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1 **Highly CO<sub>2</sub>-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 CO<sub>2</sub> degassing occurring in active tectonic intracontinental settings. We propose new  
25 constraints on the CO<sub>2</sub> 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 CO<sub>2</sub> 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 CO<sub>2</sub>  
36 and H<sub>2</sub>O in glass veins and pockets range between  $0.27 \pm 0.01$  (1 $\sigma$ ) and  $0.96 \pm 0.02$  (1 $\sigma$ ), and  
37  $0.54 \pm 0.26$  (1 $\sigma$ ) and  $4.25 \pm 0.27$  (1 $\sigma$ ) wt. %, respectively, and have higher CO<sub>2</sub> contents than  
38 typical arc magmas (0-0.25 wt. % CO<sub>2</sub> and 1.9-6.3 wt. % H<sub>2</sub>O). 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 CO<sub>2</sub>/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 CO<sub>2</sub> supersaturated  
44 in their source region. We suggest that slab derived melts initially containing up to 25 wt. %  
45 of CO<sub>2</sub> migrated congruently into the lithospheric mantle, and were subsequently fractionated  
46 by CO<sub>2</sub> segregation in secondary fluid inclusions by fracturing and creeping of the peridotite  
47 mineral matrix. A first order estimation of the bulk minimal CO<sub>2</sub> 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; CO<sub>2</sub>-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 CO<sub>2</sub>  
61 degassing at mid-ocean ridges, CO<sub>2</sub>-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 CO<sub>2</sub> 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 <sup>3</sup>He/<sup>4</sup>He  
69 and δ<sup>13</sup>C 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 CO<sub>2</sub> 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 CO<sub>2</sub> towards  
80 the surface and, more importantly, transient storage of CO<sub>2</sub> 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 CO<sub>2</sub> 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 CO<sub>2</sub> 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 CO<sub>2</sub> through the lithosphere. This study develops an innovative  
94 multi-technique approach in order to investigate the abundance and significance of CO<sub>2</sub> 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  $\mu\text{m}$ . 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<sup>®</sup> and Avizo<sup>®</sup> 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<sup>®</sup> (Fig. 3). Then the  
173 3-D volumes of each petrographic phase were defined with the Avizo<sup>®</sup> 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<sup>+</sup> 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 <sup>12</sup>C, <sup>16</sup>O<sup>1</sup>H<sup>-</sup>, <sup>28</sup>Si<sup>-</sup>, <sup>27</sup>Al<sup>16</sup>O<sup>-</sup> and <sup>56</sup>Fe were collected using electron multipliers over 3 x  
188 3 μm<sup>2</sup> square using the beam blanking mode (1.5 x 1.5 μm<sup>2</sup>) to reduce surface contamination  
189 (Stephant et al., 2014). A presputtering on a 3 × 3 μm<sup>2</sup> 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<sup>-10</sup> 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 CO<sub>2</sub> densities in inclusions were calculated from the two Raman  
222 bands of CO<sub>2</sub> 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 CO<sub>2</sub>-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 CO<sub>2</sub>)  
230 with a heating rate of 0.5-2 °C.min<sup>-1</sup>. Final homogenization occurred to the liquid phase  
231 (T<sub>h</sub>L). The CO<sub>2</sub> density and pCO<sub>2</sub> 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  $\mu\text{m}$ ,  
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<sup>-3</sup>), Opx (3.29 to 3.32 g.cm<sup>-3</sup>) and Cpx (3.42 to 3.43 g.cm<sup>-3</sup>)  
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<sup>-3</sup> (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<sup>-3</sup>) 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<sup>-3</sup>), 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 CO<sub>2</sub>  
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 CO<sub>2</sub> 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 (R<sup>2</sup>). 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<sup>-3</sup>  
322 <sup>3</sup>, 0.48 and 0.82 g.cm<sup>-3</sup> and 0.30 and 1.06 g.cm<sup>-3</sup> (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 CO<sub>2</sub> and H<sub>2</sub>O contents. H<sub>2</sub>O contents vary from  $0.54 \pm 0.26$  (1 $\sigma$ ) to  $4.25 \pm 0.27$  (1 $\sigma$ ) wt.  
328 % in glass from veins and from  $0.55 \pm 0.26$  (1 $\sigma$ ) to  $2.76 \pm 0.28$  (1 $\sigma$ ) wt. % in glass from melt  
329 pockets. CO<sub>2</sub> contents vary between  $0.27 \pm 0.01$  (1 $\sigma$ ) and  $0.48 \pm 0.02$  (1 $\sigma$ ) wt. % in glass from  
330 veins and from  $0.38 \pm 0.01$  (1 $\sigma$ ) to  $0.96 \pm 0.02$  (1 $\sigma$ ) wt. % in glass from melt pockets (Table  
331 1).

332

## 333 4 Discussion

334

### 335 4.1 Magma CO<sub>2</sub>-supersaturation conditions

336

337 Several petrographic lines of evidence point to CO<sub>2</sub> supersaturation in the melt trapped within  
338 the studied xenoliths at lithospheric depths. Such evidence includes 1) the occurrence of CO<sub>2</sub>  
339 FI with densities above  $1.0 \text{ g.cm}^{-3}$  (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 CO<sub>2</sub> and H<sub>2</sub>O 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, CO<sub>2</sub> 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 H<sub>2</sub>O and CO<sub>2</sub> 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 CO<sub>2</sub>-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 CO<sub>2</sub> in the high  
352 pressure range of 0.5-3.0 GPa, as also mentioned by [Papale et al. \(2006\)](#). Based on CO<sub>2</sub>-  
353 saturation isotherms, vesicle-melt equilibrium pressures were calculated from the CO<sub>2</sub>  
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<sub>g</sub>) 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). CO<sub>2</sub>  
362 appears to have almost no effect on the glass transition of silicate melts unlike H<sub>2</sub>O, 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 CO<sub>2</sub> 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 CO<sub>2</sub> 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<sup>-3</sup> 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 CO<sub>2</sub> 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 CO<sub>2</sub> and H<sub>2</sub>O 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 CO<sub>2</sub> concentrations of ~1 wt. % are in  
391 agreement with the maximum CO<sub>2</sub> content measured in glass for this study in sample SZG23  
392 from the same xenolith locality (0.8-1.2 wt. % CO<sub>2</sub>; 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 CO<sub>2</sub>-rich  
395 percolating melts analyzed in this study.

396

397 4.2 CO<sub>2</sub> 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 CO<sub>2</sub>. This  
409 assumption is consistent with the high H<sub>2</sub>O and CO<sub>2</sub> contents in the glass determined by  
410 nanoSIMS.

411 The CO<sub>2</sub>/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 CO<sub>2</sub>;  $M(\text{Glass})$  : mass of glass;  $V(\text{CO}_2)$  : volume of CO<sub>2</sub>;  $V(\text{Glass})$  : volume  
417 of glass;  $\rho(\text{CO}_2)$  : density of CO<sub>2</sub>;  $\rho(\text{Glass})$  : 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 CO<sub>2</sub>/melt mass ratios range from 9.0 to 25.4 wt. % (Table 1). These ratios,  
424 which can be considered as the CO<sub>2</sub> 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 CO<sub>2</sub>/melt mass ratios calculated in this study  
430 implies that CO<sub>2</sub> 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 CO<sub>2</sub>/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 CO<sub>2</sub>-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 CO<sub>2</sub>-rich metasomatic agent during melt-rock reactions due to carbon saturation and vapor  
445 immiscibility in the percolating silicate melts. Thus, the variability in CO<sub>2</sub> 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 CO<sub>2</sub> 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 CO<sub>2</sub> mass fraction.

455

#### 456 4.2.2. Fluid inclusions and physical fractionation

457

458 The large abundance of CO<sub>2</sub>-rich FIs trapped in minerals along fractures and the absence of  
459 comparably abundant silicate melt inclusions indicate that CO<sub>2</sub> 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 SiO<sub>2</sub>), 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 CO<sub>2</sub> 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 CO<sub>2</sub> in dunite ([Watson & Brenan, 1987](#)). Consequently, melt  
479 inclusions only rarely trap melts with their primary CO<sub>2</sub> 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 CO<sub>2</sub> 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 CO<sub>2</sub>-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 CO<sub>2</sub>  
494 and H<sub>2</sub>O 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 CO<sub>2</sub> 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 H<sub>2</sub>O 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 CO<sub>2</sub>

500 content of arc magmas sampled by SMIs may only represent a negligible fraction of the CO<sub>2</sub>  
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 CO<sub>2</sub>-rich melt may be trapped as SMIs due to the pre-entrapment supersaturation in  
504 CO<sub>2</sub> and the preferential accumulation of most of the CO<sub>2</sub> 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 CO<sub>2</sub>/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 CO<sub>2</sub> vesicles and silicate melt during magma ascent to the crust, 2)  
510 important dilution of CO<sub>2</sub>-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 H<sub>2</sub>O and CO<sub>2</sub> 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 CO<sub>2</sub> and H<sub>2</sub>O contents like those calculated in this  
518 study (0.55-4.25 wt. % H<sub>2</sub>O and 9.0-25.4 wt. % CO<sub>2</sub>; Figure 15 and Table 1). If the CO<sub>2</sub>  
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 CO<sub>2</sub> 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 CO<sub>2</sub>/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 CO<sub>2</sub>/peridotite have been calculated for each xenoliths which provide  
531 values between 2700 and 24,400 ppm by weight of CO<sub>2</sub> 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 CO<sub>2</sub> content present in the Pannonian lithospheric mantle.  
538 Thus, we estimate that quantity to be 1935 ppm CO<sub>2</sub> ( $0.2 \times 9557 \text{ ppm} + 0.8 \times 30 \text{ ppm} = 1935$   
539 ppm). We consider this first order CO<sub>2</sub> 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 CO<sub>2</sub> (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 CO<sub>2</sub> 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-CO<sub>2</sub> 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 CO<sub>2</sub>-rich fluid inclusions, CO<sub>2</sub>-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 CO<sub>2</sub>-supersaturation of the silicate  
569 melts, which allows to calculate melt saturation pressures based on the dissolved CO<sub>2</sub> content  
570 of the glass.

571 The CO<sub>2</sub>/glass mass ratio corresponding to the melt + vesicle system is estimated from the  
572 microtomography, NanoSIMS analyses and thermodynamic modeling. The CO<sub>2</sub>/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 CO<sub>2</sub> 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

## 593 7 References

594

595 Anderson A. T. (1975) Some basaltic and andesitic gases. *Rev. Geophys.* **13**, 37.

596 Bagdassarov N. S., Maumus J., Poe B., Slutskiy A. B. and Bulatov V. K. (2004) Pressure  
597 dependence of T<sub>g</sub> in silicate glasses from electrical impedance measurements. *Phys.*  
598 *Chem. Glas. - Eur. J. Glas. Sci. Technol. Part B*, **45**, 197–214.

