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## Deep sourced fluids for peridotite carbonation in the shallow mantle wedge of a fossil subduction zone: Sr and C isotope profiles of OmanDP Hole BT1B

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1 **Deep sourced fluids for peridotite carbonation in the shallow mantle wedge of a fossil**  
2 **subduction zone: Sr and C isotope profiles of OmanDP Hole BT1B**  
3

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6 **Phase 1 Science Party<sup>†</sup>**

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12

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15

16

17 **Key Points:**

- 18 • Strontium and Carbon were added to the peridotites during alteration of mantle peridotite  
19 with carbonated fluid derived from decarbonation reaction.  
20

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109

## 110 **Abstract**

111 Completely carbonated peridotites represent a window to study reactions of carbon-rich fluids  
 112 with mantle rocks. Here we present details on the carbonation history of listvenites close to the  
 113 basal thrust in the Samail ophiolite. We use samples from Oman Drilling Project Hole BT1B,  
 114 which provides a continuous record of lithologic transitions, as well as outcrop samples from  
 115 listvenites, metasediments and metamafics below the basal thrust of the ophiolite.  $^{87}\text{Sr}/^{86}\text{Sr}$  of  
 116 listvenites and serpentinites, ranging from 0.7090 to 0.7145, are significantly more radiogenic  
 117 than mantle values, Cretaceous seawater, and other peridotite hosted carbonates in Oman. The  
 118 Hawasina sediments that underlie the ophiolite, on the other hand, show higher  $^{87}\text{Sr}/^{86}\text{Sr}$  values  
 119 of up to 0.7241.  $\delta^{13}\text{C}$  values of total carbon in the listvenites and serpentinites range from -  
 120 10.6‰ to 1.92‰. We also identified a small organic carbon component with  $\delta^{13}\text{C}$  as low as -  
 121 27‰. Based on these results, we propose that during subduction at temperatures above  $>400^\circ\text{C}$ ,  
 122 carbon-rich fluids derived from decarbonation of the underlying sediments migrated updip and  
 123 generated the radiogenic  $^{87}\text{Sr}/^{86}\text{Sr}$  signature and the fractionated  $\delta^{13}\text{C}$  values of the serpentinites  
 124 and listvenites in core BT1B.

125

## 126 **Plain Language Summary**

127 Samples from Oman Drilling Project Hole BT1B provide a record of interactions of fluids rich in  
 128 carbon dioxide with mantle rocks. This interactions lead to the formation of listvenites, rocks  
 129 composed mainly by magnesite and quartz. Here we describe the formation of listvenites in the  
 130 Oman ophiolite using Strontium and Carbon isotopes to characterize the source and nature of the  
 131 fluid that pervasively transform the mantle rocks that now store vast amounts of carbon dioxide.

## 132 **1 Introduction**

133 Hydration and carbonation of ultramafic rocks are important processes in the carbon and  
 134 water cycle of our planet (Alt et al., 2013; Fruh-Green et al., 2004). These alteration reactions are  
 135 sinks of water and carbon where peridotites are exposed on the seafloor forming alteration  
 136 minerals like serpentine and carbonates (Alt et al., 2013; Klein et al., 2020; Macdonald & Fyfe,  
 137 1985; Paulick et al., 2006) which are carried back into the mantle in convergent margins. Fluids  
 138 derived from the subducted slab can migrate and interact with the mantle wedge in subduction  
 139 zones, so that the “leading edge of the mantle wedge” aka the “cold nose”, can be partially  
 140 hydrated and carbonated (Blakely et al., 2005; Hyndman & Peacock, 2003; Kelemen &  
 141 Manning, 2015). This is usually inferred from seismic data (e.g. DeShon and Schwartz, 2004;  
 142 Kamiya and Kobayashi, 2000; Tibi et al., 2008; Tsuji et al., 2008). Understanding the interaction  
 143 of carbon-bearing hydrous fluids with peridotites is important to supplement and constrain  
 144 geophysical observations.

