Carbon concentration increases with melting depth in Earth's upper mantle

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Carbon in the upper mantle controls incipient melting of carbonated peridotite and so acts as a critical driver of plate tectonics. The carbon-rich melts that form control the rate of volatile outflux from the Earth's interior, contributing to climate evolution over geological times. However, attempts to constrain the carbon concentrations of the mantle source beneath Oceanic Islands and Continental Rifts is complicated by pre-eruptive volatile loss from magmas. Here, we compile literature data on magmatic gases, as surface expression of the pre-eruptive volatile loss, from 12 Oceanic Island and continental Rift volcanoes. We find the levels of carbon enrichment in magmatic gases correlate with the trace element signatures of the corresponding volcanic rocks, implying a mantle source control. We use this global association to estimate that the mean carbon concentration in the upper mantle, down to 200 km depth, is approximately 350 ppm (range, 117-669 ppm). We interpret carbon mantle heterogeneities to reflect variable extents of mantle metasomatism from carbonated silicate melts. Finally, we find that the extent of carbon enrichment in the upper mantle positively correlates with the depth at which melting starts. Our results imply a major role of C in driving melt formation in the upper mantle.

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Measuring the C concentration in mantle-sourced magmas[1-3] (Fig. 1) is, together with melting

experiments[4-6] and the recovery of mantle-derived diamonds[7] and xenoliths[8], the main way to assess

the C-rich nature of the Earth's mantle. However, owing to limited solubility in silicate melts under

crustal conditions, erupted magmas are extensively depleted in C by degassing, to the point that

even the most deeply trapped melt inclusions (MIs) in crystals fail to record the initial (mantle-

inherited) C cargo[9]. Thus, while consensus exists that the majority of C on Earth is deeply stored[10-11], the mantle C budget remains a matter of debate[12-14].

To correct for pre- and post-entrapment gas loss to bubbles[15-16] and therefore estimate the initial C

content of parental melts and their mantle sources, geochemists pair CO_2 in MIs with incompatible

(and nonvolatile) trace elements[13,17] that behave similarly to C during partial melting, such Nb and

Ba (Fig. 1b-c). Reconstructing the mantle CO_2/Ba ratio[2,13] (Fig. 1c) from analysis of rare

undegassed MIs[2,18] and glasses[19] in mid-ocean ridge basalts (MORBs) has constrained the average

C content of the depleted MORB mantle (DMM) at ~20-56 ppm[1,20]. However, the wide inter-ridge

variability[20] (range 3-540 ppm C) implies an heterogeneous[2] DMM, with the highest C contents in

MOR segments near active hot spots[20] suggesting a more C-rich deeper mantle.

The C content of deeper regions of the convecting mantle can potentially be traced by plume-related

mid-ocean ridges (e.g., Iceland[21]), Oceanic Island Basalts (OIBs; ref. 16,22-26) and Continental Rift

(CR) magmas[27], as these melts are extracted from deeper (>50 km) sources than MORBs. However,

the C-enriched signature[1,16] of OIBs cause their volatile saturation deep in the crust/mantle, making

MIs often degassed[24-26] (Fig. 1b). The case is even more exacerbated in CR setting, where the

prevailing silica-undersaturated alkaline erupted magmas imply a metasomatised C-rich mantle

source[27], but even the most CO_2-rich (up to ~ 1 wt%) MIs[28-29] are affected by pre-entrapment C-loss

(Fig. 1b, c). Nevertheless, a better knowledge of the C content in mantle sources underneath rifting

cratons[27] is vital to modelling the deep C cycle[30-34].

51   Carbon that is lost to vapour during decompression-driven magma degassing, while predominantly

52   escaping the MI record, can be directly quantified by measuring the shallow release of magmatic

53   gas CO_2 through fumaroles and plumes during both volcano quiescence and eruption[31]. Considering

54   the low residual volatile contents in erupted magmas[9], the magmatic gas output corresponds to the

55   total volatile budget of the magma batch under the condition that closed-system degassing has

56   prevailed during ascent, followed by near-surface gas-melt separation (as observed at mafic

57   volcanoes[35-36]). In worldwide subduction-zone volcanoes[37-38] for example, the abundance of CO_2

58   scaled to more magma-soluble sulphur S (i.e., the CO_2/S_T ratio, where $S_T = SO_2 + H_2S$) correlates

59   with the trace element slab-fluid proxies in magmas, implying an essential control exerted by the

60   volatile signature of the source mantle wedge[39].

61   Non-arc (e.g., MOR, OIB and CR-related) magmatic gases are typically richer[40] in CO_2 compared

62   with their arc counterparts[37], with extreme CO_2 enrichments observed in gas released by alkaline

63   magmatic systems of the East African Rift (EAR), where the source mantle is expected to retain

64   high C amounts[27]. However, the link between C-rich magmatic gases and their mantle source

65   characteristics has not been quantitatively explored.

66

67   **Magmatic gas compositions and parental melt C contents**

68   There are only 12 volcanoes in plume-related (MOR and intraplate) and CR settings for which high-

69   temperature (>450 °C) gas composition information is available (see Extended Data Fig. 1-2 and

70   Extended Data Table 1-2 for location and data provenance). For this volcano selection, we illustrate

71   in Fig. 2 a,b (and Extended Data Fig. 3) the relationship between the time-averaged (mass)

72   magmatic gas CO_2/S_T ratios and the trace element compositions of the corresponding volcanic rocks

73   (see Methods). Among these, we focus on Sr, Nd, and Sm whole-rock ratios because these elements

74   exhibit similar behaviour (i.e., similar incompatibility) during magma generation in the mantle.

75   Thus, the spread of Sr/Sm and Sr/Nd ratios cannot be explained either by variable degrees of

76　melting of the same mantle source or by different degrees of magma differentiation (Extended Data

77　Fig. 4).

78　Indeed, the resulting correlations (Figs. 2a,b) suggest that (i) OIB and CR gases are C-enriched

79　(relative to S_T) compared to the composition of the DMM, and (ii) the level of such C enrichment

80　scales with the extent of enrichment in Sr (and other large ion lithophile elements, LILEs) in the

81　source magmas.

82　Ratios between incompatible/nonvolatile LILEs and light rare earth elements (LREE), which are

83　unaffected by magma differentiation/degassing upon ascent and crustal emplacement (Extended

84　Data Fig. 4), are typically used to constrain the enriched/depleted nature of their mantle sources

85　[37,41]. Thus, the correlations in Fig. 2a-b ultimately imply that the magmatic gas CO_2/S_T ratios are

86　also controlled by source mantle characteristics, e.g., that degassing-driven fractionation has little

87　control on time-averaged gas compositions[37]. The corollary is that magmatic gas CO_2/S_T ratios may

88　serve as proxies to estimate the parental C content in both undegassed magmas and the source

89　mantle.