599 Bali E., Szabó C., Vaselli O. and Torok K. (2002) Significance of silicate melt pockets in  
600 upper mantle xenoliths from the Bakony – Balaton Highland Volcanic Field , Western  
601 Hungary. *Lithos* **61**, 79–102.

602 Bali E., Zanetti A., Szabó C., Peate D. W. and Waight T. E. (2008) A micro-scale  
603 investigation of melt production and extraction in the upper mantle based on silicate melt  
604 pockets in ultramafic xenoliths from the Bakony–Balaton Highland Volcanic Field  
605 (Western Hungary). *Contrib. to Mineral. Petrol.* **155**, 165–179.

606 Ballentine C. J., O’Nions R. K., Oxburgh E. R., Horvath F. and Deak J. (1991) Rare gas  
607 constraints on hydrocarbon accumulation, crustal degassing and groundwater flow in the  
608 Pannonian Basin. *Earth Planet. Sci. Lett.* **105**, 229–246.

609 Balogh K. and Németh K. (2005) Evidence for the Neogene small-volume intracontinental  
610 volcanism in Western Hungary: K/Ar geochronology of the Tihany Maar Volcanic  
611 Complex. *Geol. Carpathica* **56**, 91–99.

612 Balogh K. and Pécskay Z. (2001) K/Ar and Ar/Ar geochronological studies in the Pannonian-  
613 Carpathians-Dinarides (PANCARDI) region. *Acta Geol. Hung.* **44**, 281–299.

614 Bartoli O., Cesare B., Remusat L., Acosta-Vigil A. and Poli S. (2014) The H<sub>2</sub>O content of  
615 granite embryos. *Earth Planet. Sci. Lett.* **395**, 281–290.

616 Baruchel J., Buffiere J.-Y., Maire E., Merle P. and Peix G. (2000) X-ray Tomography in  
617 Material Science. *Hermes Sci. Publ.*

618 Berkesi M., Hidas K., Guzmics T., Dubessy J., Bodnar R. J., Szabó C., Vajna B. and  
619 Tsunogae T. (2009) Detection of small amounts of H<sub>2</sub>O in CO<sub>2</sub>-rich fluid inclusions  
620 using Raman spectroscopy. *J. Raman Spectrosc.* **40**, 1461–1463.

621 Bodnar R. J. (2003) Re-equilibration of fluid inclusions. *Fluid Inclusions Anal. Interpret.*  
622 *Short Course Ser. Samson I, Anderson A, Marshall D Miner. Assoc Can, Vancouver* **32**,  
623 213–231.

- 624 Bonelli R. and Frezzotti M. . (2003) Raman Spectra Database.  
625 <http://www.dst.unisi.it/geofluidslab/Raman%20intro.html>.
- 626 Botcharnikov R., Behrens H. and Holtz F. (2006) Solubility and speciation of C–O–H fluids  
627 in andesitic melt at T=1100–1300°C and P=200 and 500MPa. *Chem. Geol.* **229**, 125–  
628 143.
- 629 Bruce Watson E. and Brenan J. M. (1987) Fluids in the lithosphere, 1. Experimentally-  
630 determined wetting characteristics of CO<sub>2</sub>H<sub>2</sub>O fluids and their implications for fluid  
631 transport, host-rock physical properties, and fluid inclusion formation. *Earth Planet. Sci.*  
632 *Lett.* **85**, 497–515.
- 633 Burton M. R., Sawyer G. M. and Granieri D. (2013) Deep Carbon Emissions from Volcanoes.  
634 *Rev. Mineral. Geochemistry* **75**, 323–354.
- 635 Cartigny P., Pineau F., Aubaud C. and Javoy M. (2008) Towards a consistent mantle carbon  
636 flux estimate: Insights from volatile systematics (H<sub>2</sub>O/Ce, δD, CO<sub>2</sub>/Nb) in the North  
637 Atlantic mantle (14° N and 34° N). *Earth Planet. Sci. Lett.* **265**, 672–685.
- 638 Chalot-Prat F. and Arnold M. (1999) Immiscibility between calciocarbonatitic and silicate  
639 melts and related wall rock reactions in the upper mantle: a natural case study from  
640 Romanian mantle xenoliths. *Lithos* **46**, 627–659.
- 641 Creon L., Delpech G., Rouchon V. and Guyot F. Melt-peridotite interaction in the lithospheric  
642 mantle - Genesis of the Pannonian calc-alkaline suites revealed by Alkali basalt hosted  
643 xenoliths. *this Vol.*
- 644 Dasgupta R. (2013) Ingassing, Storage, and Outgassing of Terrestrial Carbon through  
645 Geologic Time. *Rev. Mineral. Geochemistry* **75**, 183–229. Dasgupta R. and Hirschmann  
646 M. M. (2010) The deep carbon cycle and melting in Earth's interior. *Earth Planet. Sci.*  
647 *Lett.* **298**, 1–13.
- 648 Dasgupta R., Mallik A., Tsuno K., Withers A. C., Hirth G. and Hirschmann M. M. (2013)

649 Carbon-dioxide-rich silicate melt in the Earth's upper mantle. *Nature* **493**, 211–215.

650 Demény A., Dallai L., Frezzotti M.-L., Vennemann T. W., Embey-Isztin A., Dobosi G. and  
651 Nagy G. (2010) Origin of CO<sub>2</sub> and carbonate veins in mantle-derived xenoliths in the  
652 Pannonian Basin. *Lithos* **117**, 172–182.

653 Demény A., Vennemann T. ., Hegner E., Nagy G., Milton J. ., Embey-Isztin A., Homonnay Z.  
654 and Dobosi G. (2004) Trace element and C–O–Sr–Nd isotope evidence for subduction-  
655 related carbonate–silicate melts in mantle xenoliths (Pannonian Basin, Hungary). *Lithos*  
656 **75**, 89–113.

657 Dixon J. E., Stolper E. M., Holloway J. R. and Science A. (1995) An Experimental Study of  
658 Water and Carbon Dioxide Solubilities in Mid-Ocean Ridge Basaltic Liquids . Part II :  
659 Applications to Degassing. *J. Petrol.* **36**, 1607–1631.

660 Dunn P. K. (2007) tweedie: Tweedie exponential family models.

661 Embey-Isztin a, Dobosi G., Altherr R. and Meyer H.-P. (2001) Thermal evolution of the  
662 lithosphere beneath the western Pannonian Basin: evidence from deep-seated xenoliths.  
663 *Tectonophysics* **331**, 285–306.

664 Embey-Isztin A., Downes H., James D. E. E., Upton B. G. J. G. J., Dobosi G., Ingram G. A.  
665 a., Harmon R. S. S. and Scharbert H. G. G. (1993) The petrogenesis of Pliocene alkaline  
666 volcanic rocks from the Pannonian Basin, Eastern central Europe. *J. Petrol.* **34**, 317–343.

667 Embey-Isztin A., Sharbert H. G., Dietrich H. and Poultidis H. (1990) Mafic granulites and  
668 clinopyroxenite xenoliths from the Transd- nubian Volcanic Region (Hungary):  
669 implications for the deep structure of the Pannonian Basin. *Mineral. Mag.* **54**, 463–483.

670 Fall A., Tattitch B. and Bodnar R. J. (2011) Combined microthermometric and Raman  
671 spectroscopic technique to determine the salinity of H<sub>2</sub>O–CO<sub>2</sub>–NaCl fluid inclusions  
672 based on clathrate melting. *Geochim. Cosmochim. Acta* **75**, 951–964.

673 Falus G., Szabó C., Kovács I., Zajacz Z. and Halter W. (2007) Symplectite in spinel lherzolite

674 xenoliths from the Little Hungarian Plain, Western Hungary: A key for understanding  
675 the complex history of the upper mantle of the Pannonian Basin. *Lithos* **94**, 230–247.

676 Fodor L., Csontos L., Bada G., Györfi I. and Benkovics L. (1999) Tertiary tectonic evolution  
677 of the Pannonian basin and neighbouring orogenes: a new synthesis of palaeostress data.  
678 In: Durand, B., Jolivet, L., Horváth, F., Séranne, M. (Eds.), *The Mediterranean Basins: Tertiary Extension with Alpine Orogen*. *Geol. Soc.* **156**, 295–334.

680 Fyfe W. S., Price N. J. and Hompson A. B. (1978) Fluids in the Earth's Crust: Their  
681 Significance in Metamorphic, Tectonic, and Chemical Transport Processes. *Elsevier*,  
682 *Amsterdam*, 384 pp.

683 Ghiorso M. S. and Gualda G. A. R. (2015) An H<sub>2</sub>O-CO<sub>2</sub> mixed fluid saturation model  
684 compatible with rhyolite-MELTS. *Contrib. to Mineral. Petrol.*

685 Giordano D., Russell J. K. and Dingwell D. B. (2008) Viscosity of magmatic liquids: A  
686 model. *Earth Planet. Sci. Lett.* **271**, 123–134.

687 Gorman P. J., Kerrick D. M. and Connolly J. a D. (2006) Modeling open system metamorphic  
688 decarbonation of subducting slabs. *Geochemistry, Geophys. Geosystems* **7**.

689 Gualda G. A. R., Ghiorso M. S., Lemons R. V. and Carley T. L. (2012) Rhyolite-MELTS: a  
690 Modified Calibration of MELTS Optimized for Silica-rich, Fluid-bearing Magmatic  
691 Systems. *J. Petrol.* **53**, 875–890.

692 Guillot B. and Sator N. (2011) Carbon dioxide in silicate melts: A molecular dynamics  
693 simulation study. *Geochim. Cosmochim. Acta* **75**, 1829–1857.

694 Hammouda T. (2003) High-pressure melting of carbonated eclogite and experimental  
695 constraints on carbon recycling and storage in the mantle. *Earth Planet. Sci. Lett.* **214**,  
696 357–368.

697 Harangi S. and Lenkey L. (2007) Genesis of the Neogene to Quaternary volcanism in the  
698 Carpatian Pannonian region : Role of subduction , extension , and mantle plume. *Geol.*

699 *Soc. Am. Special Pa*, 67–92.

700 Hidas K., Guzmics T., Szabó C., Kovács I., Bodnar R. J., Zajacz Z., Nédli Z., Vaccari L. and  
701 Perucchi A. (2010) Coexisting silicate melt inclusions and H<sub>2</sub>O-bearing, CO<sub>2</sub>-rich fluid  
702 inclusions in mantle peridotite xenoliths from the Carpathian–Pannonian region (central  
703 Hungary). *Chem. Geol.* **274**, 1–18.

704 Horvath F. (1993) Towards a mechanical model for the formation of the Pannonian Basin.  
705 *Tectonophysics* **226**, 333–357.

