

1 **SUPPLEMENTARY INFORMATION**

2 **METHODS**

3           Table DR1 contains the major, trace and volatile element compositions of 205  
4 melt inclusions from NAL709, a quickly cooled tephra sample collected from the  
5 eruption vent of Borgarhraun, a monogenetic post-glacial eruption located in the  
6 Theistareykir segment of northern Iceland (Lat. 65.8234°, Long. -16.8665°). Major  
7 element compositions of melt inclusions and host olivines were determined by electron  
8 probe microanalysis (EPMA) at the Geophysical Laboratory, Carnegie Institution, using  
9 the following conditions: 15 kV accelerating voltage, 30 nA beam, spot mode (for the  
10 olivine) or beam defocused to 10 µm diameter (for the glasses). We processed the olivine  
11 analyses for matrix correction using the set of absorption coefficients from Pouchon and  
12 Pichoir (1991). Combined accuracy and precision is 2% for the major elements (all  
13 quoted uncertainties are 2-sigma); MnO was determined by laser ablation inductively  
14 coupled mass spectrometry (see below).

15           Volatiles (CO<sub>2</sub>, H<sub>2</sub>O, F, S, Cl) were determined by SIMS using methods described  
16 in Hauri et al. (2002, 2006) with the modification that <sup>16</sup>OH is collected for H<sub>2</sub>O  
17 abundances rather than <sup>1</sup>H; total uncertainty (precision + accuracy) is ±7% for all  
18 volatiles. Selected trace elements (Li, Be, B, P, K, Sc, Ti, Cr, Sr, Y, Zr, Nb, Ba, La, Ce,  
19 Nd, Sm, Eu, Gd, Dy, Er, Yb, Hf) were determined by SIMS using a 10 nA beam of O<sup>-</sup>  
20 (10–15 µm diameter) and detection of positive secondary ions with a nominal

21 acceleration voltage of + 10 kV. Energy filtering was employed ( $-135 \pm 50$  eV) as well  
22 as a moderate mass resolving power sufficient to resolve  $^{27}\text{Al}^{3+}$  from  $^9\text{Be}$ ; calibration was  
23 performed in every session using the MPI-DING glasses KL2g, ML3Bg, GOR128g,  
24 GOR132g and BM90-21g; in addition we analyzed the in-house MORB glass standard  
25 ALV519-4-1 as a monitor of reproducibility. Trace element detection limits were  
26 measured using Herasil glass, and are very low ( $\sim 50$  ppb for Sr and Ba;  $\sim 20$  ppb for Hf  
27 and REE;  $\sim 5$  ppb for Nb). Combined accuracy and precision is 10% for all the trace  
28 elements reported.

29 Selected trace elements that can be difficult to measure accurately by SIMS (V,  
30 Mn, Co, Ni, Cu, Zn, Rb, Cs, Pb, Th, U), as well as several elements overlapping with  
31 SIMS (Nb, Ba, Hf) were determined by laser-ablation inductively coupled plasma mass  
32 spectrometry (LA-ICPMS) using a Photon Machines 193nm excimer laser and a Thermo  
33 iCapQ ICP-MS. A 35  $\mu\text{m}$  diameter beam was used to ablate material into a pure He gas  
34 stream (0.6 liters/min) injected into a pure Ar gas stream (1.4 liters/min) at a rate of 40  
35 Hz for a total of 400 shots (ablation rate 0.1  $\mu\text{m}/\text{shot}$ ). Calibration was performed in  
36 every session using the MPI-DING glasses (KL2g, ML3Bg, GOR128g, GOR132g and  
37 BM90-21g) and USGS glasses (BCR-2g, BHVO-2g, BIR-1g); in addition we analyzed  
38 the in-house MORB glass standard VE-32 as a monitor of reproducibility. Detection  
39 limits were in the single parts-per-billion range except for Sc which has an isobaric  
40 interference from SiO; ablation of pure forsterite (0.07 ppm Sc) was used to correct the  
41 SiO interference on Sc. Combined accuracy and precision is 6% or better for these trace  
42 elements; SIMS and LA-ICPMS data agree to within 8% for Nb, 10% for Ba and 15% for  
43 Hf.

44 For melt inclusions that contained a vapor bubble, bubble diameters were either  
45 measured by SEM when exposed at the surface, or by optical microscopy ( $\pm 10\text{-}20\%$ )  
46 using a calibrated scale; melt inclusion axes were measured with an SEM with an  
47 uncertainty of  $\pm 3\%$ . We estimated the amount of vapor and the moles of  $\text{H}_2\text{O}$  and  $\text{CO}_2$  in  
48 the shrinkage bubbles using the ideal gas law (IGL,  $n = PV/RT$ ) where  $n$  is the total moles  
49 of gas in the shrinkage bubble. For the IGL calculation, our pressure estimate ( $P$ ) and  
50 molar proportions of  $\text{H}_2\text{O}$  and  $\text{CO}_2$  come from the vapor saturation pressure determined  
51 from the  $\text{H}_2\text{O}$  and  $\text{CO}_2$  contents measured on the melt inclusion glass applied to the vapor  
52 saturation model of Dixon et al. (1995). We used a closure temperature of  $900^\circ\text{C}$  for  
53  $\text{H}_2\text{O}$ - $\text{CO}_2$  exchange in these rapidly-cooled inclusions, and the melt inclusion volume was  
54 calculated from the equation for the volume of an ellipsoid, using the measured long and  
55 short axes of the melt inclusion and assuming the two short axes of the ellipsoid are  
56 equal. From these constraints, we calculated the masses of  $\text{H}_2\text{O}$  and  $\text{CO}_2$  in each  
57 shrinkage bubble, and added these masses back into the composition of the melt  
58 inclusion, thereby calculating a bulk inclusion  $\text{H}_2\text{O}$  and  $\text{CO}_2$  content for each melt  
59 inclusion that contained a shrinkage bubble. In these calculations, the amount of  $\text{H}_2\text{O}$  in  
60 the shrinkage bubble was vanishingly small, and  $\text{CO}_2$  contained in the shrinkage bubble  
61 never exceeded 50% of the bulk  $\text{CO}_2$  content.