145

146 Fully carbonated peridotites, also known as listvenites (Halls & Zhao, 1995), provide a  
 147 window into the alteration processes that occur in the “cold nose” of the mantle wedge above  
 148 subduction zones, where mantle peridotite reacts with hydrous and carbonated fluids likely  
 149 derived from the subducting slab at moderate temperatures and pressures (Beinlich et al., 2012;  
 150 Boskabadi et al., 2017, 2020; Falk & Kelemen, 2015; Menzel et al., 2018). Formation of  
 151 listvenites appears to be restricted to particular conditions requiring high carbon concentrations

152 in the fluid (Falk & Kelemen, 2015; Hansen et al., 2005; Kelemen et al., 2021; Menzel et al.,  
 153 2018). For example, complete carbonation of peridotites to form listvenites is not observed near  
 154 the surface in ophiolites (e.g. Clark and Fontes, 1990; de Obeso and Kelemen, 2020, 2018;  
 155 Garcia del Real et al., 2016; Kelemen and Matter, 2008; Noël et al., 2018; Quesnel et al., 2016;  
 156 Schwarzenbach et al., 2016). It is also not observed near the seafloor, where serpentinization and  
 157 formation of carbonate veins are common (e.g. Bach et al., 2011; Delacour et al., 2008;  
 158 Schwarzenbach et al., 2013). Fluid sources for complete carbonation have been associated with  
 159 subduction in different listvenite localities. Menzel et al. (2018) attributed carbonation of  
 160 harzburgite in the Advocate ophiolite (Canada) to fluxing by slab-derived, CO<sub>2</sub>-rich fluids.  
 161 Isotopic data ( $\delta^{13}\text{C}$ ,  $\delta^{18}\text{O}$  and  $^{87}\text{Sr}/^{86}\text{Sr}$ ) of listvenites in the Late Cretaceous ophiolites of eastern  
 162 Iran point to carbon-bearing fluids derived from subducted sedimentary units as the source of  
 163 carbon (Boskabadi et al., 2020).

164 In Oman, listvenites occur along the basal thrust of the ophiolite (Nasir et al., 2007;  
 165 Wilde et al., 2002). Previous studies have investigated their formation conditions and the nature  
 166 of the carbonation fluids without reaching conclusive answers on the source of the fluids  
 167 (Beinlich et al., 2020; Falk & Kelemen, 2015; Nasir et al., 2007; Stanger, 1985). In this paper,  
 168 we present  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $\delta^{13}\text{C}$  data on samples from Oman Drilling Project Hole BT1B and the  
 169 underlying sediments of the Hawasina Formation. We show that devolatilization of the subducting  
 170 sediments similar to the Hawasina Formation likely generated the carbonation fluids which  
 171 reacted with the mantle wedge in this fossil subduction zone to form listvenites. These processes  
 172 probably operate in subduction zones worldwide, where fluids migrate updip along the slab  
 173 mantle interface, and then react with hanging wall peridotites storing large amounts of carbon.

174

## 175 **2 Geological setting**

### 176 *2.1 The Samail ophiolite*

177 The Samail ophiolite, along the northeast coast of Oman and the United Arab Emirates  
 178 (UAE), is one of the best-exposed block of oceanic crust and its underlying mantle in the world.  
 179 It was thrust over adjacent oceanic lithosphere soon after magmatic formation, and then onto the  
 180 margin of the Arabian subcontinent in the late Cretaceous. The mantle section of the ophiolite is  
 181 mainly composed of highly depleted, residual mantle peridotites (mostly harzburgites, e.g.  
 182 (Godard et al., 2000; Hanghøj et al., 2010; Monnier et al., 2006), together with 5 to 15% dunite  
 183 (Braun, 2004; Braun & Kelemen, 2002; Collier, 2012). Near the basal thrust, interlayered dunites  
 184 and refertilized harzburgites comprise the distinctive “Banded Unit” (Khedr et al., 2014). The  
 185 peridotites are pervasively serpentinized, with serpentine ( $\pm$  brucite) making up  $\sim$  30-100 wt% of  
 186 “fresh” rock (Godard et al., 2000; Hanghøj et al., 2010; Monnier et al., 2006) and/or completely  
 187 carbonated to form listvenites (Falk & Kelemen, 2015; Nasir et al., 2007; Stanger, 1985; Wilde  
 188 et al., 2002). The listvenites only occur within a few km of the basal thrust of the ophiolite, and

189 within the tectonic melanges with a serpentine matrix just below the base of the ophiolite (Nasir  
190 et al., 2007; Stanger, 1985).