706 Horváth F., Musitz B., Balázs a., Végh A., Uhrin a., Nádor A., Koroknai B., Pap N., Tóth T.  
707 and Wórum G. (2015) Evolution of the Pannonian basin and its geothermal resources.  
708 *Geothermics* **53**, 328–352.

709 Jarrard R. D. (2003) Subduction fluxes of water, carbon dioxide, chlorine, and potassium.  
710 *Geochemistry, Geophys. Geosystems* **4**.

711 Johnston F. K. B., Turchyn A. V. and Edmonds M. (2011) Decarbonation efficiency in  
712 subduction zones: Implications for warm Cretaceous climates. *Earth Planet. Sci. Lett.*  
713 **303**, 143–152.

714 Kazmer M. and Kovacs S. (1985) Eocene–Paleogene paleogeography along the Eastern part  
715 of the Insubric–Periadriatic Lineament system: evidence for continental escape of the  
716 Bakony- Drauzug unit. *Acta Geol. Hung.* **1-2**, 71–84.

717 Kelemen P. B. and Manning C. E. (2015) Reevaluating carbon fluxes in subduction zones,  
718 what goes down, mostly comes up. *Proc. Natl. Acad. Sci.*, 201507889.

719 Kovacs I., Falus G., Stuart G., Hidas K., Szabó C., Flower M. F. J., Hegedűs E., Posgay K.  
720 and Zilahi-Sebess L. (2012) Seismic anisotropy and deformation patterns in upper  
721 mantle xenoliths from the central Carpathian–Pannonian region: Asthenospheric flow as  
722 a driving force for Cenozoic extension and extrusion? *Tectonophysics* **514-517**, 168–179.

723 Kovács I. and Szabó C. (2008) Middle Miocene volcanism in the vicinity of the Middle

724 Hungarian zone: Evidence for an inherited enriched mantle source. *J. Geodyn.* **45**, 1–17.

725 Labiche J.-C., Mathon O., Pascarelli S., Newton M., Guilera Ferre G., Curfs C., Vaughan G.,  
726 Homs A. and D. F. C. (2007) The fast readout low noise camera as a versatile X-ray  
727 detector for time resolved dispersive extended X-ray absorption fine structure and  
728 diffraction studies of dynamic problems in materials science, chemistry, and catalysis.  
729 *Rev. Sci. Instrum.* **78**, 091301–091311.

730 Marty B. (2012) The origins and concentrations of water, carbon, nitrogen and noble gases on  
731 Earth. *Earth Planet. Sci. Lett.* **313-314**, 56–66.

732 Marty B. and Tolstikhin I. N. (1998) CO<sub>2</sub> fluxes from mid-ocean ridges, arcs and plumes.  
733 *Chem. Geol.* **145**, 233–248.

734 Mercier J. C. and Nicolas A. (1975) Textures and Fabrics of Upper-Mantle Peridotites as  
735 Illustrated by Xenoliths from Basalts. *J. Petrol.* **16**.

736 Morizet Y., Nichols A. R. L., Kohn S. C., Brooker R. a. and Dingwell D. B. (2007) The  
737 influence of H<sub>2</sub>O and CO<sub>2</sub> on the glass transition temperature: insights into the effects of  
738 volatiles on magma viscosity. *Eur. J. Mineral.* **19**, 657–669.

739 Paganin D., Mayo S. C., Gureyev T. E., Miller P. R. and Wilkins S. W. (2002) Simultaneous  
740 phase and amplitude extraction from a single defocused image of a homogeneous object.  
741 *J. Microsc.* **206**, 33–40.

742 Papale P., Moretti R. and Barbato D. (2006) The compositional dependence of the saturation  
743 surface of H<sub>2</sub>O+CO<sub>2</sub> fluids in silicate melts. *Chem. Geol.* **229**, 78–95.

744 Pineau F., Shilobreeva S., Hekinian R., Bideau D. and Javoy M. (2004) Deep-sea explosive  
745 activity on the Mid-Atlantic Ridge near 34°50'N: a stable isotope (C, H, O) study. *Chem.*  
746 *Geol.* **211**, 159–175.

747 Plank T. and Langmuir C. H. (1998) The chemical composition of subducting sediment and  
748 its consequences for the crust and mantle. *Chem. Geol.* **145**, 325–394.

- 749 Poinar K. (2012) The Carbon Cycle in the Mantle. , 1–11.
- 750 Praus O., Pěčová J., Petr V., Babuška V. and Plomerová J. (1990) Magnetotelluric and  
751 seismological determination of the lithosphere-asthenosphere transition in Central  
752 Europe. *Phys. Earth Planet. Inter.* **60**, 212–228.
- 753 Richet P., Whittington A., Holtz F., Behrens H., Ohlhorst S. and Wilke M. (2000) Water and  
754 the density of silicate glasses. *Contrib. to Mineral. Petrol.* **138**, 337–347.
- 755 Roedder E. (1984) Fluid inclusions. *Rev. Mineral.* **12**.
- 756 Saal A. E., Hauri E. H., Langmuir C. H. and Perfit M. R. (2002) Vapour undersaturation in  
757 primitive mid-ocean-ridge basalt and the volatile content of Earth's upper mantle. *Nature*  
758 **419**, 451–5.
- 759 Sanchez S., Ahlberg P. E., Trinajstic K. M., Mirone A. and Tafforeau P. (2012) Three  
760 Dimensional Synchrotron Virtual Paleohistology: A New Insight into the World of  
761 Fossil Bone Microstructures. *Microsc. Microanal.* **18**, 1095–1105.
- 762 Sano Y. and Marty B. (1995) Origin of carbon in fumarolic gas from island arcs. *Chem. Geol.*  
763 **119**, 265–274.
- 764 Schiano P., Provost A., Clocchiatti R., Faure F., Schiano P., Provost A., Clocchiatti R. and  
765 Faure F. (2006) Transcrystalline Melt Migration and Earth's Mantle. *Science (80-. )*.  
766 **314**, 970–974.
- 767 Shcheka S. S., Wiedenbeck M., Frost D. J. and Keppler H. (2006) Carbon solubility in mantle  
768 minerals. *Earth Planet. Sci. Lett.* **245**, 730–742.
- 769 Sherwood Lollar B., Ballentine C. J. and Onions R. K. (1997) The fate of mantle-derived  
770 carbon in a continental sedimentary basin: Integration of relationships and stable isotope  
771 signatures. *Geochim. Cosmochim. Acta* **61**, 2295–2307.
- 772 Shishkina T. a., Botcharnikov R. E., Holtz F., Almeev R. R. and Portnyagin M. V. (2010)  
773 Solubility of H<sub>2</sub>O- and CO<sub>2</sub>-bearing fluids in tholeiitic basalts at pressures up to

774 500MPa. *Chem. Geol.* **277**, 115–125.

775 Sleep N. H., Zahnle K. J. and Neuhoff P. S. (2001) Initiation of clement surface conditions on  
776 the earliest Earth. *Proc. Nat. Acad. Sci.* **98**, 3666–3672.

777 Spakman W. (1990) Tomographic images on the upper mantle below Europe and the  
778 Mediterranean. *Ter. Nov.* **2**, 542–553.

779 Span R. and Wagner W. (1996) A New Equation of State for Carbon Dioxide Covering the  
780 Fluid Region from the Triple-Point Temperature to 1100 K at Pressures up to 800 MPa.  
781 *J. Phys. Chem.* **25**, 1509–1596.

782 Stephant A., Remusat L., Thomen A. and Robert F. (2014) Reduction of OH contamination in  
783 quantification of water contents using NanoSIMS imaging. *Chem. Geol.* **380**, 20–26.

784 Sterner S. M. and Pitzer K. S. (1994) An equation of state for carbon dioxide valid from zero  
785 to extreme pressures. *Contrib. to Mineral. Petrol.* **117**, 362–374.

786 Szabó C., Harangi S. and Csontos L. (1992) Review of Neogene and Quaternary volcanism of  
787 the Carpathian-Pannonian region. *Tectonophysics* **208**, 243–256.

788 Szabó C., Kovacs I., Dégi J., Kothay K., Torok K., Hidas K. K., Konya P., Berkesi M., Degi  
789 J., Kothay K., Torok K., Hidas K. K., Konya P. and Berkesi M. (2010) From maars to  
790 lava lakes: Ultramafic and granulite xenoliths associated with the alkaline basaltic  
791 volcanism of the Pannonian Basin. In *Mineralogica-petrographica field guide series* pp.  
792 1–32.

793 Tašárová A., Afonso J. C., Bielik M., Götze H.-J. and Hók J. (2009) The lithospheric structure  
794 of the Western Carpathian–Pannonian Basin region based on the CELEBRATION 2000  
795 seismic experiment and gravity modelling. *Tectonophysics* **475**, 454–469.

796 Wallace P. J. (2005) Volatiles in subduction zone magmas: concentrations and fluxes based  
797 on melt inclusion and volcanic gas data. *J. Volcanol. Geotherm. Res.* **140**, 217–240.

798 Wijbrans J., Németh K., Martin U. and Balogh K. (2007)  $^{40}\text{Ar}/^{39}\text{Ar}$  geochronology of

799 Neogene phreatomagmatic volcanism in the western Pannonian Basin, Hungary. *J.*  
800 *Volcanol. Geotherm. Res.* **164**, 193–204. Youxue Zhang and Zindler A. (1993)  
801 Distribution and evolution of carbon and nitrogen in Earth. *Earth Planet. Sci. Lett.* **117**,  
802 331–345.  
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 CO<sub>2</sub> and H<sub>2</sub>O 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. \(2010\)](#)]. CO<sub>2</sub> and H<sub>2</sub>O were normalized to SiO<sub>2</sub>.

