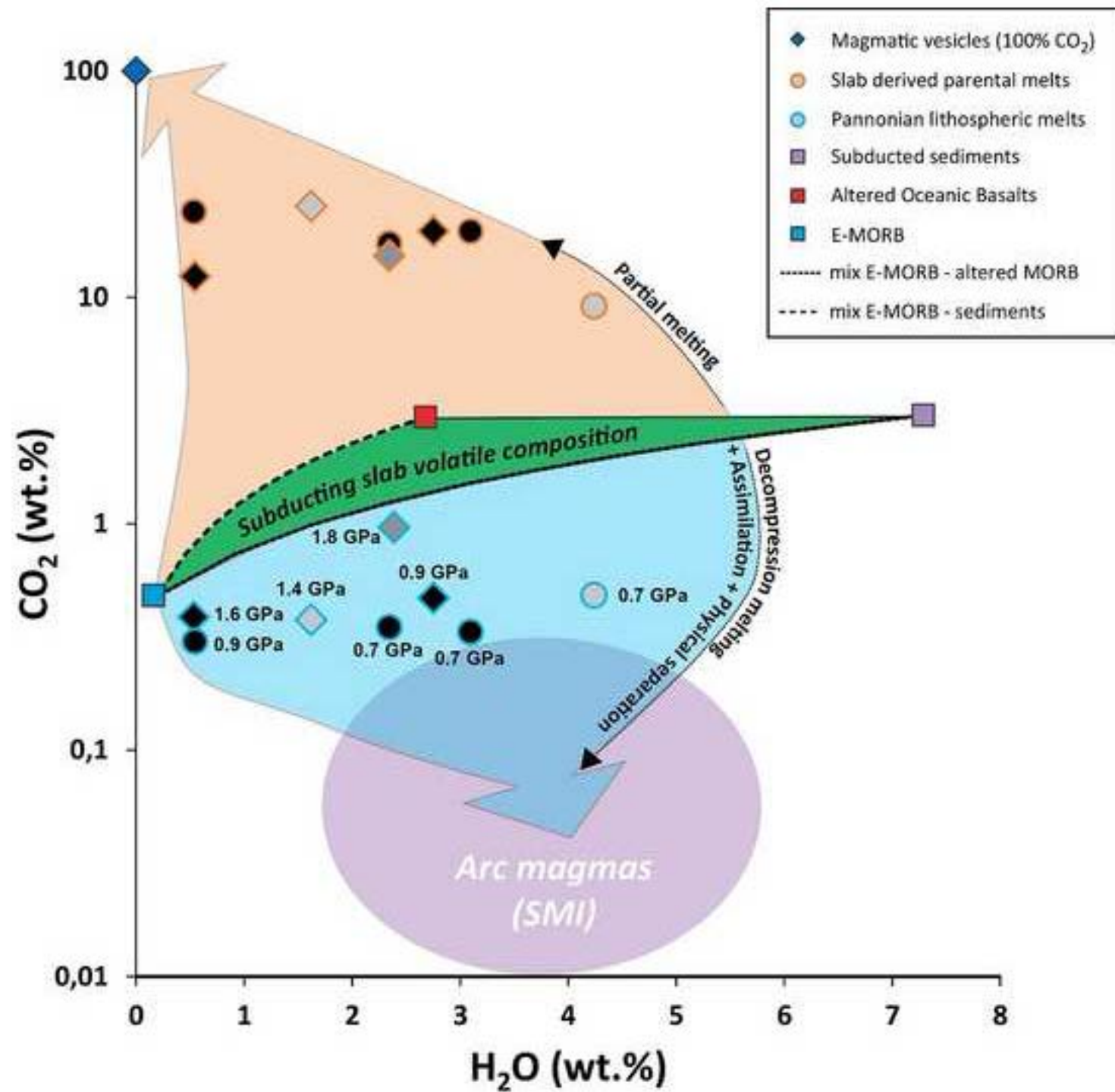


Table 1

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							Vol. (cm ³)	Glass volume (cm ³)			Fluid volume (cm ³)			Density (g/cm ³)			Mass (g)				
Sample	MP	SiO ₂	H ₂ O	Error (1σ)	CO ₂	Error (1σ)	Minerals	Best estimate	Min (%)	Max (%)	Best estimate	Min (%)	Max (%)	Glass	Vesicles	FI (Max)	Glass	CO ₂ (vesicle)	CO ₂ (dissolved)	CO ₂ /glass (wt. %)	CO ₂ /π (ppm)
SZB16	no	56.6	0.54	0.26	0.29	0.14	4.6E-03	9.1E-05	3.7E+01	1.3E+01	5.5E-05	1.5E+01	3.3E+01	2.599	1.022	0.573	2.4E-04	5.6E-05	6.8E-07	24.1	3723
SZB50	no	55.4	0.59	0.26	0.39	0.14	3.1E-02	6.4E-03	6.6E+00	6.4E+00	1.6E-03	1.3E+00	1.4E+01	2.740	1.295	0.617	1.8E-02	2.1E-03	6.8E-05	12.2	21287
SZB51-v	no	61.9	3.1	0.26	0.33	0.16															
SZB51-MP	yes	56.6	2.76	0.28	0.46	0.15	6.0E-03	2.1E-04	1.4E+01	2.6E-04	1.1E-04	3.2E+01	2.0E+01	2.496	0.970	1.024	5.3E-04	1.0E-04	2.1E-06	19.9	5409
SZB51-Av	yes	59.6	2.95	0.28	0.39	0.16															
SZB52	no	57.9	2.35	0.26	0.34	0.15	2.3E-02	7.8E-04	1.3E+01	1.0E-03	3.4E-04	5.8E+00	3.4E+01	2.452	0.953	1.114	1.9E-03	3.3E-04	6.5E-06	17.4	4422
SZG23	yes	56.3	2.38	0.26	0.96	0.20	3.5E-02	6.1E-03	2.3E+00	1.6E-03	2.0E-03	3.2E+00	1.3E+01	2.744	1.330	0.703	1.7E-02	2.6E-03	1.6E-04	16.5	24383
SZG44	yes	51.7	3.45	0.24	0.53	0.14	nd		0.0E+00	0.0E+00	1.3E-04	nd	nd	0.000	1.045	0.824	nd	1.4E-04	nd	nd	nd
FT12	yes	54.8	0.55	0.26	0.38	0.14	3.1E-02	7.8E-04	4.2E+00	9.7E-04	4.0E-04	1.0E+01	1.5E+01	2.542	1.228	0.661	2.0E-03	5.0E-04	7.5E-06	25.4	4946
FT01P	no	57.8	4.25	0.27	0.48	0.16	3.6E-02	1.4E-03	9.3E+00	1.7E-03	3.3E-04	4.9E+01	3.9E+01	2.611	0.930	1.057	3.6E-03	3.0E-04	1.7E-05	9.0	2727

Table 1: Summary of petrographic observations, chemical compositions and phase volumes, densities and masses. V – vein; MP – melt pocket; Av – average composition; Vol. – volume; Min & Max – Relative determined errors for each best estimate; vs - vesicle; FI – fluid inclusion; gl – glass; π - peridotite