90　For each of the 12 volcanoes we convert the CO_2/S_T ratios into parental melt C contents (Fig. 1) by

91　multiplying by the corresponding parental melt S contents. The latter are inferred from the measured

92　S contents (0.07 to 0.37 wt. %; Extended Data Table 1) in the most mafic (e.g., primitive) olivine-

93　hosted MIs for each volcano. Being generally entrapped at relatively high pressure (> 100 MPa),

94　these MIs have escaped S loss to gas[9]. Our inferred parental melt CO_2 contents range from 0.12 to

95　6.2 wt% (Extended Data Table 3), increase with magma alkalinity and are systematically higher

96　than (or, in a few cases, at the upper range of) those measured in MIs (Fig. 1a,b). This mismatch,

97　and especially the fact that our gas-derived CO_2 contents convert into CO_2/Ba and CO_2/Nb within

98　the mantle range (Figs 1b, c), while MI-based CO_2 contents do not, provides confidence in our

99　magmatic gas-based methodology and confirms that MIs are in most cases extensively degassed

100　(and C depleted relative to nonvolatile Ba and Nb; ref. 1-3).

101   Our results constrain the CO_2 transport capacity of CR magmas at 1-6 wt%, within the range of

102   parental melt CO_2 contents (of 3–24 wt%) predicted on the basis of CO_2/trace element ratios in MIs

103   from EAR magmas[29]. We emphasise that while correcting MIs for pre-entrapment CO_2 loss[1]

104   requires a priori assumptions on the source CO_2/trace element proxy ratio (which may vary in a

105   heterogeneous mantle[2]), our method does not. We also infer parental melt CO_2 contents of 0.12-1.1

106   wt% for plume-related tholeiitic magmas, with the lower and upper ranges being consistent with

107   recent independent estimates for Iceland[21] and Hawaii[22], respectively (Extended Data Table 3).

108

109   **Mantle C contents**

110   We next infer the C content of the mantle sources underneath the 12 volcanoes (Fig. 3 and Extended

111   Data Table 3) by applying a batch-melting equation (see Methods) to our derived parental melt CO_2

112   contents and using estimated mantle melting fractions (F) for each locality (listed in Extended Data

113   Table 3). As disparate and often limited information exists for mantle source characteristics

114   (mineral modal abundances and chemical composition), we cautiously determine F for each of the

115   12 volcanoes from modelling Zr partitioning between primitive melts and two possible end-member

116   mantle sources (primitive and enriched mantle; see Methods).

117   Our best-estimated F values range from 0.03 (Erebus) to 0.18 (Piton de la Fournaise) (Extended

118   Data Table 3). From these, and the parental melt C contents, we infer C contents in the mantle

119   sources ranging between 117 and 669 ppm (Extended Data Table 3). Our results unambiguously

120   demonstrate that, as recently suggested[12,22,42], mantle sources sampled by OIB and CR magmas are

121   systematically C-richer than the "averaged" DMM (20-56 ppm C, ref. 1,20,43). If deeper (than

122   normally sampled by MORBs) portions of the convecting mantle are C-rich, then the few C-

123   enriched DMM segments[20] that plot within the OIB/CR range (Fig. 3) must result from lateral

124   transport of enriched plume-related melts towards ridges[44].

125   In Fig. 3, our estimated mantle C contents positively correlate with the pressure of initiation of

126   mantle melting, as derived from using a P-dependent, H_2O-independent barometer (see Methods).

127   Our results thus identify a downward C increasing trend in the mantle source of OIB and CR

128   magmas (Fig. 3). We stress that the "mantle source" we here refer to corresponds to the region of

129   mantle melting and magma formation that, especially in the case of OIB, is shallower than the

130   initial source area of mantle plumes (which are typically rooted in the deep mantle).

131   We integrate the inferred C vs. depth dependence ($[C_{,ppm}] = 330 \cdot \ln(\text{depth}_{,km}) - 1120$; Fig. 3) down

132   to 200 km depth to estimate that the mantle melting regions that source OIB and CR magmas (Fig.

133   3) contain ~$1.2 \cdot 10^{23}$ g C (confidence interval, 0.95 to $1.4 \cdot 10^{23}$ g). Extrapolating the same function

134   to depths > 200 km is geochemically less constrained owing to the lack of recently erupted lavas

135   originating from such deep mantle sources and because redox freezing potentially limits deeper

136   infiltration of subducted carbonates[45]. Our calculations indicate that the upper (< 200 km) mantle

137   contains ~352 ppm C on average (confidence interval, 284 to 419). We conclude that the average

138   mantle C concentration exceeds that predicted (110±40 ppm C) using CO_2/Ba systematics of

139   minimally degassed magmas[13] and is more likely within the range obtained using the nitrogen

140   terrestrial budget[12] (337-479 ppm C). Recent mantle C estimates from OIBs (201-344, ref. 22; 368

141   ppm C, ref. 42) also match our estimate closely.

142

143   **Modelling the C vs. trace element association.**

144   Our parental melt/source mantle C estimates are based on the assumption that the consistent and

145   parallel enrichments in C and LILEs (relative to DMM) observed in plume-related (Iceland and

146   OIBs) and CR magmas (Fig. 2) are source controlled. There are obvious potential drawbacks to this

147   approach. For example, the magma CO_2/S_T budget can be potentially altered by interaction with

148   crustal C-rich rocks[37]. However, chemical and isotopic tracers (Sr-Nd-Pb-C) indicate that

149   assimilation of C-rich crustal lithotypes is negligible (if any) at most of our selected volcanoes

150   (Supplementary Information). We cannot exclude that at volcanoes overlying sedimentary

151   successions (e.g., Etna), a minor CO_2 fraction is derived from de-carbonatation reactions of crustal

152   limestones[37]. However, our CO_2 vs. Nb-Ba (Fig. 1) and Sr (Fig. 4) relationships rule out the

153   possibility that crustal limestones are major C sources that impact the gas CO_2/S_T ratio. Modelling

154   also shows that sulphide saturation in magmas has little control on magmatic gas CO_2/S_T

155   (Supplementary Information). We conclude, therefore, that the C-LILE/REE association (Fig. 2)

156   reflects their common derivation from a fertile (C-rich) source.