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: CO<sub>2</sub> 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 H<sub>2</sub>O  
66 contents obtained by Nano-SIMS measurements. Saturation pressure estimates of the melt  
67 prior to quenching are defined by the intercept of the CO<sub>2</sub> contents in glass (NanoSIMS data)  
68 and the CO<sub>2</sub> saturation curves (grey lines). Black diamonds: SZB xenoliths), dark-grey  
69 diamonds: SZG xenoliths) and light-grey diamonds: FT xenoliths)

70

71 Figure 14: H<sub>2</sub>O vs. CO<sub>2</sub> 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 CO<sub>2</sub> 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 CO<sub>2</sub> 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: H<sub>2</sub>O vs. CO<sub>2</sub> diagram illustrating the NanoSIMS data from peridotite hosted glass,  
88 and the reconstructed CO<sub>2</sub> 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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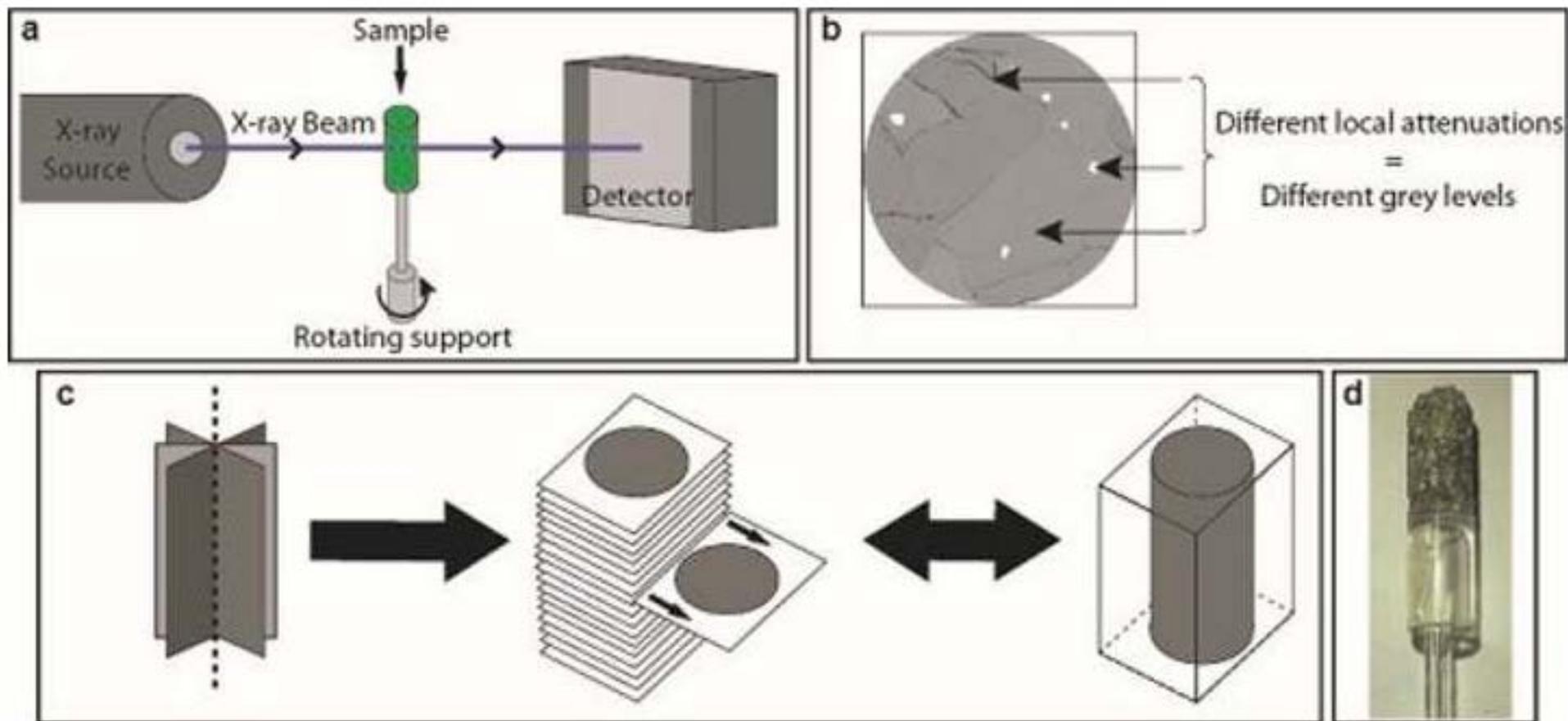


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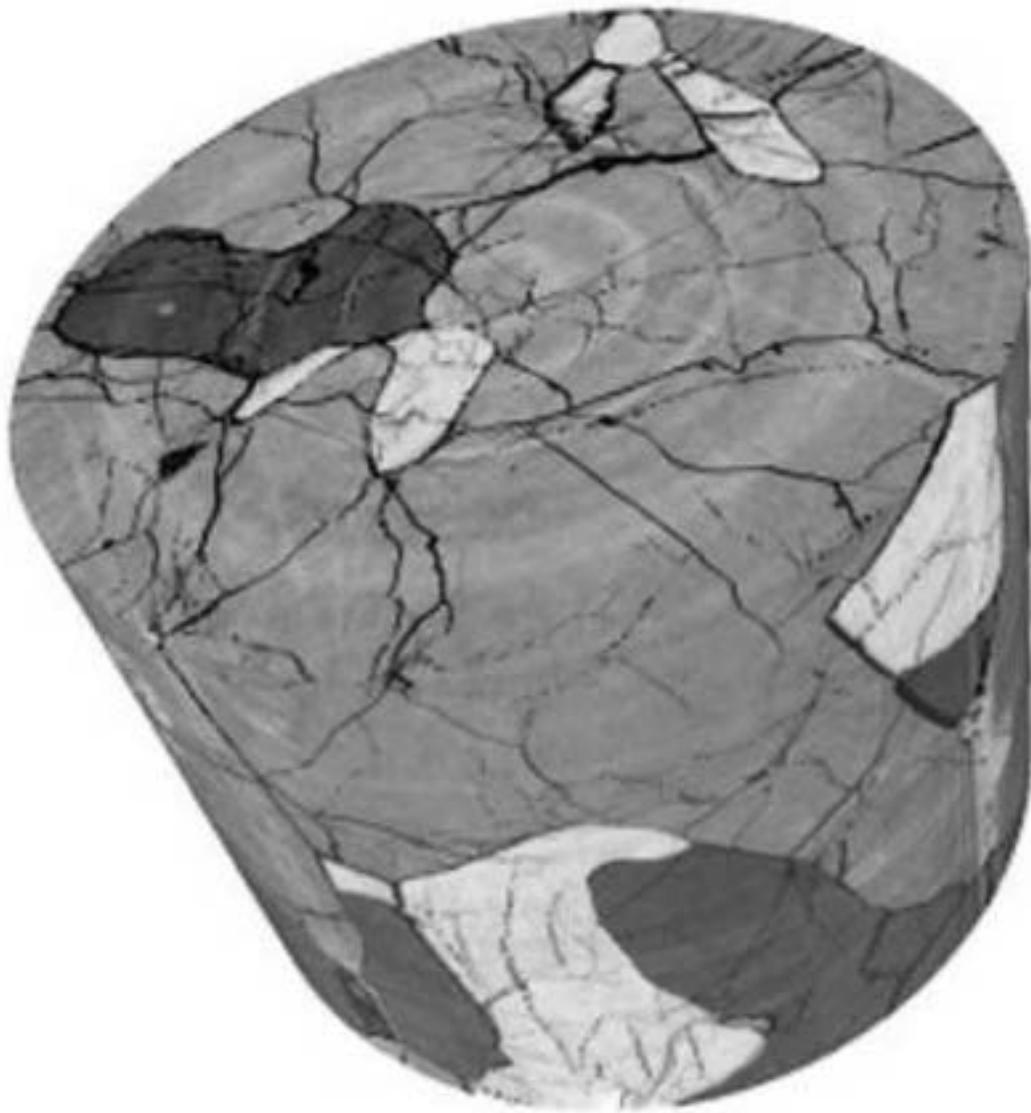


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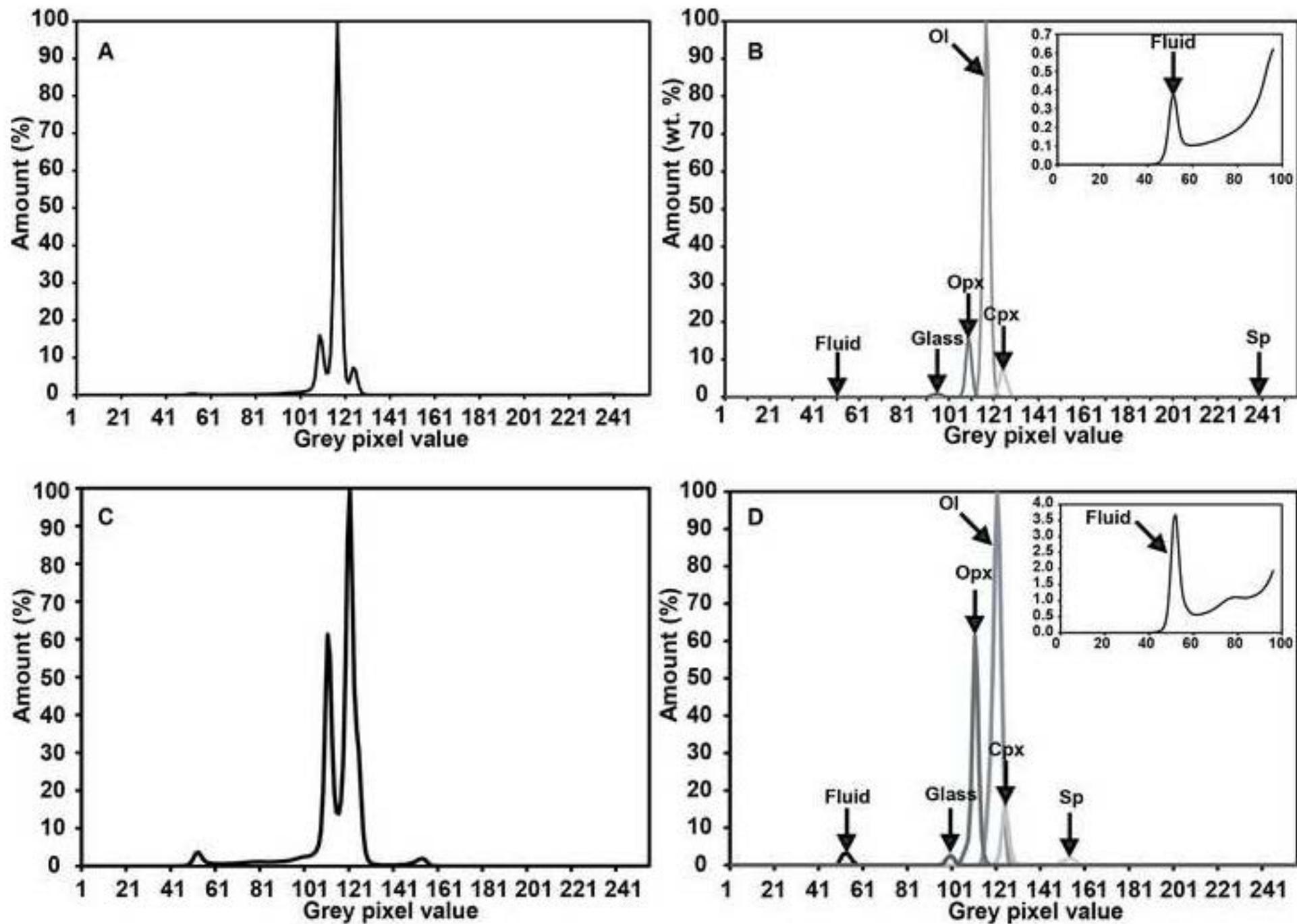