62 Table DR2 contains major, volatile and trace element abundances (and radiogenic  
63 isotope data where it exists) for mid-ocean ridge basalts containing  $\text{CO}_2$  and Nb data on  
64 the same sample, as recorded in the PetDB petrological database (161 samples). These  
65 samples were chosen by ranking the samples by  $\text{CO}_2/\text{Nb}$  data and selecting the samples  
66 in the upper 90<sup>th</sup> percentile, excluding samples from transform fault systems (the

67 Siqueiros data of Saal et al. (2002) and QDG data of Shimizu et al. (2016) are considered  
68 separately). The data set was filtered to include only samples with CO<sub>2</sub> determined by  
69 either SIMS or FTIR, and contains all of the vapor-undersaturated MORB samples (5  
70 samples) described in Michael and Graham (2015) which are not from transform fault  
71 regions or from the study of Shimizu et al. (2016). **In this group of 15 samples, we did  
72 not use the pressure of vapor saturation to evaluate whether or not they were  
73 undersaturated in CO<sub>2</sub>. Instead we relied on their correlated abundances of CO<sub>2</sub> and Nb-  
74 Rb-Ba as an indication that the samples did not degas CO<sub>2</sub>. There exist several models  
75 based on experimental data that calibrate vapor saturation pressure with CO<sub>2</sub>+H<sub>2</sub>O  
76 contents in mafic melts (Holloway & Blank, 1994; Dixon & Stolper, 1995; Papale, 1999;  
77 Newman & Lowenstern, 2002; Papale et al., 2006; Iacono-Marziano et al., 2012; Ghiorso  
78 & Gualda, 2015), and they all give somewhat different vapor saturation pressures, thus  
79 we have not relied on estimated vapor saturation pressures to determine whether samples  
80 have lost CO<sub>2</sub> by degassing.**

81 To determine the CO<sub>2</sub>/Nb ratio of the melt inclusion population, we calculated the  
82 CO<sub>2</sub>/Nb ratio of each melt inclusion and then calculated the simple arithmetic mean of  
83 the population. The same method was used to calculate the CO<sub>2</sub>/Nb ratio of melt  
84 inclusion populations from the Siqueiros Fracture Zone (Saal et al., 2002) and the  
85 equatorial mid-Atlantic Ridge (Le Voyer et al. 2016), and the vapor-undersaturated  
86 MORB sample suites described by Cartigny et al. (2008) and Shimizu et al. (2016), and  
87 the top 10% MORB samples described above. Uncertainties on the CO<sub>2</sub>/ITE ratios of the  
88 data populations were calculated as 2 standard errors of the population.

## 89 **COMPARISON WITH PUBLISHED DATA**

90 Studies reporting CO<sub>2</sub> abundances in submarine glasses and melt inclusions  
91 normally display large scatter due to heterogeneous distributions of CO<sub>2</sub>-rich bubbles  
92 (vesicles) within submarine samples, and the presence of shrinkage bubbles in melt  
93 inclusions whose presence is not typically accounted for (Moore et al., 2015). As a result,  
94 for the global MORB database as a whole, neither submarine MORB glasses nor most  
95 MORB melt inclusions display correlations of CO<sub>2</sub> with non-volatile trace elements (Fig.  
96 4), although certain samples plot at CO<sub>2</sub>/ITE ratios similar to the Borgarhraun melt  
97 inclusions.

98 Several prior studies have focused attention on the highest CO<sub>2</sub>/Nb ratios among a  
99 group of melt inclusions, arguing that melt inclusions with the highest CO<sub>2</sub>/Nb most  
100 closely approach the composition of the undegassed magma, and that lower CO<sub>2</sub>/Nb  
101 ratios are produced by degassing of CO<sub>2</sub> during magma ascent from depth (Wanless and  
102 Shaw, 2012; Wanless et al., 2014; Rosenthal et al., 2015). This approach is problematic;  
103 the highest CO<sub>2</sub>/Nb ratio among the Borgarhraun melt inclusions (961) occurs in a spinel-  
104 hosted inclusion corrected for the presence of a shrinkage bubble, and is more than twice  
105 the average CO<sub>2</sub>/Nb of the population. The highest bubble-free melt inclusion from  
106 Borgarhraun has a CO<sub>2</sub>/Nb of 598, which is still 50% higher than the population average.  
107 Further insight into the origin of the data scatter is obtained from the systematics of the  
108 non-volatile trace elements; (Slater et al., 2001; MacLennan et al., 2003) demonstrated  
109 that the rare-earth element (REE) patterns of many populations of Iceland melt inclusions  
110 could be accurately fit by invoking incomplete mixing of polybaric near-fractional melts  
111 derived from adiabatic upwelling of a column of mantle. In Figure 3 we have tested this  
112 model for the CO<sub>2</sub>-Nb variation, using appropriate C and Nb partition coefficients

113 between mantle minerals and silicate melts (Rosenthal et al., 2015). We used pHMELTS  
114 (Asimow et al., 2001) (Smith and Asimow, 2005) to model the 1D adiabatic melting  
115 process, with a mantle potential temperature of 1410°C (corresponding to 1450°C at 3  
116 GPa), and a residual melt porosity of 1.5% which serves as a threshold above which melt  
117 ascent and mixing becomes possible. In this model melting begins at 3.0 GPa and ceases  
118 at 0.85 GPa, a pressure that corresponds approximately to the base of the crust beneath  
119 northern Iceland, yielding an overall degree of melting of 17%. For a mantle source with  
120 75.4 ppm CO<sub>2</sub> and 0.193 ppm Nb, this melting model produces a curved trajectory of  
121 incremental melt compositions that skirts the lower bound of the CO<sub>2</sub>-Nb data, while  
122 families of linear mixing trajectories among the incremental melt compositions explains  
123 well the scatter of the Borgarhraun CO<sub>2</sub>-Nb data (Fig. 3).