157   Experimental and petrological evidence suggests that the most common C-LILE carriers and

158   metasomatic agents[46] in the mantle are C-rich melts[6], formed via incipient melting of either

159   carbonated peridotites[4-5] and/or subducted carbonated oceanic crust[47]. Such incipient mantle melts

160   form a continuous trend from carbonatitic to carbonated silicate (kimberlite-like) melts[4-6], and

161   erupted carbonatitic (Cb) to kimberlitic (Kb) rocks thus represent their best surface-emplaced

162   analogues. These rocks are C-rich and have high Sr/Nd and Sr/Sm ratios (Fig. 4 and Extended Data

163   Fig. 5), and mantle metasomatism via carbonatitic/kimberlitic melts is therefore a suitable

164   mechanism to explain our CO_2/S_T vs. Sr/LREE association (Fig. 2). We therefore back-process our

165   gas vs. trace element data (Fig. 2) (i) to test whether the compositional trends exhibited by the 12

166   volcanoes correspond to those of mantle-derived melts formed by a suite of mantle sources variably

167   enriched in C and LILE and (ii) to verify whether the mantle enrichment process is consistent with

168   the addition, to a DMM-like mantle, of small amounts of metasomatic C-rich melts (mCm), an

169   analogue of natural kimberlitic (Kb) to carbonatitic (Cb) (Fig. 4, Extended Data Fig. 5) melts, and

170   (iii) to derive the trace element signature of these mCm by data regression.

171   The procedure (see Methods) involves calculation of $(CO_2/S_T)_M$ and $(Sr/X)_M$ ratios (where X is a

172   given trace element; Extended Data Fig. 6) of the mantle sources (M) at equilibrium with the

173   original volcanic gas vs. rock dataset (Fig. 2). This was obtained by applying a batch-melting

174   equation and using a range of melting fractions F (0.025, 0.5, 0.1, 0.2) covering the mantle melting

175   conditions observed at the studied volcanoes (Extended Data Table 3). Then, the obtained

176    $(CO_2/S_T)_M$ and $(Sr/X)_M$ ratio population was fitted using a best-fit mixing equation (see Methods;

177   Extended Data Fig. 6). This data regression is initialised with a set of assigned C, S, and Sr values

178   for the mCm (Extended Data Table 4) based on the composition of natural kimberlitic and

179   carbonatitic rocks and/or experimental melts[4-6] (Fig. 4, Extended Data Fig. 5). From this, the whole

180   trace element compositional suite of the mCm is obtained by regression (Extended Data Table 4 and

181   Fig. 5). We consider five distinct model scenarios (Cb1, Cb2, Kb1, Kb2, and Kb3) to cover the

182   possible C, S, and Sr mCm ranges (see Methods).

183   The results (Fig. 5a,b) show that our model curves well match the gas (CO_2/S_T) vs. trace element

184   global populations. Therefore, partial melting of a suite of variably C-LILE-enriched mantle sources

185   derived from 0.01-0.1 wt% (tholeiites) to 0.1-2.0 wt% (alkaline CR magmas) mCm addition to the

186   DMM is a suitable mechanism to explain the global C-LILE enrichment pattern (Figs. 1,4-5 and

187   Extended Data Fig. 5). The derived primordial mantle (PM)-normalised trace element compositions

188   of our mCms share high chemical affinity with natural (erupted) kimberlites (Fig. 5c), suggesting

189   that carbonated silicate melts (rather than carbonatitic melts) are the most likely metasomatic fluids

190   in the mantle. Carbonatitic[47] and kimberlitic[48] melts have both been found trapped in sub-

191   lithospheric diamonds and, thus, are associated with metasomatic fluids permeating the lower

192   portion of the upper mantle. The stability of these CO_2(-H_2O)-bearing fluids is linked to P-T-f_{O_2}

193   conditions required for melting of diamond-bearing peridotites to occur. For a lithospheric mantle

194   whose redox state decreases with depth[49], kimberlitic melts (approximately 10-20 wt% CO_2; our

195   Kb1 and Kb2 modelled compositions) would form in equilibrium with a diamond-bearing source at

196   $-3 \leq f_{O_2}$ (ΔFMQ) ≤ -2 log units[49].

197   Our results provide compelling evidence for the role of mCm in determining the C-LILE

198   heterogeneity of the Earth's upper mantle (Figs. 5-6). Even more importantly, the dependence

199   between mantle C content and depth/pressure on initiation of melting we bring to light (Fig. 3)

200   provides independent observational confirmation for the experimentally determined[4-5] role of C in

201   driving melting in the mantle. It has been experimentally demonstrated[4-5] that trace C in the upper

202   mantle forces mantle peridotites to start melting significantly deeper than would occur in volatile-

203   free conditions. It has been argued, in particular, that 0.1-1 wt% carbonated silicate melts can form

204   at 100-250 km depth in a moderately wet (0-200 ppm H_2O) carbonated mantle containing 27-81

205   ppm C (ref. 4). Our results (Fig. 3) provide evidence for C-rich mantle regions melting deeper than

206   DMM. If, as our data suggest, the Earth's mantle contains ~400 to ~700 ppm C in the 100-250 km

207   mantle window (Fig. 3), then C-induced incipient melting of carbonated peridotite may advance to

208   melt fractions higher (even by two orders of magnitude) than previously thought[5]. If we assume, for

209   instance, that incipient melts that form are kimberlite-like magmas with 10 to 20 wt% CO_2 (ref. 4-

210   5), then a 400-700 ppm mantle C range would imply melting fractions of 0.7-2.6 wt%. Redox

211   melting may also play a major role, provided redox conditions required to oxidise a diamond-

212   bearing mantle source ($Fe^{3+}/\sum Fe \gg 4\%$) persist. Indeed, the addition of C to an Fe^{3+}-rich mantle

213   source would cause the onset of (redox) melting at much deeper conditions[49]. Upon ascent, the so-

214   formed carbonated silicate melts may thus exert a key control on the mineralogical[46] and chemical[41]

215   heterogeneity of the overlying mantle, sampled by OIBs and MORBs (Fig. 6). If mCms are the

216   main C carrier in the deep upper mantle (Figs. 4-6), then their limited transport capacity in the

217   shallow lithospheric mantle may ultimately control the C downward trend (Fig. 3). The C-rich

218   mantle our results evoke, and the incipient melting that (in combination with H_2O) this C would

219   lead to, may also well explain[1,2,4] the geophysical anomalies in the upper mantle that, to be

220   observable, would require higher melt fractions. Ultimately, our results emphasise the central role C

221   plays in driving the geodynamics and long-term evolution of our planet.

222

223   **Data availability**

224   All data generated or analysed during this study are included in this published article (Extended

225   Data Tables 1-4). The dataset is also publicly available in the Earthchem data repository

226   (https://www.earthchem.org/ecl/).

227

228   **Code availability**

229   The code that supports the findings of this study and used to generate Figs. 1,5 and Exstended Data

230   Fig. 6 is available from author Giancarlo Tamburello (giancarlo.tamburello@ingv.it) upon request.

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Author contribution

A.A. devised the study concept. A.A., F.C., M.C., V.S. and G.T. contributed to refinement of the initial concept, and to data analysis and interpretation. A.A. drafted the original version of the manuscript with contributions from all coauthors.