## 191 *2.2 Lithologies below the Samail ophiolite nappe*

192 Beneath the mantle section of the Samail ophiolite is a locally preserved “metamorphic  
193 sole”. This sole is exposed discontinuously along the basal thrust, juxtaposed with the overlying  
194 Banded Unit at the base of the Samail mantle section. It records peak metamorphic temperatures  
195 of 700-900°C and imprecise peak pressures of 0.8 to 1.4 GPa (Cowan et al., 2014; Hacker &  
196 Mosenfelder, 1996; Searle & Cox, 2002; Searle & Malpas, 1980; Soret et al., 2017). A lower  
197 temperature unit (~450-550°C) with similar peak pressures (0.8 to 1.2 GPa) has also been  
198 identified from the metamorphic section of Oman DP Hole BT1B (Kotowski et al., 2021). The  
199 base of the sole is in fault contact with low grade allochthonous sediments of the Hawasina  
200 formation, which is composed of pelagic clastic units interlayered with limestones (Bechennec et  
201 al., 1988, 1990) deposited from the late Permian to the Cretaceous. The Hawasina sedimentary  
202 units were thrust over autochthonous Mesozoic to Proterozoic platform sediments of the  
203 Arabian continental margin, forming nappes between the autochthon and the ophiolite.

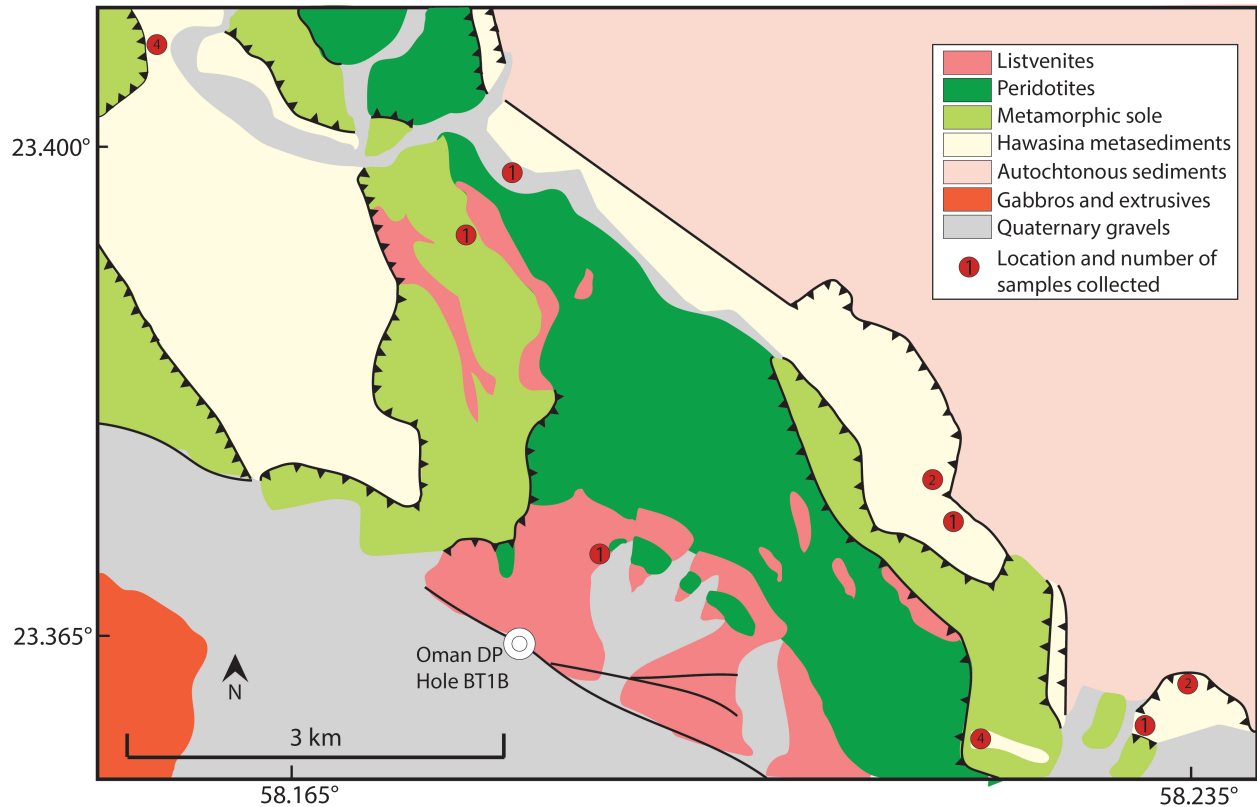
## 204 *2.3 OmanDP Hole BT1B and Oman Listvenites*