124 We illustrate this point to emphasize that erroneous estimates of the CO<sub>2</sub>/Nb of  
125 mantle sources can arise when focusing on the highest CO<sub>2</sub>/Nb ratio among a population  
126 of melt inclusions. While such variations can be due to degassing (Wanless and Shaw,  
127 2012; Wanless et al., 2014), they can also result as a normal part of the range in CO<sub>2</sub>/Nb  
128 ratios of incremental melts produced during near-fractional melting due to the slight  
129 difference melt-solid partition coefficients of CO<sub>2</sub> and Nb, and it is these near-fractional  
130 incremental melt compositions (and their mixtures) that are often preserved in melt  
131 inclusions hosted by primitive high-Mg# phenocrysts. The best approach to estimating  
132 the CO<sub>2</sub>/Nb ratio of the mantle source is to first account for CO<sub>2</sub> partitioning into melt  
133 inclusion shrinkage bubbles, then to look for a correlation of CO<sub>2</sub> with Nb or other non-  
134 volatile trace elements. Well-correlated CO<sub>2</sub> and Nb indicate the absence of degassing  
135 and thus a record of the mantle source ratio, but if no correlation is found then it can be

136 reasonably assumed that the melt had lost CO<sub>2</sub> via degassing prior to inclusion  
137 entrapment. As a result, for a population of degassed melt inclusions it is not possible to  
138 recover the pre-degassing CO<sub>2</sub>/Nb ratio with any degree of precision.

### 139 **AVERAGE AND LOCAL MANTLE CO<sub>2</sub> ABUNDANCES**

140 Stracke et al. (2003) observed that whole rock samples from the Borgarhraun flow  
141 were isotopically heterogeneous, and Maclennan et al. (2003) used major and trace  
142 element compositions of melt inclusions from Borgarhraun to demonstrate that the array  
143 of melt compositions present in the Borgarhraun mantle came from the same distribution  
144 of melt compositions that represent the Theistareykir segment as a whole. McKenzie et  
145 al. (2004) demonstrated that isotopic variations correlate with the concentrations of  
146 incompatible trace elements both in the Borgarhraun flow and in the Theistareykir  
147 segment, while Maclennan (2008) showed that correlated variations in Pb isotopes and  
148 trace elements in melt inclusions from the Reykjanes Peninsula demonstrate a dominant  
149 role for mixing of melts from heterogeneous sources in generating the chemical and  
150 isotopic variability of Icelandic basalts. Thus some of the small variability in CO<sub>2</sub>/Nb  
151 ratios may represent melt mixing superimposed upon populations of near-fractional melts  
152 from a heterogeneous mantle, and if so then it is probable that the CO<sub>2</sub>/Nb ratio of the  
153 Borgarhraun melt inclusion population is dominated by melts from the more enriched  
154 mantle components beneath the Theistareykir segment.

155 We use all four non-volatile trace elements (Th, Nb, Rb, Ba) to provide multiple  
156 estimates of mantle source CO<sub>2</sub> content. The CO<sub>2</sub> and ITE contents observed in the  
157 Borgarhraun melt inclusions span half of the range displayed by MORB, between the  
158 depleted Siqueiros magmas (CO<sub>2</sub>/Nb = 230) and the enriched magmas of the 14°N MAR

159 region ( $\text{CO}_2/\text{Nb} = 534$ ). Given that the six sample suites examined here span the entire  
160 range of trace element depletion and enrichment observed in MORB, we conclude that  
161 upper-mantle  $\text{CO}_2/\text{Nb}$  ratios at other MOR segments are unlikely to vary significantly  
162 outside this range (factor of 2.4). Assuming a global average  $\text{CO}_2/\text{Nb}$  ratio of 435 (Table  
163 1, ALL-MORB), with a melt production rate of  $21 \pm 3 \text{ km}^3/\text{yr}$  (Crisp, 1984), crustal  
164 density of  $2700 \text{ kg/m}^3$ , and an average MORB Nb content of  $3.62 \pm 0.36 \text{ ppm}$  (Gale et al.,  
165 2013), the global average Nb flux across the crust-mantle boundary at mid-ocean ridges  
166 amounts to  $2.23 \pm 0.36 \times 10^9 \text{ mol/yr}$ , and this translates into an average global  $\text{CO}_2$  flux of  
167  $2.03 \pm 0.36 \times 10^{12} \text{ mol/yr}$  for the present-day mid-ocean ridge system. This flux is within  
168 20% of that calculated by Michael and Graham (2015). The average  $\text{CO}_2$  flux estimated  
169 from Nb, Th, Rb and Ba fluxes at mid-ocean ridges is  $2.38 \pm 0.46 \times 10^{12} \text{ mol/yr}$  (Table 1).  
170 The average MOR  $\text{CO}_2$  flux thus calculated is at the low end of the range of most  
171 published estimates, but within the ranges estimated by Cartigny et al. (2008) and Burton  
172 et al. (2013). With an average  $\text{CO}_2/{}^3\text{He}$  molar ratio of  $2.5 \pm 1.0 \times 10^9$  (Marty and Tolstikhin,  
173 1998), this constrains the average MORB  ${}^3\text{He}$  flux to be  $953 \pm 325 \text{ mol/yr}$ .