385   **Figure Captions**

386   **Figure 1 – CO_2 contents in mafic melts from plume-related and Continental Rift volcanoes.**

387   Our estimated parental melt CO_2 contents (Extended Data Table 3) are shown on the vertical axis,

388   as determined by combining measured volcanic gas CO_2/S_T ratios with S contents in primitive MIs.

389   **(a)** The calculated parental melt CO_2 contents are plotted against measured CO_2 contents in MIs in

390   the same volcanoes (x-scale), where available; the latter, being systematically lower, imply

391   extensive CO_2 loss prior to/after MI entrapment. The derived CO_2 content of Ol Doinyo Lengai

392   overestimates the real parental melt CO_2 (the gas compositions used include low-temperature gases

393   where S has been scrubbed; see Extended Data Table 1). The measured Ardoukoba gas is a residual

394   (CO_2-depleted) gas (Extended Data Table 1); as such, we instead use the CO_2/S_T ratio inferred from

395   the gas-trace element relationship (Fig. 2): $[(CO_2/S_T)_i = 0.35 \cdot exp(0.03 \cdot (Sr/Sm)_m))$. The same

396   equation is used to calculate parental melt CO_2 contents for the Laki and Nabro eruptions, for which

397   no volcanic gas data exist. **(b)** Nb vs. CO_2 plot, in which the majority of the "degassed" MIs plot

398   below (at lower CO_2/Nb ratios lower than) the CO_2-Nb mantle array (grey area), as defined by

399   undegassed MORB MIs and glasses (characteristic upper mantle CO_2/Nb ratios are from ref.

400   2,3,13,50). Our inferred parental melt CO_2 contents, when plotted in tandem with measured Nb

401   contents in primitive MIs or whole/rocks (Extended Data Table 1), extend the mantle array to more

402   C-Nb enriched compositions; stars indicate the derived composition of kimberlitic (Kb) to

403   carbonatitic (Cb) metasomating C-rich melts (mCms), see text; **(c)** same as (b), but using Ba in

404   primitive MIs or whole/rocks as trace element proxy. Our parental melt CO_2-Ba population falls

405   within the CO_2/Ba mantle range (74-120). In (b) and (c) the compositional field of crustal

406   limestones is from ref. 37.

407

408 **Figure 2 – Time-averaged volcanic gas CO_2/S_T ratios vs. mean whole-rock Sr/Sm and Sr/Nd**

409 **ratios.** Scatter plots contrasting, for the 12 volcanoes, the time-averaged volcanic gas CO_2/S_T ratios

410 with the corresponding mean whole-rock Sr/Sm and Sr/Nd ratios (Extended Data Table 1). The

411 best-fit regression lines (with equations and regression coefficients) are shown in red. The averaged

412 volatile composition of the DMM is calculated by combining data in ref. 1,3,18,20.

413

414 **Figure 3 – C contents in the source mantles of plume-related (OIB and Iceland) and**

415 **Continental Rift volcanoes.** C contents (Extended Data Table 3) derived by applying a batch-

416 melting equation to the parental melt CO_2 contents and using an appropriate melting fraction (F) for

417 each volcano (Extended Data Table 3 and Methods). For each volcano, the symbol identifies our

418 best-estimated C content (the horizontal bar identifies the possible range for the possible F range),

419 and is plotted against pressure/depth of initiation of melting (Methods). The C vs. depth population

420 was fit by data regression (we also included the C= 20 ppm mode[20] of the C frequency distribution

421 in the DMM in the data fit). This identifies a downward increase in C concentrations in the upper

422 mantle (equation shown with 2.5% and 97.5% confidence intervals). Our inferred melting depth vs.

423 C dependence is statistically significant, as a one-tailed Spearman test yields a p-value of 0.0004848

424 (e.g., there is <5% probability that the two variables are uncorrelated).

425 For comparison, we also show the mantle C contents for 711 individual MOR segments[20] and for

426 some plume-related OIB volcanoes (see legend, parental melt CO_2 contents from ref. 1 were

427 converted into source mantle C using the batch melting equation and inferred melting fractions; see

428 Extended Data Table 3 for data sources).

429

430 **Figure 4 - The composition of natural carbonatites and kimberlites**. These are used to constrain

431 the C, S and Sr composition of our metasomatic C-rich melts (mCm) Cb1-2 to Kb1-3. (**a**) CO_2 vs.

432 Sr global distribution of natural carbonatitic and kimberlitic rocks (source GeoRoc;

433 http://georoc.mpch-mainz.gwdg.de/georoc/). These define a compositional array overlapping (but

434 extending to more enriched compositions) the compositional array exhibited by our parental CO_2

435 (inferred, Fig. 1 and Extended Data Table 3) vs. Sr trend for the 12 selected volcanoes. The high

436 correlation exhibited by the latter supports the accuracy of our estimated parental CO_2 contents. The

437 best fit equation (regression coefficient shown in figure) is used to extrapolate the Sr concentrations

438 at 20 and 40 wt. % CO_2, used for our mCm in all scenarios (except Cb1). MORBs and DMM plot at

439 less enriched (C-Sr-poorer) compositions. The compositional field of crustal limestones[37] is also

440 shown. (**b**) Derived Sr/Sm and Sr/Nd ratios for the 5 mCm. Sm and Nd are derived from data

441 regression (see Methods). Similar Sr/Sm and Sr/Nd ratios are obtained in the 5 model scenarios

442 explored.

443

444 **Figure 5 – Modelling the gas-rock association.** The volcanic gas CO_2/S_T ratio vs. (**a**) Sr/Sm and

445 (**b**) Sr/Nd ratio global association (same as Fig. 2) compared with the results of the melting/mixing

446 model. Our OIB/CR volcanoes identify a compositional array that spreads between the C-Sr-poor

447 MORB and DMM (see legend for data sources; grey diamonds are from the GeoRoc MORB pre-

448 compiled file; http://georoc.mpch-mainz.gwdg.de/georoc/) and a C-Sr-rich end-member. This trend

449 is inconsistent with variable degrees of melting of the same DMM mantle source (see the DMM

450 melting line). We interpret our OIB/CR array as being due to melting (melting fractions F shown)

451 of a heterogeneous C-LILE-enriched mantle source formed by small additions (mixing fractions X,

452 shown in red) of metasomatic C-rich melts (mCms; Cb1 and Kb1) to the DMM. The composition of

453 these mCms is estimated by applying a nonlinear least-squares procedure to the gas/whole-rock

454 population (see Methods). By sequentially applying the same procedure to a set of trace element

455 ratios, the entire suite of trace elements is derived for the mCms, as shown in (**c**). This spider

456 diagram compares the inferred primitive mantle (PM)-normalised trace element composition of the

457 C-rich metasomatic melts (mCm) (Extended Data Table 4) with the composition of natural

458 carbonatitic and kimberlitic melts (mean compositions of GeoRoc precompiled files;

459 http://georoc.mpch-mainz.gwdg.de/georoc/). For clarity, only two models (Cb1 and Kb1) are

460   shown, although additional scenarios are also explored (see Fig. 4 and Methods). For each of the 2

461   models, the mean (circles) and range (min/max; shaded red and white areas) are indicated.