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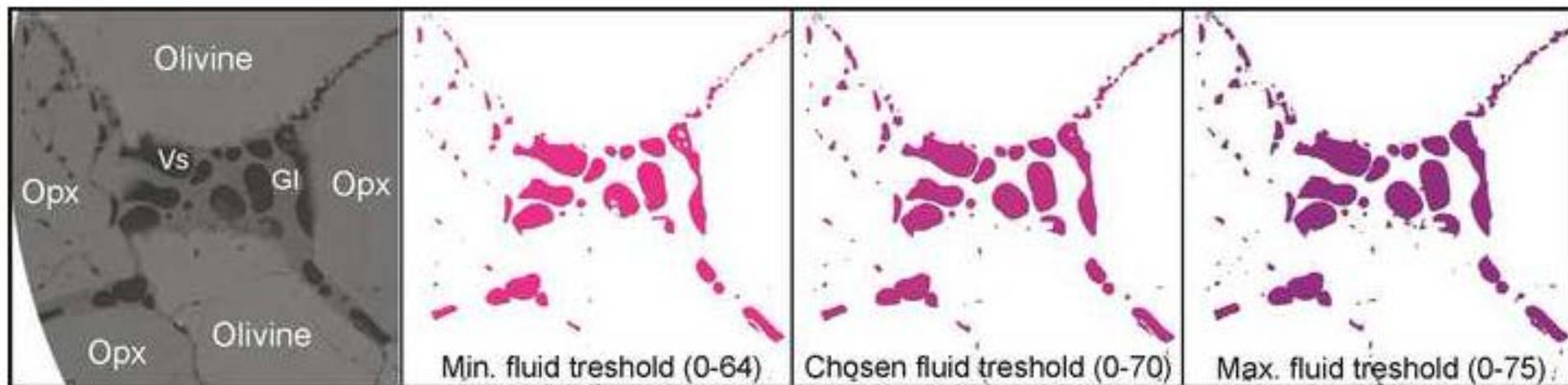


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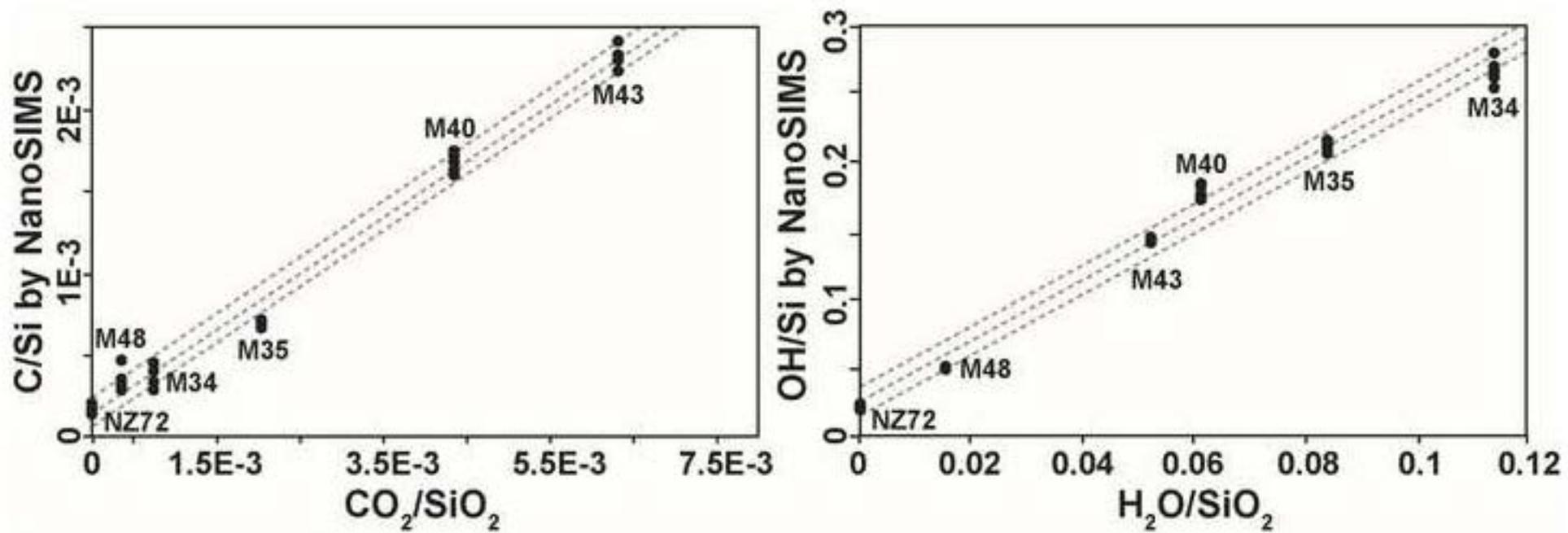


Figure 6  
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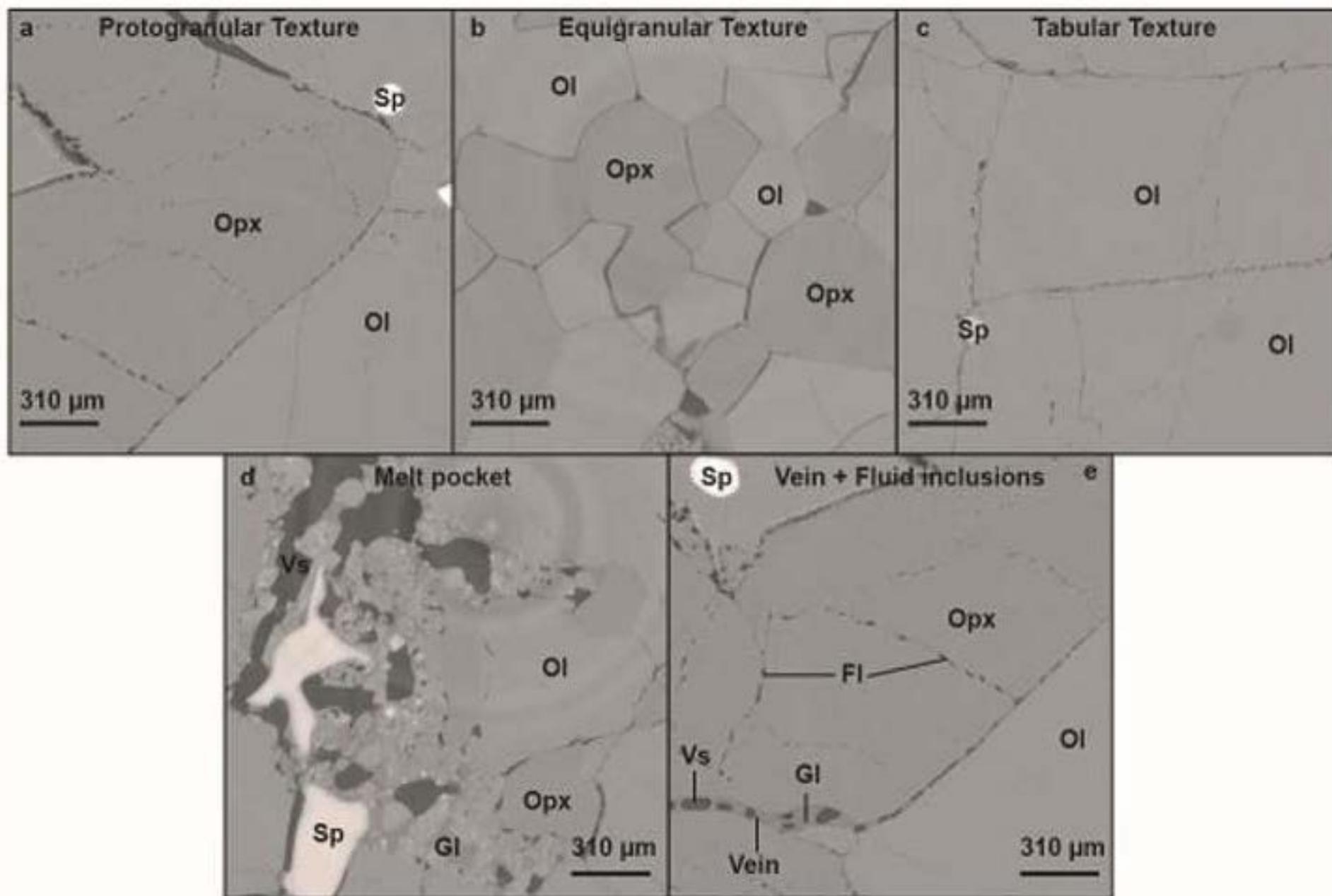


Figure 7  
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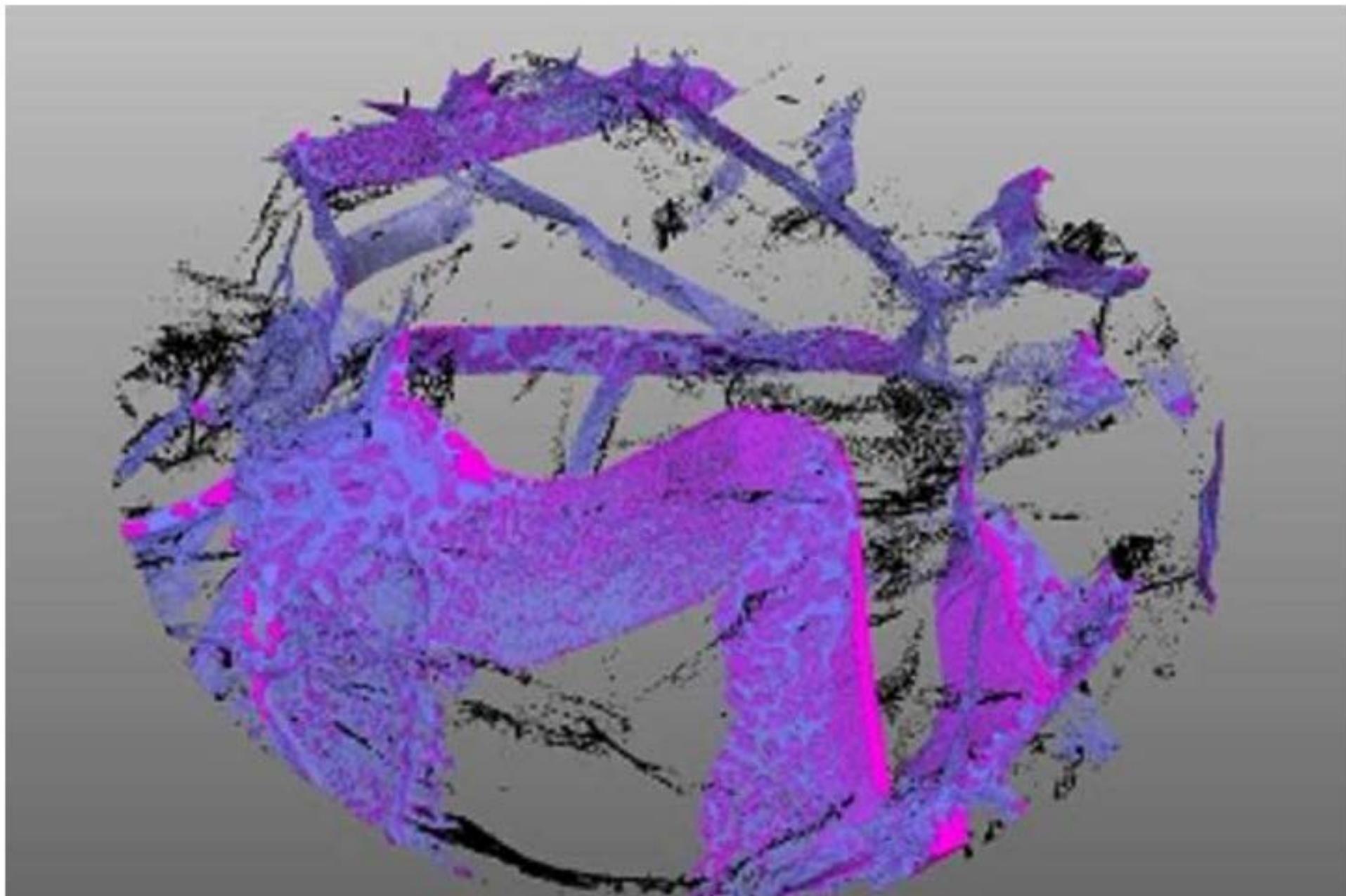