205 Hole BT1B was drilled in March 2017 in Wadi Mansah (23.364374°N, 58.182693°E),  
206 which yielded a total length of 300.1 m with 100% recovery (Kelemen et al., 2020). The upper 6  
207 meters are composed of alluvial gravels followed by an ultramafic sequence comprised of  
208 listvenites (carbonated peridotites) interlayered with two serpentinite bands (80-100 m depth and  
209 181 to 186 m depth). A thick (0.42 m) layer of grey-green fault gouge at 196.6 m–197.1 m depth  
210 separates the ultramafic units from the metamorphic sole composed of fine-grained metabasalts  
211 (Godard et al., 2021; Kelemen et al., 2021; 2020). To the first order, alteration of peridotite to  
212 form serpentinite and listvenite in Hole BT1B was nearly isochemical except for the addition of  
213 H<sub>2</sub>O and CO<sub>2</sub>. Average bulk rock Mg/Si, Fe/Si, Al/Si, Fe/Mg, and Cr/Al ratios in serpentinite  
214 and listvenite are close to the average composition of the Samail peridotite (Kelemen et al. 2020,  
215 2021, Godard et al. 2021) and similar to the composition of previously studied listvenites from  
216 the outcrops extending north and northeast from the drill site (Falk & Kelemen, 2015). The core  
217 provides a unique record of the interaction between peridotites in the leading edge of the mantle  
218 wedge and hydrous fluids rich in CO<sub>2</sub>. For an expanded version of the geology of Oman DP Hole  
219 BT1B and MoD mountain we refer the reader to the Proceedings of the Oman Drilling Project  
220 (Kelemen et al., 2020) and Kelemen et al. (2021).

## 221 **3 Materials and Methods**

222 Samples analyzed for this study comprise a suite of drill core samples from OmanDP  
223 Hole BT1B (listvenites n=50, serpentinites n=14, metamorphic sole rocks n=11) and hand  
224 samples of the Hawasina formation (n=18) and metamorphic sole sediments (n=2). Location of  
225 Oman DP Hole BT1B and Hawasina samples are shown in Figure 1. Drill core samples  
226 encompass all the identified lithologies from Hole BT1B. Major element compositions for Hole  
227 BT1B samples were reported in the *Proceedings of the Oman Drilling Project* (Kelemen et al.,  
228 2020) with the exception of samples in the 181-186m depth interval which are reported by  
229 Godard et al. (2021). Trace element compositions of Hole BT1B samples can be found in

230 Godard et al. (2021). Trace element compositions and loss on ignition for Hawasina formation  
 231 outcrop samples were analyzed at Lamont Doherty Earth Observatory (LDEO). Rb and Sr  
 232 concentrations were analyzed using a VG PlasmaQuad ExCell quadrupole ICP-MS following  
 233 HNO<sub>3</sub>-HF digestion. Major element compositions of the Hawasina samples are available in  
 234 supplementary table 1. Additional Sr isotopes were measured on samples from outcrops  
 235 northeast of Hole BT1B (Falk & Kelemen, 2015).