462

463   **Figure 6 – Schematic representation of the vertical distribution of carbon in the upper mantle**.

464   East-to west transect from the Middle Atlantic Ridge (A) to the Java volcanic arc in the Sunda arc

465   (B) (the inset on the upper right shows the trace of the AB cross-section). The cross-section shows a

466   schematic representation of the vertical distribution of carbon in the upper mantle, as inferred from

467   the [C] vs. depth relationship of Fig. 3. Mantle C contents below specific volcanoes and ridge

468   segments are from this study and ref. 20, respectively. The structure and thickness on the Sub-

469   Continental Lithospheric Mantle are only for illustrative purposes, and are meant to highlight the

470   development of a C-enriched, metasomatised mantle source underneath the African Craton, leading

471   to C-enriched EAR volcanism upon continental rifting[27,32]. The downward increasing C in the upper

472   mantle reflects C extraction from the shallow DMM as well as variable extents of C refertilization

473   brought by incipient deep upper mantle melts (of carbonatitic to kimberlitic affinity[5]), as shown by

474   intermediate C-rich OIBs (e.g., Pico do Fogo and Piton de la Fournaise, Pdf, in the figure).

475

476

477   **Methods**

478   **Geological information.** The oceanic ridge system displayed in the upper right panel of Fig. 6 is

479   from ref. 51. The crust thickness along the A-B transect is from ref. 52, and the seismicity is from

480   ref. 53.

481   **Volcanic gas dataset.** We reviewed available data from the literature and existing catalogues[54-56] to

482   estimate the time-averaged magmatic gas CO_2/S_T ratios for a selection of CR, intraplate OIB, or

483   plume-related MOR (e.g., Iceland[57]) volcanoes. The results are shown in Fig. 2 and 5 and Extended

484   Data Table 1. For previous studies on volcanic arc settings[37,58-59], we only took into consideration

485   high-temperature (>450 °C) fumaroles and/or open-vent crater plumes in which the role of

486   hydrothermal S scrubbing[60-61] can be neglected. In these high-temperature systems, S_T normally

487   corresponds to SO_2 at surface conditions[37]. From an initial screening, only 12 volcanic systems with

488   the above characteristics were identified. Their location is shown in Extended Data Fig. 1 (base map

489   modified from ref. 62). The representativeness of the time-averaged (see Extended Data Table 1 for

490   temporal interval) compositions varies from volcano to volcano, depending on data availability. The

491   magmatic gas CO_2/S_T ratio is especially well constrained at volcanoes where permanent gas

492   monitoring (e.g., Etna[63]) or periodic surveys (e.g., Erebus[64]) have allowed for acquisition of robust

493   time series. In other volcanic systems, the available gas dataset is more limited due to either the

494   remoteness/difficulty of access (e.g., EAR volcanoes) or the lack of persistent high-temperature gas

495   emissions outside eruptions (e.g., Icelandic volcanoes, Pico do Fogo in Cape Verde). Among the

496   EAR volcanoes, Erta Ale in Ethiopia has exhibited a relatively invariant CO_2/S_T ratio composition

497   during (relatively infrequent) observations encompassing 5 decades (1971-2011). In contrast,

498   volcanic gas at Nyiragongo in the Democratic Republic of Congo has evolved towards less CO_2-

499   rich compositions from 1959-1972 (CO_2/S_T of 17.8-22.7) to 2005-2011 (CO_2/S_T of 2.7-6.1), and our

500   large standard derivation around the 1959-2011 average (CO_2/S_T of 11±6.8) reflects this trend

501   (Extended Data Table 1). Nyamuragira gas has only been measured twice to date[65], with rather

502   distinct compositions in 2014 (CO_2/S_T of ~5) and 2015 (CO_2/S_T of ~0.7), implying that the time-

503 averaged composition is poorly constrained for this volcano (2.9±3.1). Similarly, gas observations

504 at Ol Doinyo Lengai have been limited to low-temperature (< 300 °C) fumaroles[66-67] in which the

505 extremely high (>300) CO_2/S_T ratios imply scrubbing of magmatic S from the gas phase. The only

506 exception is represented by the fumarole measurements of ref. 68, for which however only the

507 average CO_2/S_T ratio (~200) of several distinct fumarolic vents (ranging in temperature from 78 to

508 519 °C) is available. As an average that also includes data for low-temperature manifestations, the

509 CO_2/S_T ratio of ~200 certainly overestimates the real magmatic gas signature for the volcano

510 (Extended Data Table 1). Among intraplate volcanoes, Pico do Fogo hosts a fumarolic field in its

511 summit crater with numerous but low-temperature (< 300 °C) vents. Information on magmatic gas

512 chemistry is thus limited to plume observations[69] during the recent 2014-2015 eruption. More

513 abundant gas information is in principle available for Piton de La Fournaise and Kilauea. In the

514 former, however, inter-eruptive gas emissions are very sluggish, while eruptive emissions are

515 logistically challenging to measure from the ground[70]. The only complete analysis of high

516 temperature magmatic gas emissions was obtained during the October 2010 eruption[71]. A vast gas

517 compositional dataset is available for Kilauea[72], but derivation of the characteristic magmatic gas

518 CO_2/S_T ratio for this volcano is problematic because separate (sequential) extraction of CO_2 (below

519 the summit caldera) and SO_2 (as magma laterally intrudes to erupt along the rift zone(s)) makes the

520 CO_2/S_T ratios of summit and rift gases different[73]. The low CO_2/S_T ratio (<1; ref. 72, 74-75)

521 signature of rift-related gases (for which the majority of the data are available) is thus

522 unrepresentative of the parental gas source. The 2008-2018 Halemaʻumaʻu summit eruption,

523 although preceded by a surge of CO_2-rich magma supply[76], was similarly associated with the

524 discharge of CO_2-poor gas and magma, implying pre-eruptive CO_2 loss[77]. As such, the early 20th

525 century lava lake gas samples remain the most representative of the parental gas source[73]. The most

526 representative restored gas analyses[78] have CO_2/S_T mass ratios ranging from 1.6 to 5.6, with the

527 latter (sample J8) corresponding to a "quenched equilibrium gas composition". We use this ratio,

528 corroborated by infrared spectroscopy results[79] for the 1967-68 summit eruption (CO_2/S_T ratio of

529   5.5), as the most representative of the parental magmatic gas signature at Kilauea. Note that the

530   reconstructed cumulative (summit + rift) CO_2 and SO_2 outputs for 2005-2007 and 2011-2017

531   (calculated from data in ref. 72, 74-76; ref. 80) lead to CO_2/S_T mass ratio of ~6, very close to our

532   inferred magmatic gas ratio of 5.6.