174 This global average of course does not apply directly to specific geographic areas  
175 such as Borgarfraun and the other locales in Table 1, as trace element compositions and  
176 radiogenic isotope ratios are known to vary significantly along the global mid-ocean  
177 ridge system (Jenner and O'Neill, 2012; Gale et al., 2013; Kelley et al., 2013); thus  
178 changes in source composition and magma production rate will cause  $\text{CO}_2$  output from  
179 ridges to vary one locale to the next. In order to estimate local  $\text{CO}_2$  fluxes, we must have  
180 knowledge of the mantle Th-Nb-Rb-Ba abundances at each location. To estimate mantle  
181 ITE compositions we approximate the compositional and isotopic range in MORB as a

182 mixture between two depleted MORB mantle sources, one representing the depleted  
183 extreme of the MORB range (D-DMM) and the other at the enriched extreme (E-DMM)  
184 as described by Shimizu et al. (2016) but with the small adjustments made to the D-  
185 DMM component in the CO<sub>2</sub> (150 ppm), Rb (0.36 ppm), Ba (3.90) and Th (0.047)  
186 concentrations, in order to provide a best fit to the six MORB population averages (Fig.  
187 6). In this model, we use the Nd isotope composition of the sample to determine the  
188 extent of mixing between D-DMM and E-DMM, and from this mixture the trace element  
189 composition can be derived given appropriate D-DMM and E-DMM endmember  
190 compositions. For D-DMM we use the trace element estimate of Workman and Hart  
191 (2005) with epsilon-Nd = +10.5 and CO<sub>2</sub> = 150 ppm, but with small adjustments made in  
192 Rb (0.36 ppm), Ba (3.90) and Th (0.047) concentrations in order to provide a best fit to  
193 the six MORB population averages (Fig. 6). The Nd isotope composition of each sample  
194 or sample population thus determines the non-volatile ITE concentrations that are used to  
195 provide estimates for mantle source CO<sub>2</sub> based on CO<sub>2</sub>/ITE ratios. These estimates are  
196 compiled in Table 1, and demonstrate that mantle CO<sub>2</sub> abundances can vary by more than  
197 a factor of ten at the local scale. Although this model fails to predict the CO<sub>2</sub>/ITE ratios  
198 of the equatorial MAR melt inclusions of Le Voyer et al. (2016), we note that this MAR  
199 sample has the highest <sup>143</sup>Nd/<sup>144</sup>Nd known among all MORB and thus may have  
200 originated from an endmember more depleted, or with older time-integrated Sm/Nd, than  
201 the endmember Nd isotope composition chosen by Shimizu et al. (2016).

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204 Supplemental References

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261 **Supplemental Figures**

262 Figure DR1. (A) Transmitted light, and (B) reflected light photos of olivine-hosted melt  
263 inclusions from Borgarhraun. The olivine crystal is 3mm across the long axis; circular  
264 black depressions are 35  $\mu\text{m}$  diameter laser ablation craters.

265

266 Figure DR2.  $\text{CO}_2$  versus Nb for Borgarhraun melt inclusions (red filled circles) compared  
267 with near-fractional melts of a mantle composition with 75 ppm  $\text{CO}_2$  and 0.193 ppm Nb  
268 (red curve, see text). Blue line is a regression line through the melt inclusion data, black  
269 line is a mixing line between the most enriched and most depleted of the incremental  
270 melts. The data are bound by the compositions of incremental melts along the red curve,  
271 and families of mixing lines connecting enriched and depleted incremental melts. Inset  
272 shows the full range of incremental melt compositions.

273

274 Figure DR3. These high-precision MORB  $\text{CO}_2$  data sets shown with the global published  
275 data on olivine-hosted melt inclusions (panel B, PetDB and Georoc databases).

276

277 Figure DR4. (A) Average  $\text{CO}_2/\text{Rb}$  and  $\text{CO}_2/\text{Ba}$  ratios for the six mid-ocean ridge regions  
278 that exhibit  $\text{CO}_2$ -ITE correlations; Atlantic locales are shown in green, Pacific locales are  
279 shown in blue.  $\text{CO}_2/\text{Rb}$  and  $\text{CO}_2/\text{Ba}$  ratios are nearly homogeneous and show no  
280 distinctions between ocean basins. (B) Average  $\text{CO}_2/\text{Nb}$  and  $\text{CO}_2/\text{Th}$  ratios are  
281 heterogeneous and correlated, with Atlantic locales being 70-100% higher than Pacific  
282 locales.

283

284 Figure DR5. La/Sm and CO<sub>2</sub>/Nb ratios of individual samples and melt inclusions from  
285 the sample groups discussed in this study, group by Atlantic (14°N MAR, equatorial  
286 MAR, Iceland) and Pacific (Siqueiros, QGD, Top 10% MORB). Average CO<sub>2</sub>/Nb and  
287 La/Sm are higher in the Atlantic vs Pacific groups, but overall there is no correlation, and  
288 the full range of CO<sub>2</sub>/Nb ratio is expressed at intermediate La/Sm that is exhibited by  
289 both groups.