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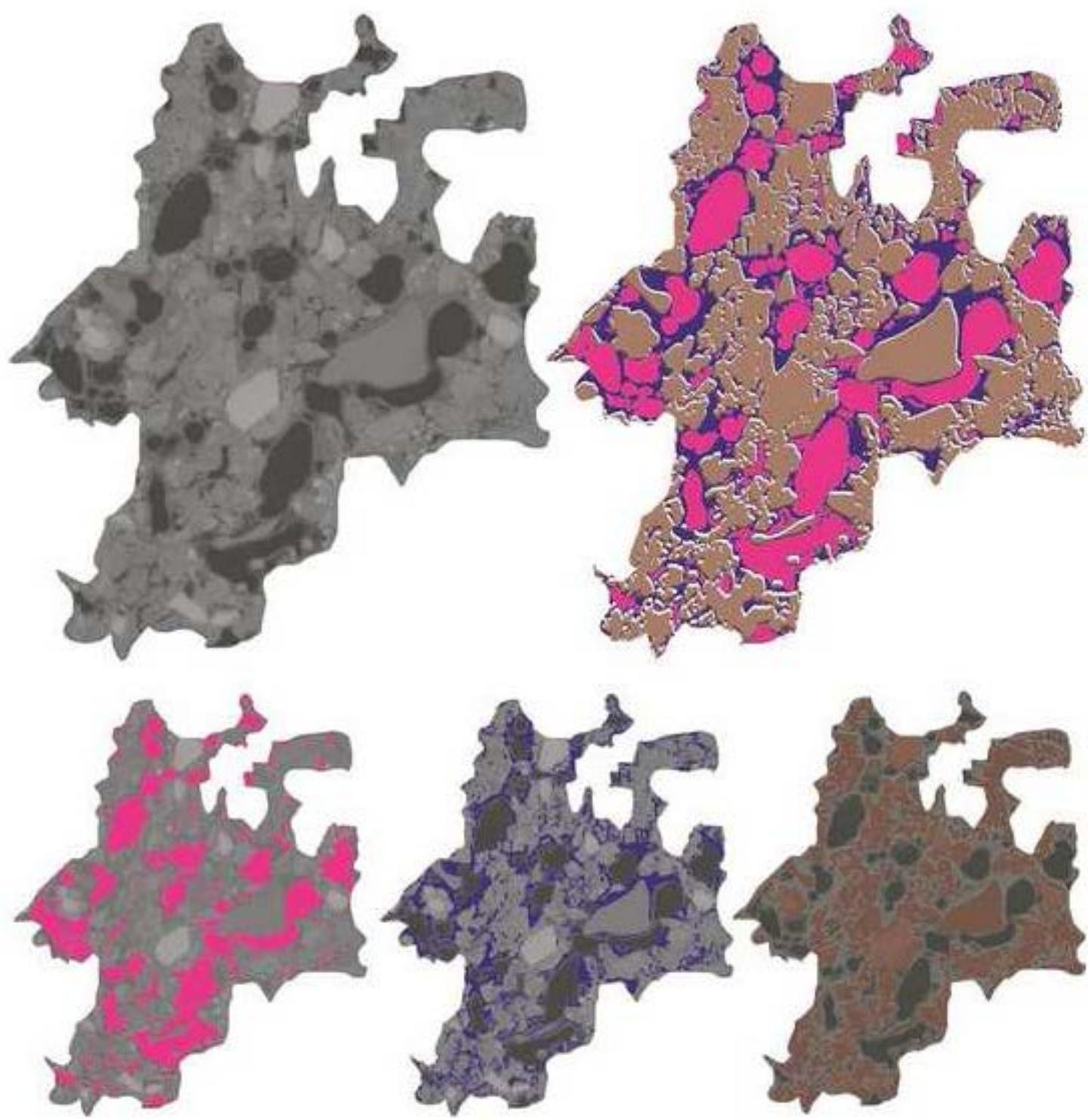


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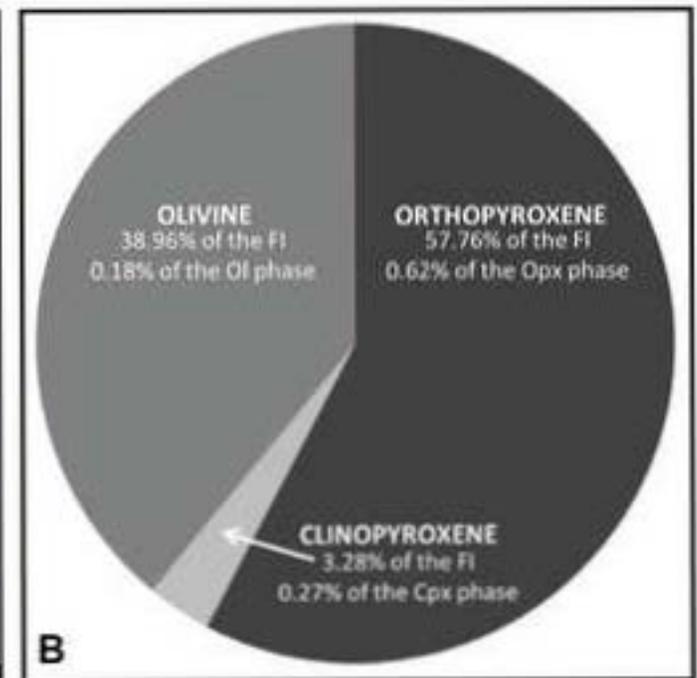
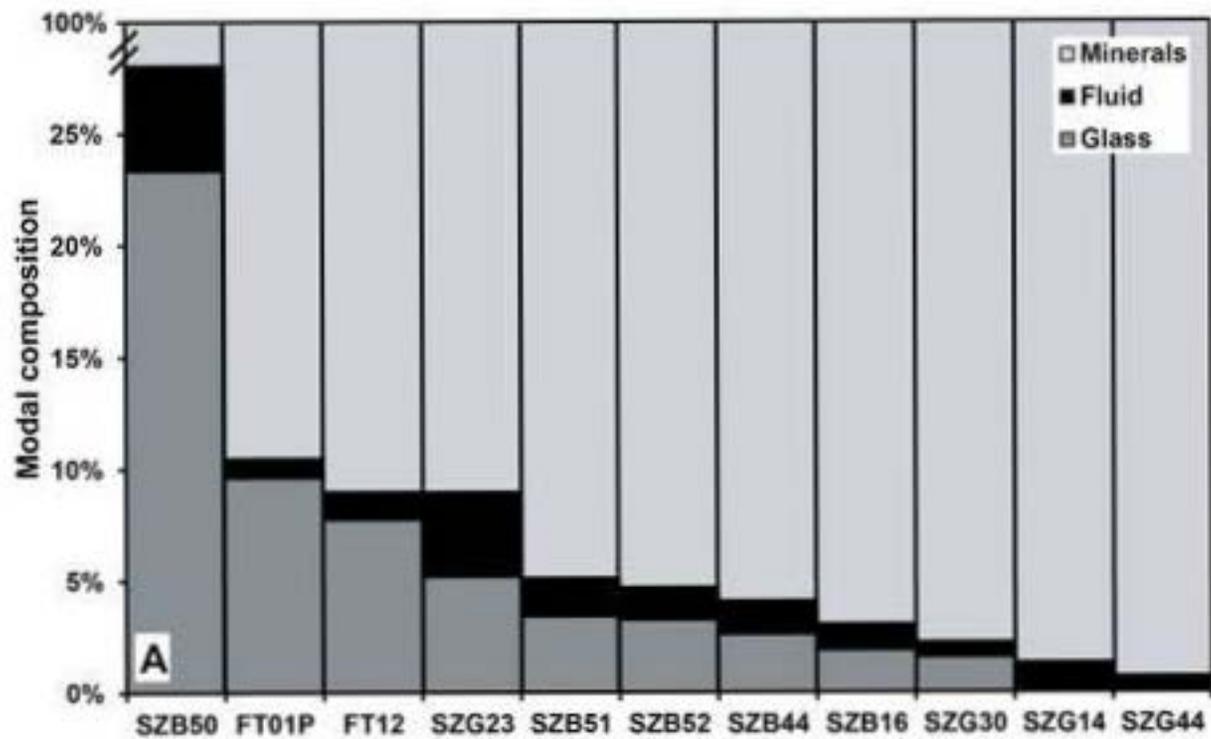