533   In addition to the measured compositions, Extended Data Table 1 shows the "predicted" volcanic

534   gas CO_2/S_T ratios for the Ardoukoba, Laki and Nabro volcanoes. The use of predicted rather than

535   measured gas compositions for Ardoukoba is motivated by the very CO_2-poor signature of the

536   latter[81] (Fig. 2), which has been explained as reflecting sampling of a residual, extensively degassed

537   magma source[40,82-83] (sampling was made at the base of the eruptive vent during the final stage of

538   the 1978 fissure eruption). No information on gas chemistry is available for Laki and Nabro, but

539   predicting CO_2/S_T ratios allows quantifying the parental melt CO_2 contents (Fig. 1a) for their 1783-

540   84 (ref. 57) and 2011 (ref. 84) eruptions. In all three cases, the volcanic gas CO_2/S_T ratios are

541   inferred using the gas vs. trace-element best-fit regression equation of Fig. 2a:

542    $$(CO_2/S_T)_i = 0.35 \cdot \exp(0.03 \cdot (Sr/Sm)_R) \quad (1)$$

543   where $(Sr/Sm)_R$ is the characteristic Sr/Sm ratio in either primitive MIs or mafic whole-rock

544   samples for each volcano. Errors in the inferred ratios are derived from uncertainty in $(Sr/Sm)_r$

545   ($\leq$10 %) and in the regression function ($\leq$20 %).

546   **Major and trace-element rock compositions.** For each of the 12 volcanoes, we inferred the time-

547   averaged magma composition by selecting the compositions of mafic (SiO_2 < 52 wt%; Extended

548   Data Fig. 2a) whole-rock (WR) samples (see Extended Data Table 1 for data sources). This same

549   methodology has been previously used in arc context[37-38]. The so-obtained trace element

550   compositions, listed in Extended Data Table 2 and illustrated in the chondrite-normalised spider

551   diagram[85] of Extended Data Fig. 2b, are used to calculate the trace element ratios shown in Fig. 2

552   and 5 (and Extended Data Fig. 3). In these figures, the trace element composition of the DMM is

553   from ref. 18, 86. Since other possible data sources (e.g., MIs) are limited or missing for several

554   volcanoes (see Extended Data Table 1), our use of a set of homogeneous WR-based trace-element

555   vs. CO_2/S_T populations appears more rigorous. Tests made for volcanoes for which robust WR and

556   MIs are simultaneously available (e.g., Kilauea, Extended Data Fig. 4) demonstrate overlapping

557   trace element ratios for the two datasets and reveal no dependence of trace element ratios on the

558   degree of magma differentiation (at least for SiO_2 <52 wt. % range considered here). This evidence

559   supports the use of averaged WR trace element ratios as representative of parental (primitive)

560   magma composition. The selection of mafic volcanic rocks in our compilation implies that rocks

561   showing typical SiO_2 enrichment owing to crystal fractionation have been excluded when present.

562   For example, data for only basanites and foidites (nephelinites) are averaged in the case of Erebus

563   and Nyiragongo, respectively, while more differentiated magmas (phonolites), and/or immiscibility

564   products (carbonatites), are not considered. Since absolute trace elements abundances vary upon

565   magma differentiation, for the sake of comparison with our inferred parental melt CO_2 contents in

566   Figs. 1b and 1c, we use our best estimates of Nb and Ba concentrations in parental magmas

567   (Extended Data Table 2), obtained by averaging the composition of the most primitive (SiO_2-poor

568   and Mg-rich) WR/MIs (see Extended Data Fig. 4 for the Kilauea example).

569   **Calculation of the mantle melting fraction.** For each volcano, we estimated the mantle source

570   partial melting degrees F, required to convert parental melt CO_2 contents into source mantle C (and

571   to generate the chosen primitive whole-rock compositions), by applying a nonmodal batch melting

572   equation[87]. We solve the equation, expressed as:

573                          $C_L = C_0/[D + F \times (1-P)]$ (2)

574   using the concentration of a selected incompatible element (Zr) in the most primitive magma of

575   each volcano. In equation 2, C_L is the Zr concentration in the magma, and C_0 the Zr concentration in

576   the mantle source. P is the partition coefficient of the element weighted for the eutectic mineral

577   melting proportions, and D is the partition coefficient weighted for the modal composition of the

578   mantle source. To obtain results comparable between each of the selected case studies and avoid

579   any possible dependency on modal composition of the mantle source/s and/or the mineral

580   proportions involved during melting, we assumed almost totally incompatible Zr behaviour. Such

581   an assumption results in the approximation of P and D to zero, thus simplifying equation 2 into C_L

582     $= C_0/F$. In this way, the partial melting degree of the mantle sources beneath each volcano can be

583   approximated as the ratio between the Zr concentrations in the source and the primitive magma ($F =$

584   C_0/C_L). Well aware that the dependency of this method is mostly on the concentration of Zr in the

585   mantle source, we conservatively calculated the partial melting degree from each primitive magma

586   composition by taking into account two different mantle source end-members, i.e. the primitive

587   mantle (PM; ref. 85), with a C_{Zr} of 11.2 ppm, and an enriched mantle (EM) analogous to that

588   proposed by ref. 88, with a C_{Zr} of 20 ppm (Extended Data Table 2). The "enriched" nature of the

589   studied magmas (Figs. 2-5 and Extended Data Fig. 2) would result in unrealistically low (0.01-0.06)

590   degrees of partial melting if a DMM end-member was considered. Our inferred F values are listed

591   (for both the considered PM and EM sources) in Extended Data Table 3. For each locality, we

592   select the most correct F solution from the scrutiny of available knowledge, e.g., existing literature

593   constraints on a PM/EM being implicated. Our F values are in good agreement with those

594   previously estimated at each locality (Extended Data Table 3).

595   **Calculation of the depth/pressure of melting initiation.** The pressure of partial melting (plotted

596   in Fig. 3) was calculated using a T-dependent, H_2O-independent barometer[89] (equation 42) applied

597   to the most primitive WR compositions from the selected volcanoes. This barometer considers the P

598   dependencies[90] of the melt silica activity, and was calibrated over a range of P, T and SiO_2

599   conditions (of respectively 0.0001-7 GPa, 825-2000°C and 31.5-70 wt.%) using a global dataset of

600   experimental partial melts. The T inputs for the barometer are the liquidus temperatures of the

601   selected primitive magmas, calculated using the Rhyolite-MELTS code[91]. To retrieve liquidus T,

602   we also consider (Extended Data Table 2) the most undegassed H_2O and CO_2 concentrations

603   reported on MIs for each of the studied magmatic suites (data-source, see Extended Data Table 1).