Figure DR1

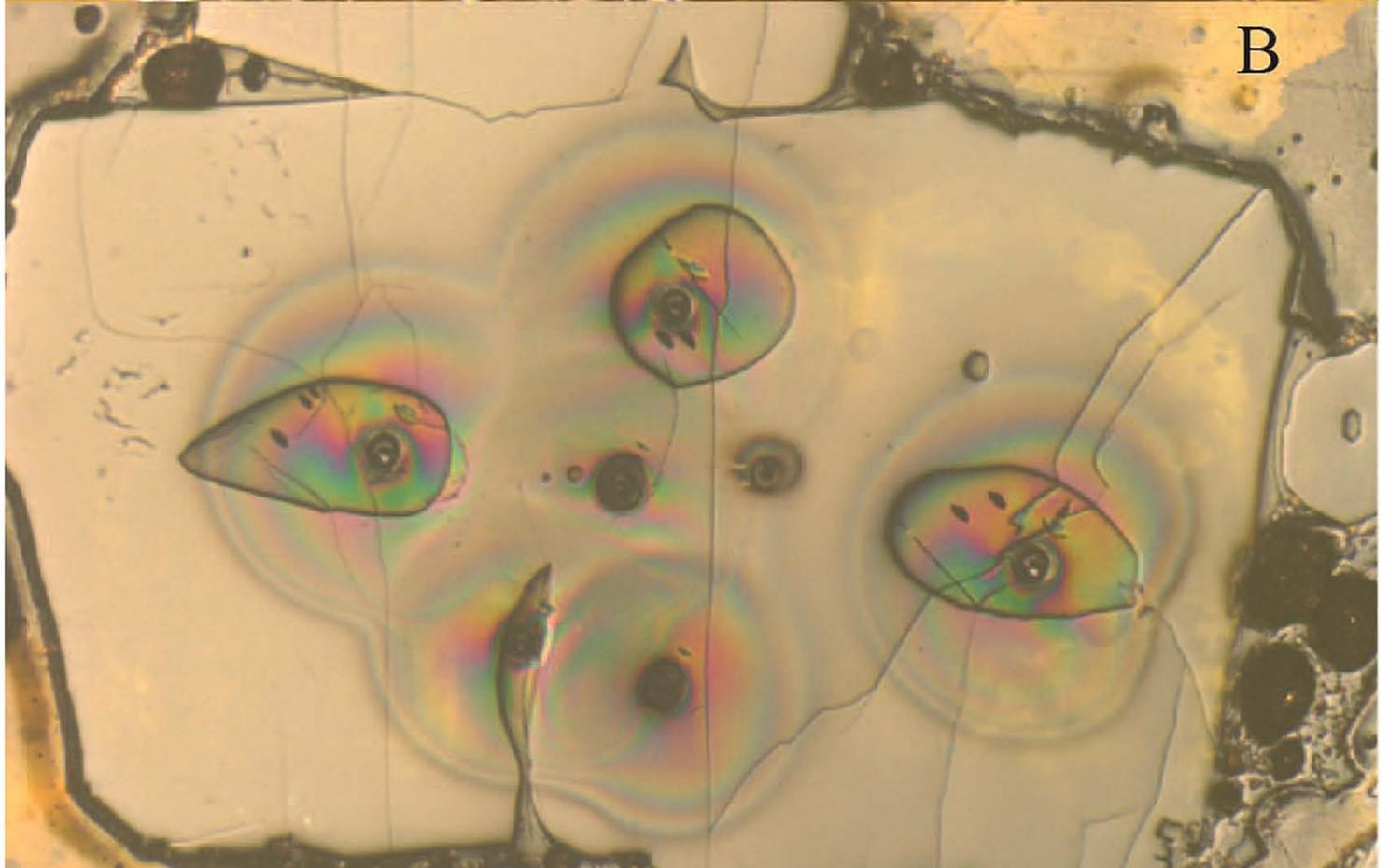
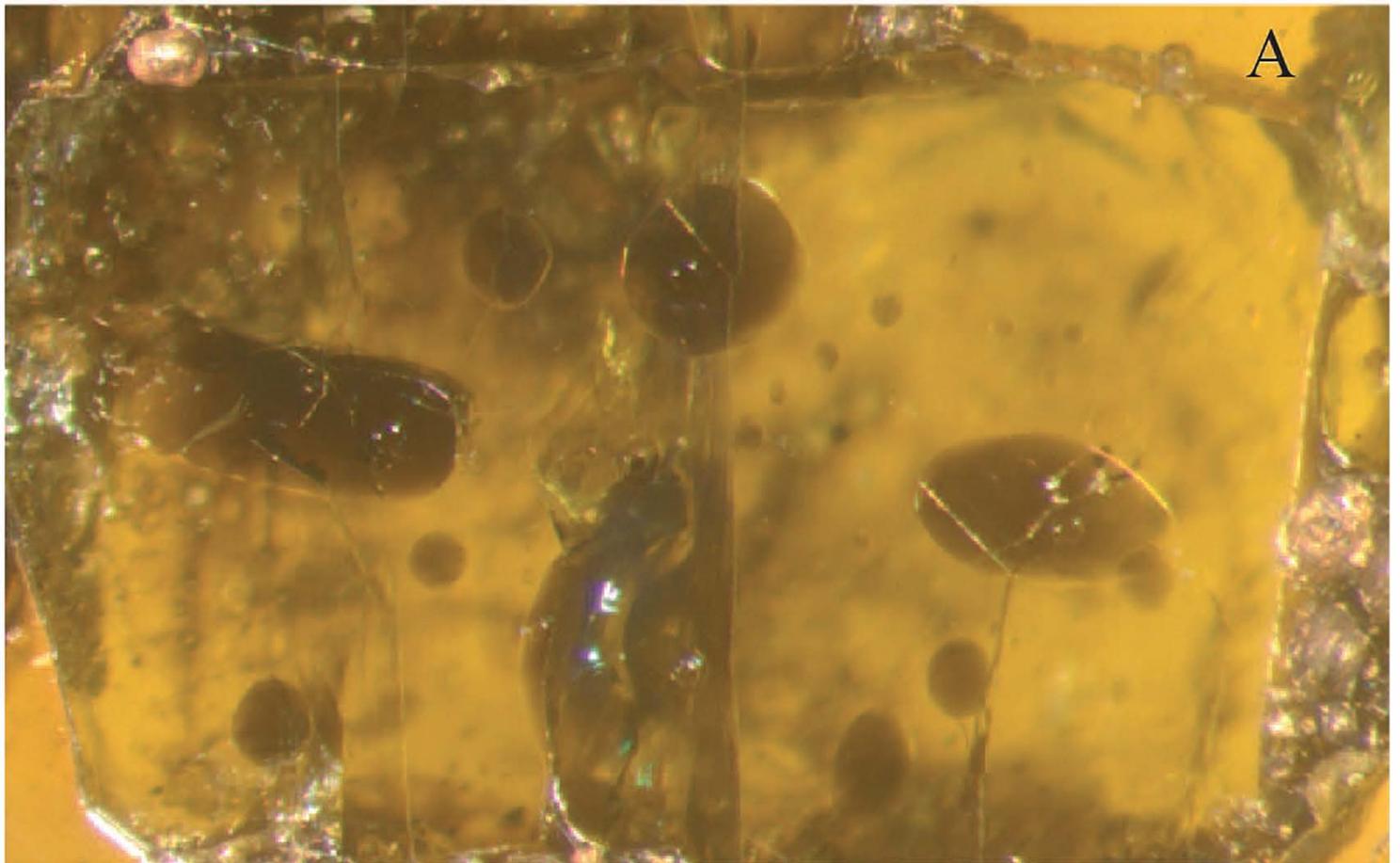