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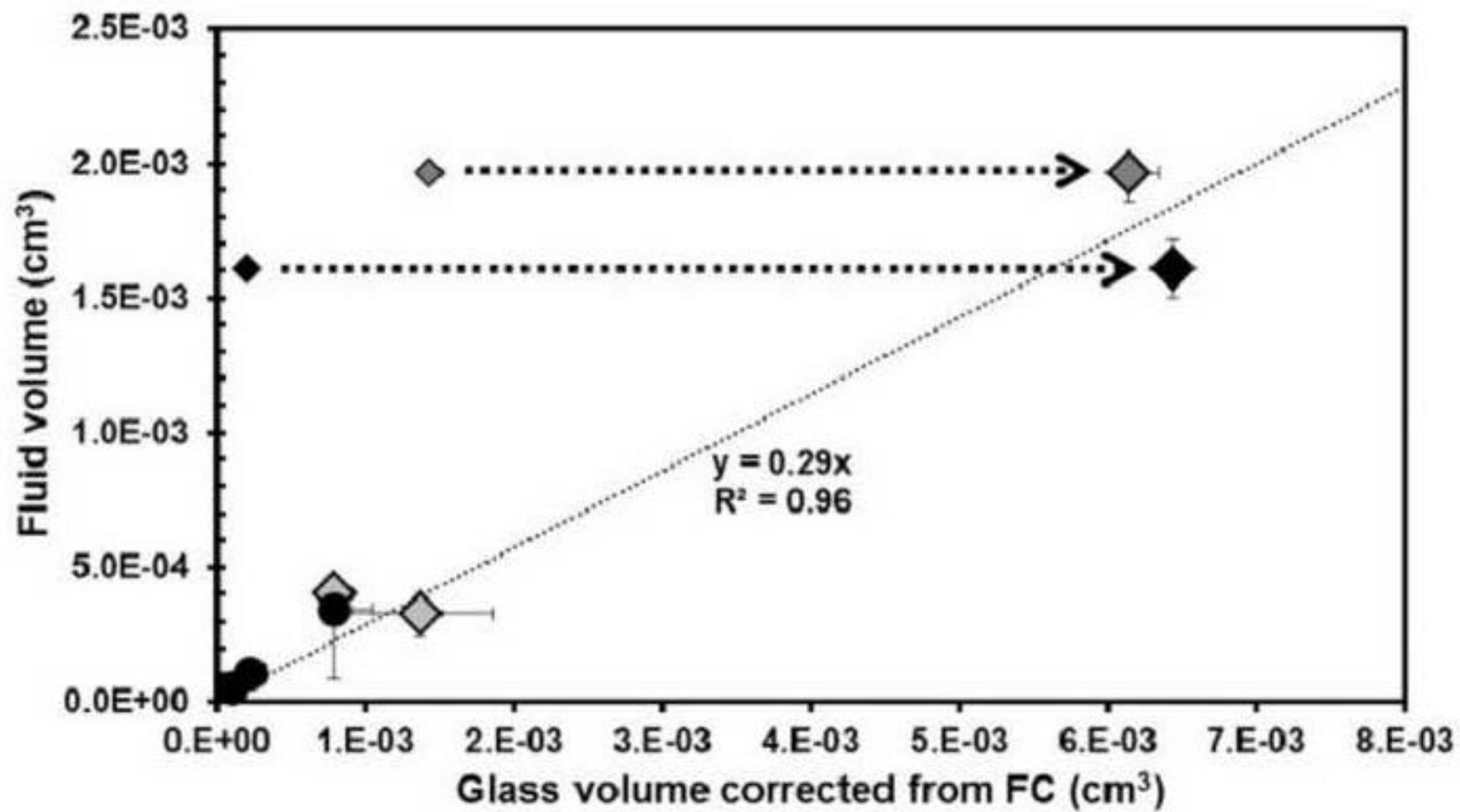


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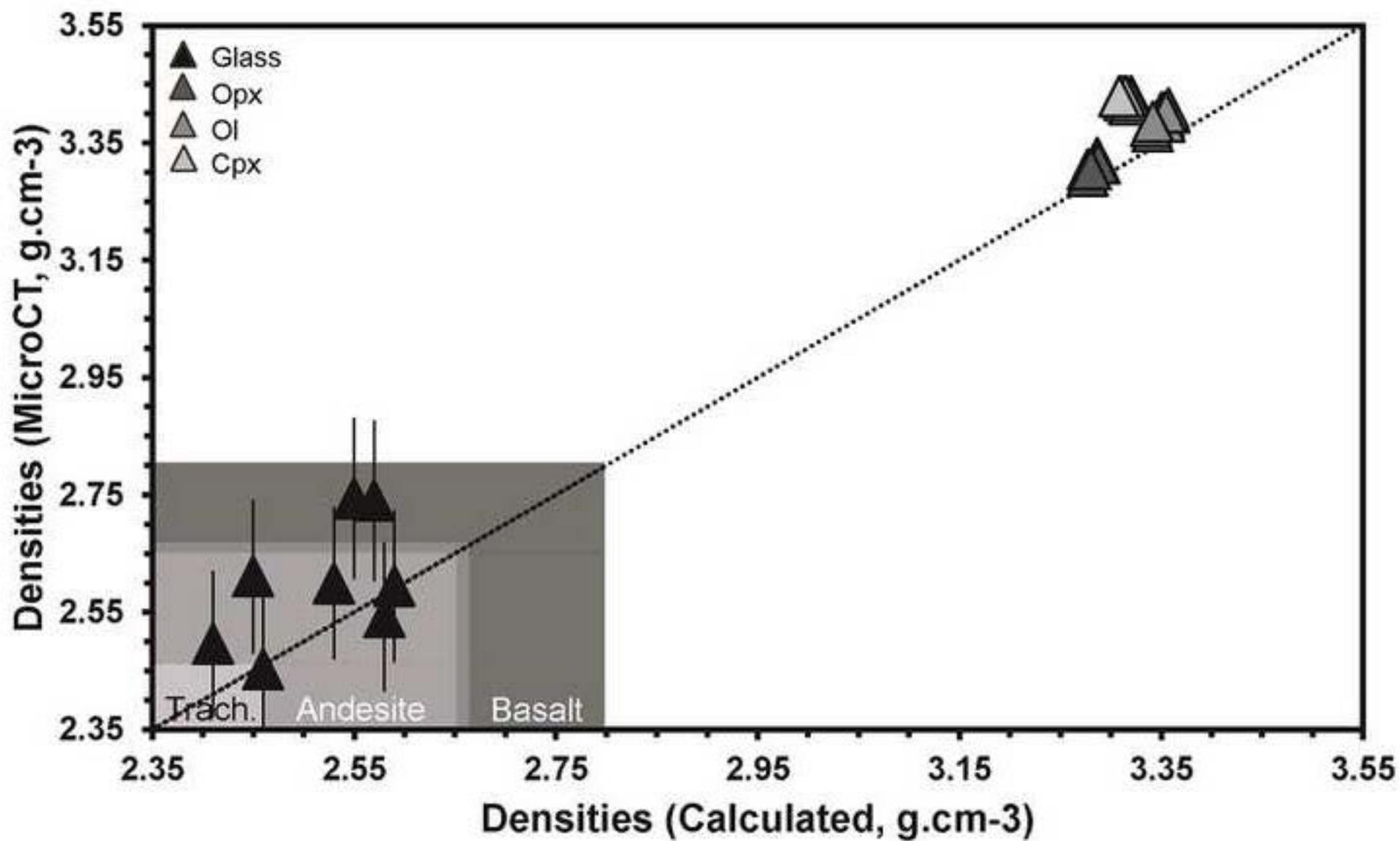


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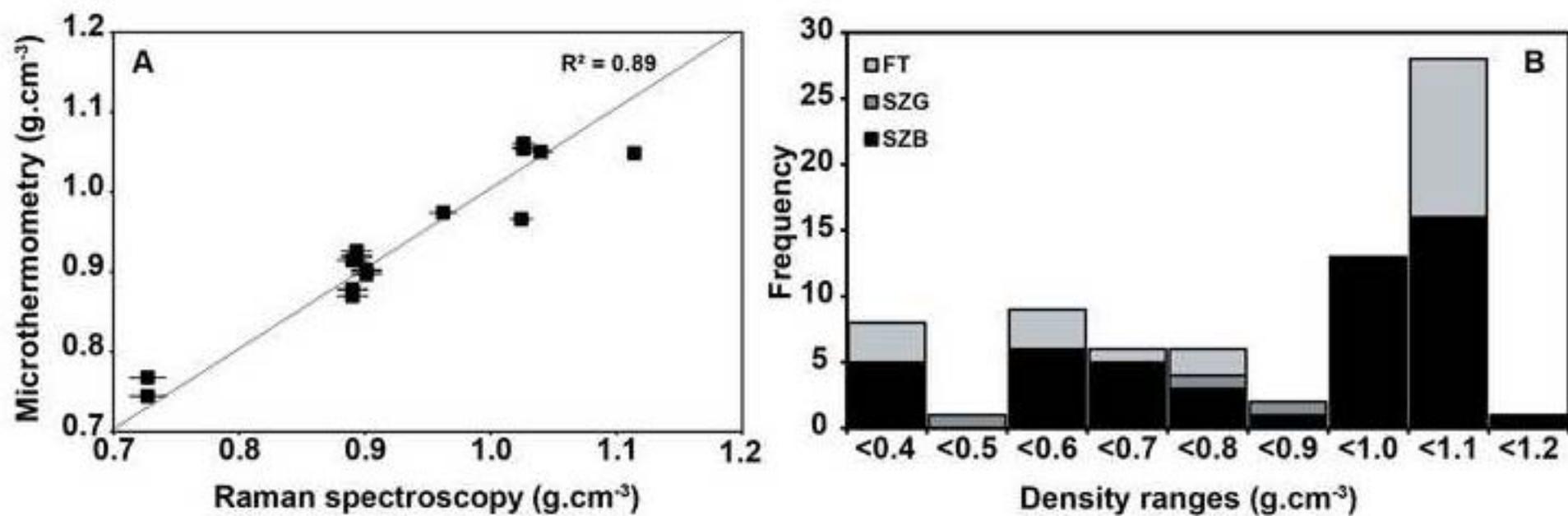