604   We stress a lack of interdependencies between thermometric and barometric equations, i.e., between

605   T and P and/or partial melting degree[92]. The same PT function[89] was also used to infer

606   depths/pressures of mantle melting initiation for the MOR segments[20] shown in Fig. 3. At this aim,

607   we used the segment-averaged potential temperatures, calculated from average ridge depths[20] and

608   the depth-temperature relationship inferred by ref. 93. The source mantle (DMM) C contents were

609   estimated[20] from MOR segment-averaged Ba contents[94], assuming a uniform CO_2/Ba mantle ratio

610   of 81. For comparison, the pressures of initiation of melting, previously inferred for our case study

611   volcanoes with a variety of geobarometric methods, are also listed in Extended Data Table 3. As an

612   example, according to recent petrogenetic models[95-96], the parental olivine melilititic magmas at

613   Nyiragongo are generated by small-fraction partial melting of a garnet- and phlogopite-bearing

614   mantle source generated by carbonate metasomatism[97]. This mantle mineral assemblage, in view of

615   experimental results on P-T conditions of peridotite melting[98], is consistent with melting depths of

616   80 (Nyamuragira) to 150 (Nyiragongo) km (ref. 97). Such depth values and associated partial

617   melting degrees are very close to those (82 and 150 km; Fs, of 7.9 and 5 %; Extended Data Table 3)

618   estimated in our study.

619   **Calculation of the trace element signature of the metasomatic agent**. The trace element

620   compositions of the endmember metasomatic C-rich melts (mCm) (Extended Data Table 4) are

621   obtained from data regression to the (CO_2/S_T) vs. trace element data. Our melting-mixing procedure

622   is illustrated in Extended Data Fig. 6 for the Sr/Sm example (an automatic calculation routine,

623   developed in R code, was applied interactively to a sequence of couples of trace element ratios). We

624   initially use the batch melting equation[87] to calculate the $(CO_2/S_T)_M$ and $(Sr/Sm)_M$ ratios of the

625   mantle source(s) from the original volcanic gas $(CO_2/S_T)_G$ and rock $(Sr/Sm)_R$ datasets (Extended

626   Data Fig. 6):

627   $$(CO_2/S_T)_M = (CO_2/S_T)_G \cdot [(Kd_C \cdot (1-F) + F)/(Kd_S \cdot (1-F) + F)] \quad (3)$$

628   $$(Sr/Sm)_M = (Sr/Sm)_R \cdot [(Kd_{Sr} \cdot (1-F) + F)/(Kd_{Sm} \cdot (1-F) + F)] \quad (4)$$

629   Calculations are performed for a set of melting fractions F (0.025, 0.5, 0.1, 0.2) covering the range

630   of mantle melting conditions at the studied volcanoes (Extended Data Table 3). In the calculations,

631   we use the trace element Kd values of C = 0.00055, S = 0.079, Sr = 0.025, and Sm = 0.045.

632   The generated $(CO_2/S_T)_M$ and $(Sr/Sm)_M$ populations (one for each F value; Extended Data Fig. 6)

633   are then fitted using an R-written routine[99] by a best-fit mixing equation[100]:

634                 $(Sr/Sm)_M = -((S_{DMM} \cdot Sr_{mCm} - S_{mCm} \cdot Sr_{DMM}) \cdot (CO_2/S_T)_M + (Sr_{DMM} \cdot C_{mCm} - Sr_{mCm} \cdot C_{DMM}))/$

635                 $((Sm_{DMM} \cdot S_{mCm} - Sm_{mCm} * S_{DMM}) \cdot (CO_2/S_T)_M + (Sm_{mCm} \cdot C_{DMM} - Sm_{DMM} * C_{mCm}))$ (5)

636   where $(CO_2/S_T)_{DMM}$ and $(Sr/Sm)_{DMM}$ refer to the characteristic ratios in the depleted MORB mantle

637   (DMM), and $(CO_2/S_T)_{mCm}$ and $(Sr/Sm)_{mCm}$ stand for the characteristic compositions of the

638   metasomatic C-rich melts (mCm). From experimental and petrological evidence[6], we assume the

639   most common mantle metasomatic agents are C-rich melts formed via incipient melting of either

640   carbonated peridotites[4-5] and/or subducted carbonated oceanic crust[101-102].

641   To solve equation 5, the C, S and Sr concentrations of the DMM are fixed averaging published

642   results[1,2,18,86], while those of the mCm are based on the composition of natural kimberlitic and

643   carbonatitic rocks and/or experimental melts formed during mantle rock melting experiments[4-6]

644   (see below). With a set of assigned C, S, and Sr values, the algorithm iteratively determines the

645   nonlinear least-squares estimate of the Sm concentrations of both DMM and mCm. This operation,

646   repeated interactively for a sequence of trace element ratios, allows deriving the whole trace

647   element composition suite of the mCm (Extended Data Table 4 and Fig. 5c).

648   Experiments[4-6,98] demonstrate that incipient mantle melts form a continuous trend from carbonatitic

649   to carbonated silicate (kimberlite-like) melts, and we therefore consider 5 distinct scenarios (models

650   Cb1, Cb2, Kb1, Kb2, and Kb3) to cover this possible range. In the Cb1 and Cb2 models (Extended

651   Data Table 4), we assume a mCm with the most CO_2-enriched (40 wt%) compositions, those of

652   natural (Fig. 4) and experimental (Extended Data Fig. 5) carbonatitic melts. Synthetic carbonatitic

653   liquids (< 10 wt% SiO_2; Extended Data Fig. 5) formed by melting of carbonated peridotites and

654   eclogites[5,102] are mostly dolomitic, implying that they do possess the Sr-rich compositional

655   signature (Fig. 4) required to explain our volcanic gas-trace-element associations (Figs. 2,5). We

656   select two "carbonatitic" model scenarios (Cb1 and Cb2) to explore the effect of Sr on our results,

657   in which the mCm Sr content is fixed at either 14,000 ppm (that of the most calcic natural

658   carbonatites; Cb1) or at 4,500 ppm (Cb2). The latter value is inferred using the Sr vs. CO_2

659   correlation we identify using our reconstructed parental melt CO_2 for plume (MOR/OIB) and CR

660   volcanoes (e.g., from extrapolating this correlation at 40 wt% CO_2) (Fig. 4).