Figure DR2

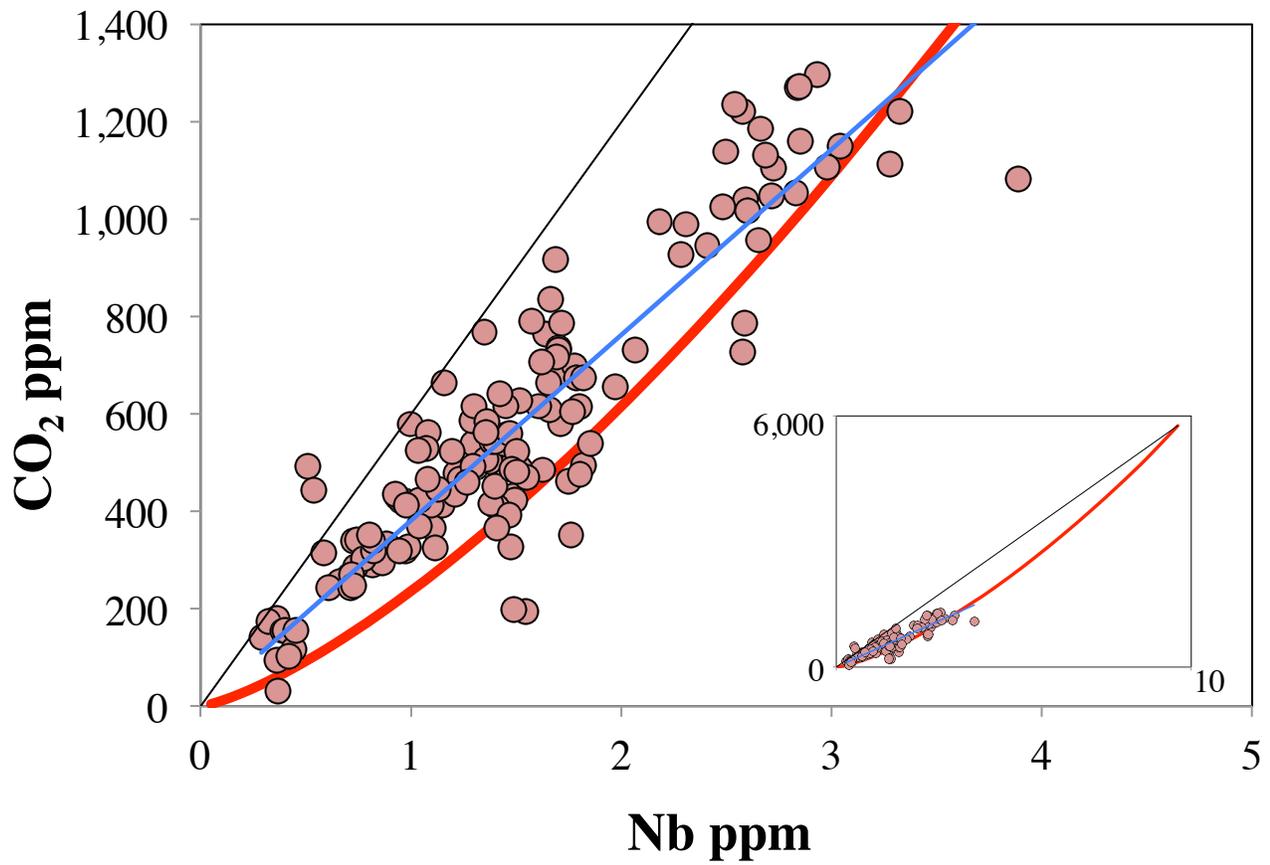


Figure DR3