236  
 237 **Figure 1.** Simplified geologic map of Wadi Mansah and vicinity of Oman DP Hole BT1B  
 238 compiled after Villey et al. (1986), Google Earth and field observations. Hand samples collected  
 239 for this study are marked with red circles with number of samples collected per location in black.

240 For Sr isotope analysis, bulk rock powder was fully digested in a HNO<sub>3</sub>-HF mixture  
 241 overnight and redissolved in 3N HNO<sub>3</sub> prior to column chemistry using the Eichrom® Sr resin.  
 242 Purified Sr were analyzed for isotopic compositions interspersed with US National Institute of  
 243 Standards and Technology (NIST) SRM 987 on a Thermo Scientific Neptune multi-collector  
 244 ICP-MS at LDEO. In-run mass fractionations were normalized to <sup>86</sup>Sr/<sup>88</sup>Sr=0.1194. Unknowns  
 245 were normalized to SRM 987 <sup>87</sup>Sr/<sup>86</sup>Sr value of 0.701248. International standards BHVO-2  
 246 yielded <sup>87</sup>Sr/<sup>86</sup>Sr value of 0.703509±41 (2σ, n = 3) and BCR-2 yielded 0.705046±34 (2σ, n = 3),  
 247 which agree with published values from Weis et al. (2006).

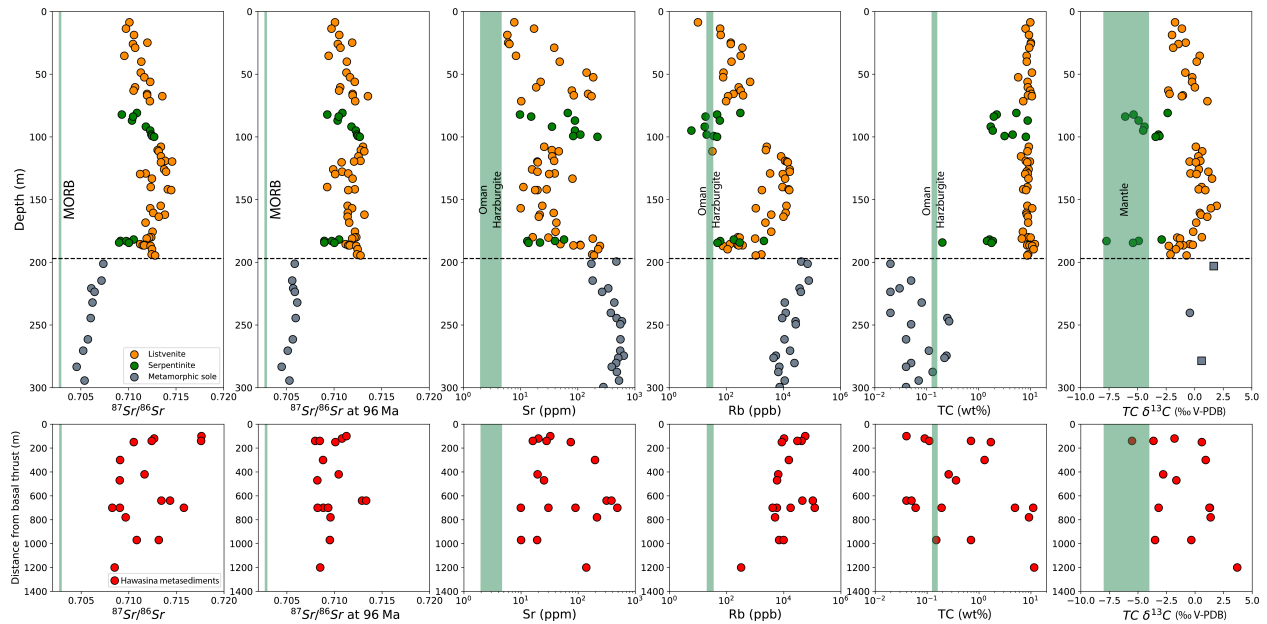
248 Total Carbon (TC) was measured from the same bulk rock powder splits as for Strontium  
 249 isotopes. Total Organic Carbon (TOC, or reduced carbon) was measured from the residual rock  
 250 powder after the removal of Inorganic Carbon (carbonate carbon) through reaction with dilute (3  
 251 N) HCl for at least 3 days, followed by washing with Millipore® water. Concentrations and δ<sup>13</sup>C