661   Natural (erupted) kimberlitic rocks have compositional signatures controlled by extents of

662   degassing, assimilation (of mantle and crustal rocks) and alteration[103], and are thus unrepresentative

663   of primary melt compositions. Nevertheless, they arguably represent the best proxy for the

664   carbonated silicate melts rising from deep in the convecting mantle[104]. Other related ultrabasic

665   rocks, such as lamproites and lamprophyres, are typically less CO_2-rich and exhibit more radiogenic

666   isotope compositions, reflecting a shallower derivation in the Sub Continental Lithospheric

667   Mantle[104-105]. The GeoRoc CO_2 average of 7 wt% for natural erupted kimberlites is considered[104] to

668   represent a lower range for their parental mantle melt CO_2 contents, owing to extensive (>10 wt%;

669   ref. 102) degassing prior to/during surface emplacement. In our "kimberlitic" simulation scenarios,

670   we therefore assume CO_2 contents of either 20 wt% (ref. 104) (Kb1 simulation) or 7 wt% (Kb2 and

671   Kb3 simulations). The Sr contents of the Kb models (Extended Data Table 4) are inferred (as

672   above) from our Sr vs. parental CO_2 correlation (Fig. 4).

673   Our mCm are assumed to have relatively low S concentrations (1000 mg/kg for Cb1, Cb2, Kb1, and

674   Kb2). This is inferred from experimentally derived sulfur concentrations at sulfide saturation

675   (SCSS) (Extended Data Fig. 5) that imply limited S transport capacity of carbonatitic to carbonated

676   silicate melts below continents and oceans[106]. A 1000 ppm S mCm is also the GeoRoc average for

677   kimberlitic-carbonatitic natural rocks (Extended Data Fig. 5). Experiments[106] suggest a positive

678   dependence of SCSS on SiO_2 melt content (Extended Data Fig. 5), and we assume 2000 ppm S in

679   Kb3 model to explore the possible effect of this dependence (Extended Data Table 4).

680   One critical aspect is that while our inferred absolute trace element mCm compositions vary in

681   between the 5 model scenarios (Extended Data Table 4), the trace element ratios we derive are more

682   uniform (Figs. 1,4b). This evidence implies that the relative trace elements proportions are more

683   strictly determined by the fitted compositional volcano trends than by the assumed composition of

684   the metasomatic agent (mCm) we use for the fitting. The spider diagram of Fig. 5c demonstrates

685   this and shows that the primitive mantle (PM)-normalised Cb1 and Kb1 trends are very similar and

686   only shifted vertically. The Cb1 and Kb1 PM-normalised trends overlap the composition of natural

687   kimberlites, while they lack the strong Ta, Zr and Hf negative peaks of natural carbonatites. Our

688   mCms thus point to a metasomatic agent more affine to a carbonated silicate melt, rather than to a

689   carbonatite.

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692   **Methods references**

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Figure legend (as labeled in the figure):

Alkaline
- Ol Doinyo Lengai
- Erebus
- Nyiragongo
- Etna
- Nyamuragira
- Pico do Fogo

Tholeiitic transitional
- Kilauea
- Piton de la Fournase
- Erta Ale
- Surtsey
- Holuhraun (Bardabunga)

- Laki
- Nabro
- Ardoukoba

MIs, Icelandic Hotspot, OIBs, C. RIFT
- Oldoynio Lengai
- Etna
- Kilauea
- Erta Ale
- Surtsey
- Holuhraun (Bardabunga)
- Laki
- Nabro

MORBs (MIs and glasses)
- 14° N MAR[50]
- Equatorial MAR[2]
- Siqueiros[?]
- Borgarhraun (N Iceland)[21]
- Undegassed MORBs[19]

Panel (a): axes — Measured CO_2 in MIs (wt%) vs Parental CO_2 (%wt); 1:1 line shown.

Panel (b): axes — Nb (ppm) vs Parental CO_2 (%wt).
Mantle CO_2/Nb: 1300[13], 557[2], 534[50], 239[3]. Crustal limestones. MORBs. OIB & CR MIs. Labels: Cb2, Cb1, Kb1, Kb3, Kb2.

Panel (c): axes — Ba (ppm) vs Parental CO_2 (%wt).
Mantle CO_2/Ba: 120[13], 106[50], 97[2], 74[3]. Crustal limestones. MORBs. Labels: Cb2, Cb1, Kb1, Kb3, Kb2.

Legend:

Alkaline
- Ol Doinyo Lengai
- Erebus
- Nyiragongo
- Etna
- Nyamuragira
- Pico do Fogo

Tholeiitic transitional
- Kilauea
- Piton de la Fournase
- Erta Ale
- Surtsey
- Holuhraun (Bardabunga)
- Ardoukoba

(a) $y = 0.35e^{0.03x}$, $R^2 = 0.9$

(b) $y = 0.05e^{0.26x}$, $R^2 = 0.9$

Axes: CO_2/S_T vs Whole-rock Sr/Sm (a); CO_2/S_T vs Whole-rock Sr/Nd (b)

DMM

Mantle Carbon (ppm)

Depth of initiation of melting (km)

Pressure (GPa)

DMM

Lateral transport from plumes

$[C] = 330 \cdot \ln(depth) - 1120$

$R^2 = 0.6$

97.5%

2.5%

MORB mantle[20]
Mean DMM[20]

This work

Pico (Azores)[25]
El Hierro (Canary)[26]
Pitcairn[1]
Society[1]
Kilauea[16]
Kilauea[22]

Erebus
Nyiragongo
Etna
Nyamuragira
Pico do Fogo

Kilauea
Piton de la Fournase
Erta Ale
Surtsey
Holuhraun (Bardabunga)
Ardoukoba

Legend (top):

Alkaline
- Ol Doinyo Lengai
- Erebus
- Nyiragongo
- Etna
- Nyamuragira
- Pico do Fogo

Tholeiitic transitional
- Kilauea
- Piton de la Fournase
- Erta Ale
- Surtsey
- Holuhraun (Bardarbunga)
- Ardoukoba

MORBs MIs
- Equatorial MAR[2]
- Siqueiros[3]
- Borgarhraun (N Iceland)[21]
- Undegassed MORBs[19]

- Carbonatites
- Kimberlites

MORBs Glasses
- MORBs[1,18,20]
- MORBs[GeoRoc]
- Mean DMM[1,3,18,20]

End-member mCm [This study]

Panel (a): $y = 592x^{0.54}$, $R^2 = 0.8$; axes Sr ppm versus CO_2 wt %. Labels: Kb2,3; Kb1; Cb1; Cb2; MORBs; DMM; Crustal limestones.

Panel (b): axes Sr/sm versus Sr/Nd. Labels: Cb2, Cb1, Kb1, Kb2, Kb3, DMM.