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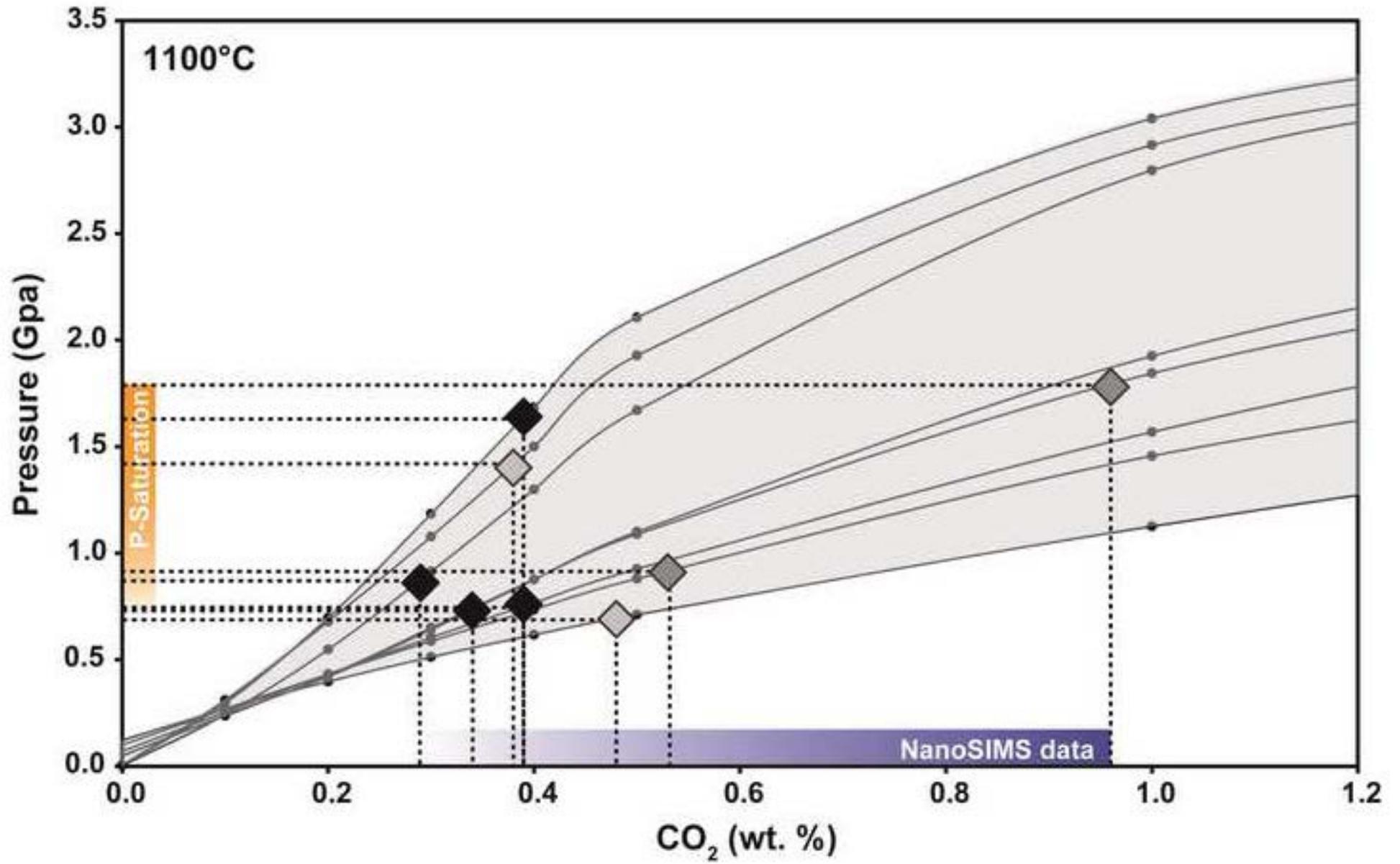
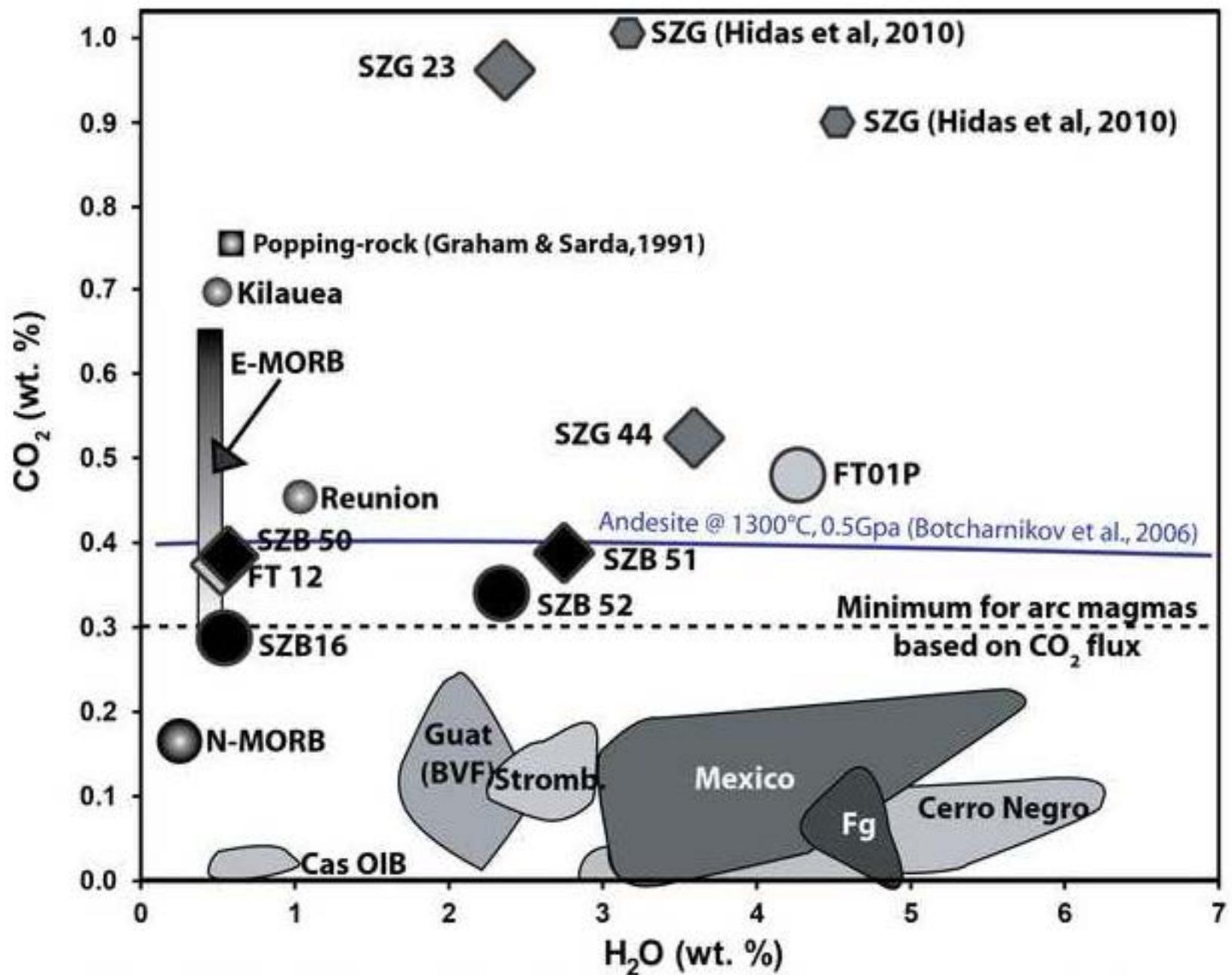


Figure 14

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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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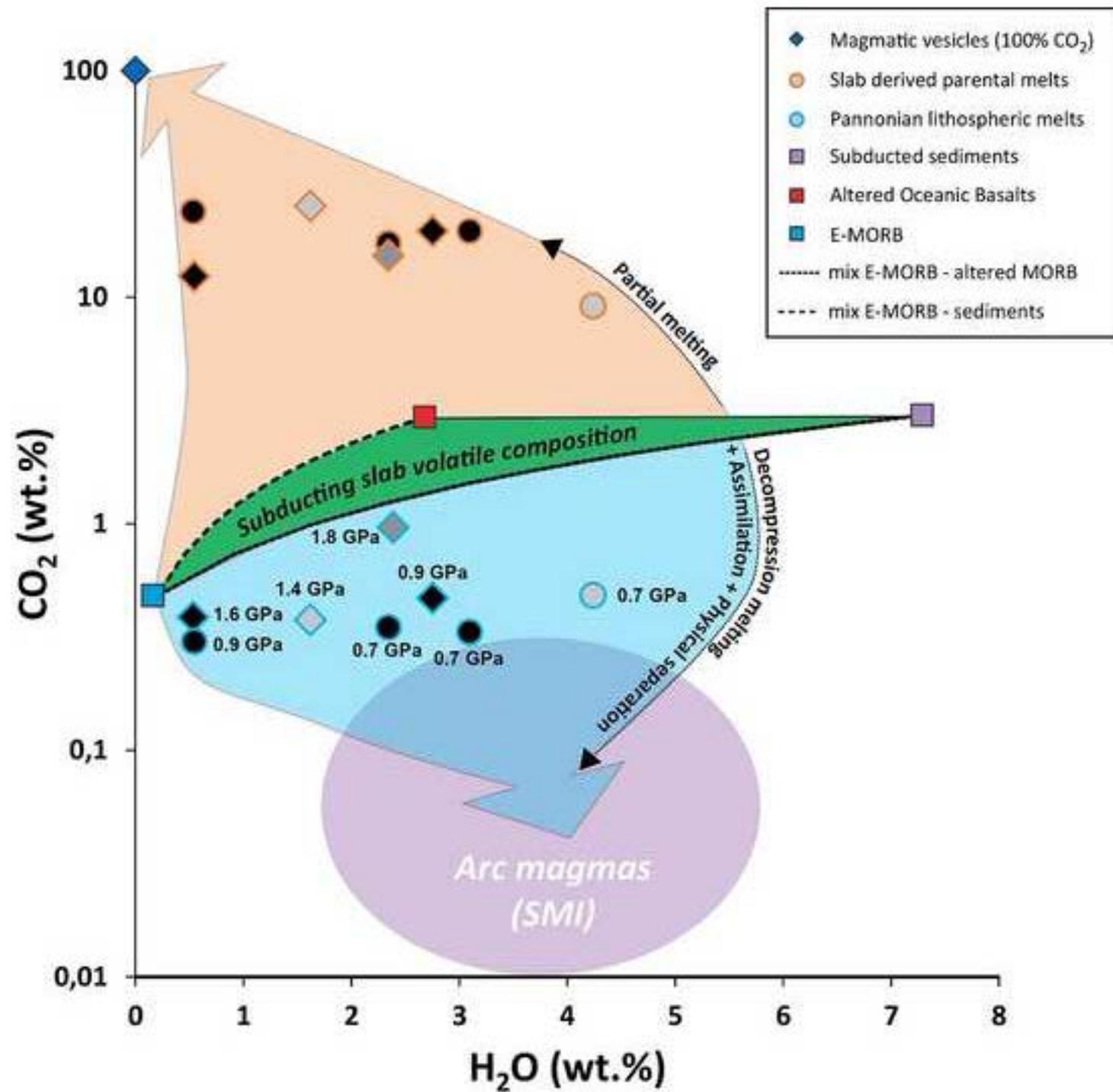


Table 1

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							Vol. (cm <sup>3</sup> )	Glass volume (cm <sup>3</sup> )			Fluid volume (cm <sup>3</sup> )			Density (g/cm <sup>3</sup> )			Mass (g)					
Sample	MP	SiO <sub>2</sub>	H <sub>2</sub> O	Error (1σ)	CO <sub>2</sub>	Error (1σ)	Minerals	Best estimate	Min (%)	Max (%)	Best estimate	Min (%)	Max (%)	Glass	Vesicles	FI (Max)	Glass	CO <sub>2</sub> (vesicle)	CO <sub>2</sub> (dissolved)	CO <sub>2</sub> /glass (wt. %)	CO <sub>2</sub> /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