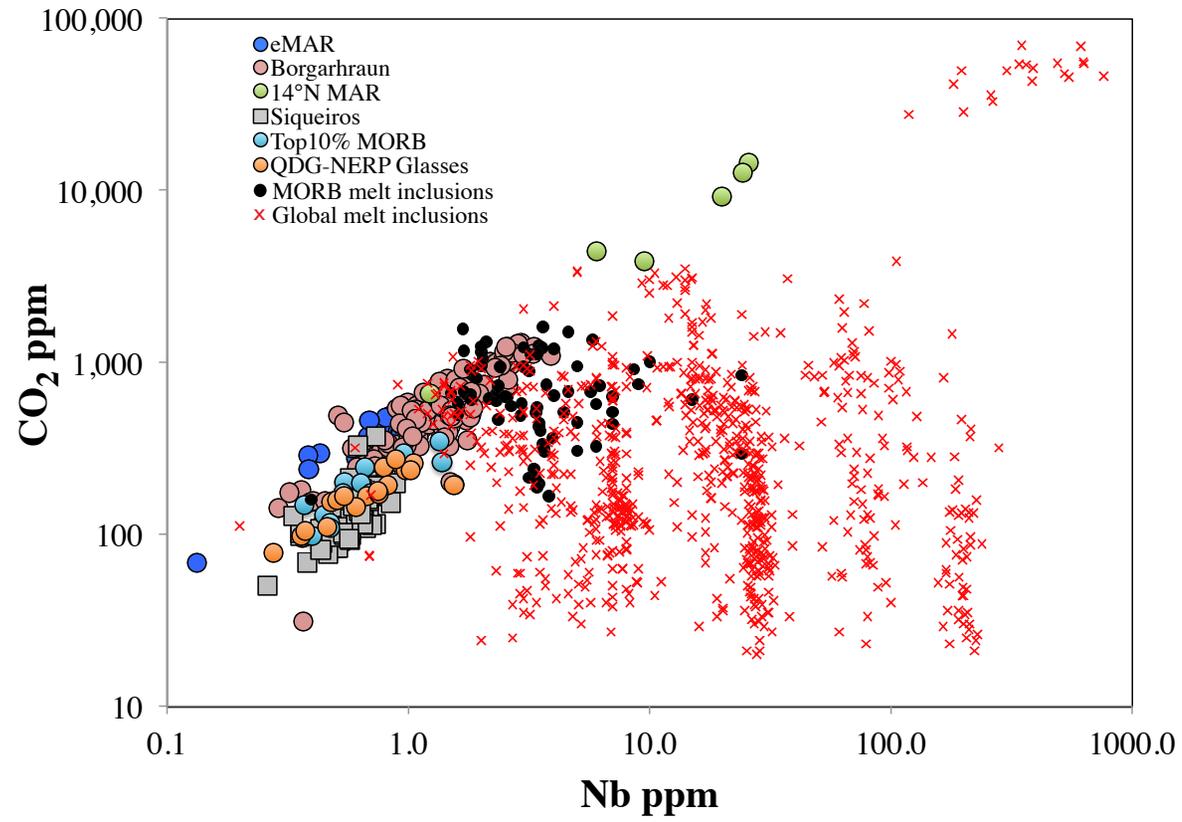


Figure DR4

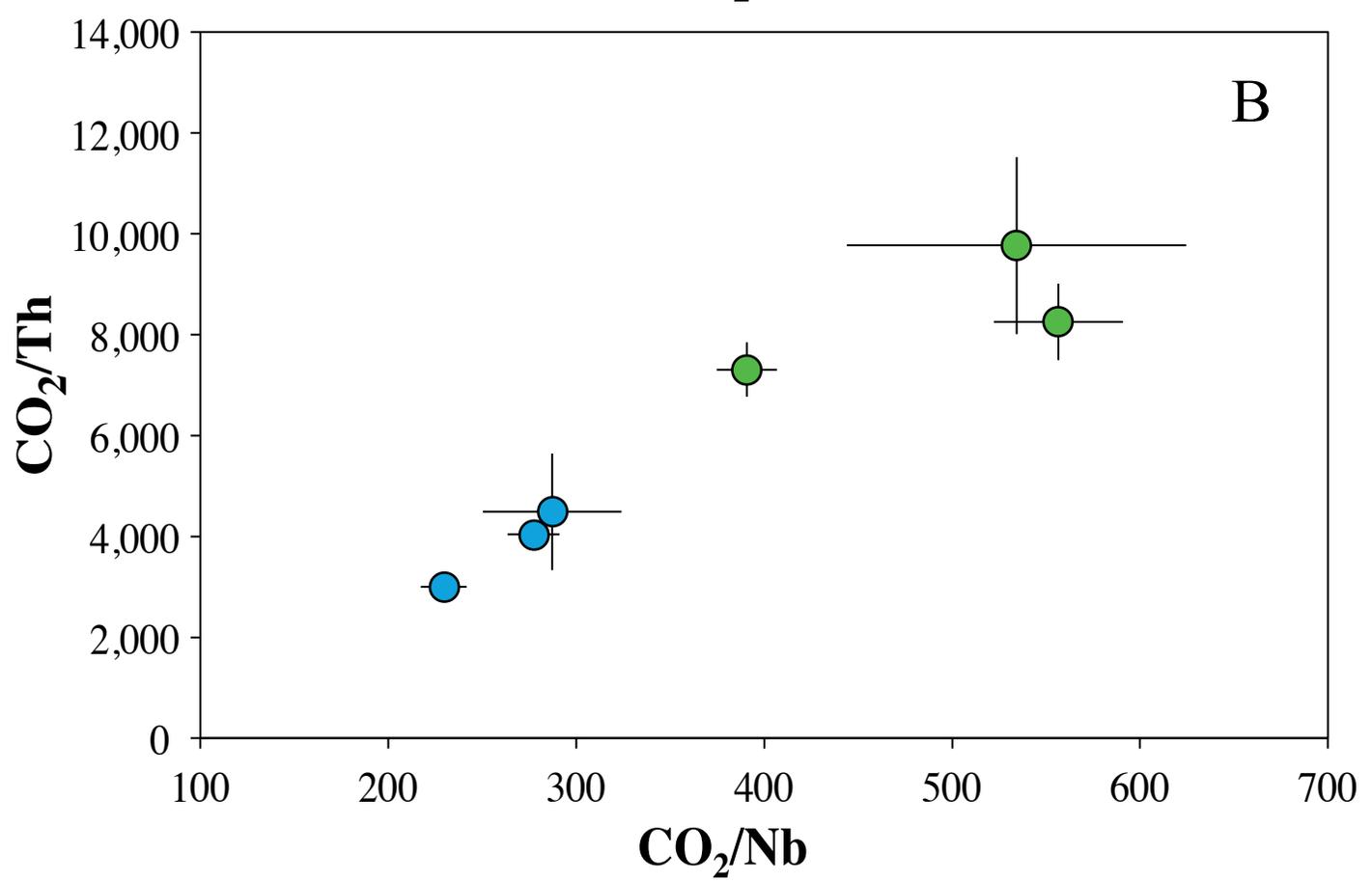
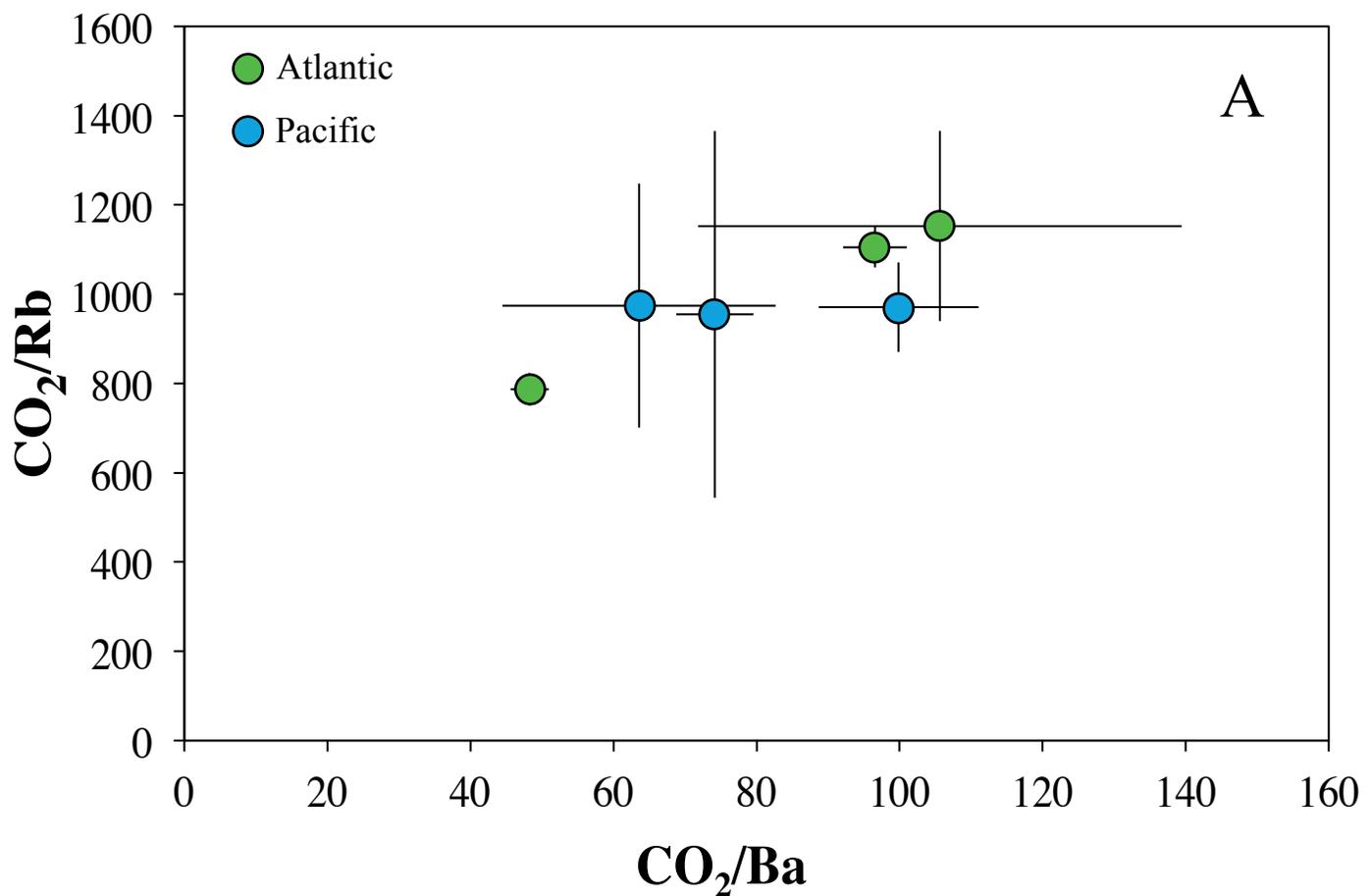


Figure DR5