252 ratios of Total Carbon (TC) and Total Organic Carbon (TOC), were determined using a Costech  
 253 element analyzer coupled with a Thermo Scientific Delta V plus mass spectrometer at LDEO.  
 254 Sample runs were calibrated using Acetanilide for carbon contents ( $R^2=0.9998$ ). For  $\delta^{13}\text{C}$  we  
 255 used USGS40 ( $\delta^{13}\text{C} = -26.77 \pm 0.16\%$  V-PDB,  $n=4$ ), USGS41 ( $\delta^{13}\text{C} = 37.63 \pm 0.12\%$  V-PDB,  $n=4$ )  
 256 and USGS24 ( $\delta^{13}\text{C} = -16.04 \pm 0.13\%$  V-PDB,  $n=4$ ). All measured values of  $\delta^{13}\text{C}$  standards agree  
 257 with accepted values reported by the United States Geological Survey (USGS). Inorganic carbon  
 258 contents and  $\delta^{13}\text{C}$  of total inorganic carbon (TIC) were estimated by mass balance between TC  
 259 and TOC.

### 260 3 Results

261 Depth profiles of  $^{87}\text{Sr}/^{86}\text{Sr}$ , Sr and Rb concentrations, carbon concentrations, Total  
 262 Carbon and Total carbon  $\delta^{13}\text{C}$  are shown in Figure 2 and Table 1 and 2. The Hawasina  
 263 metasediments are plotted as the horizontal map distance to the closest mapped metamorphic  
 264 sole/ultramafic contact.



265

266 **Figure 2.** Depth profiles of OmanDP Hole BT1B (top panels) and distance from basal thrust for  
 267 Hawasina metasediments (bottom panels). From left to right  $^{87}\text{Sr}/^{86}\text{Sr}$  (measured),  $^{87}\text{Sr}/^{86}\text{Sr}$  age  
 268 corrected at 96 Ma, Sr concentrations, Rb concentrations, total carbon (TC) concentration, total  
 269 carbon  $\delta^{13}\text{C}$ , (squares in metamorphic sole from Zeko (2021)). Black dashed line in top panels  
 270 correspond to basal thrust. Green reference bands in all panels indicate, from left to right:  
 271  $^{87}\text{Sr}/^{86}\text{Sr}$  range of MORB (Average  $\pm 2\sigma$ ) (Gale et al., 2013; Hofmann, 2013), Sr and Rb  
 272 concentration range of Oman harzburgite (Average  $\pm 2\sigma$ ) (Godard et al., 2000; Hanghøj et al.,  
 273 2010; Monnier et al., 2006), total carbon content of Oman harzburgite (Average  $\pm 2\sigma$ ) (Kelemen  
 274 & Manning, 2015) and mantle  $\delta^{13}\text{C}$  (Average  $\pm 2\sigma$ ) (Deines, 2002). The distance from the basal

275 thrust for the Hawasina metasediments is measured as the horizontal distance to the closest  
276 mapped metamorphic sole/ultramafic contact.

277 **Table 1.** Lithological information, depth, strontium, rubidium and carbon concentrations and Sr  
278 and C isotope ratios of samples from OmanDP Hole BT1B

279 *Table 1 attached as Excel spreadsheet*

280  
281 Abbreviations SE= Standard error, TC= Total Carbon, TOC= Total Organic Carbon, V-PDB= Vienna Pee Dee  
282 Belemnite standard, n.d.=not determined

283  
284  
285 **Table 2.** Lithological information, location, strontium, rubidium and carbon concentrations and  
286 Sr and C isotope data of samples for Hawasina and metamorphic sole samples

287  
288 *Table 2 attached as Excel Spreadsheet*

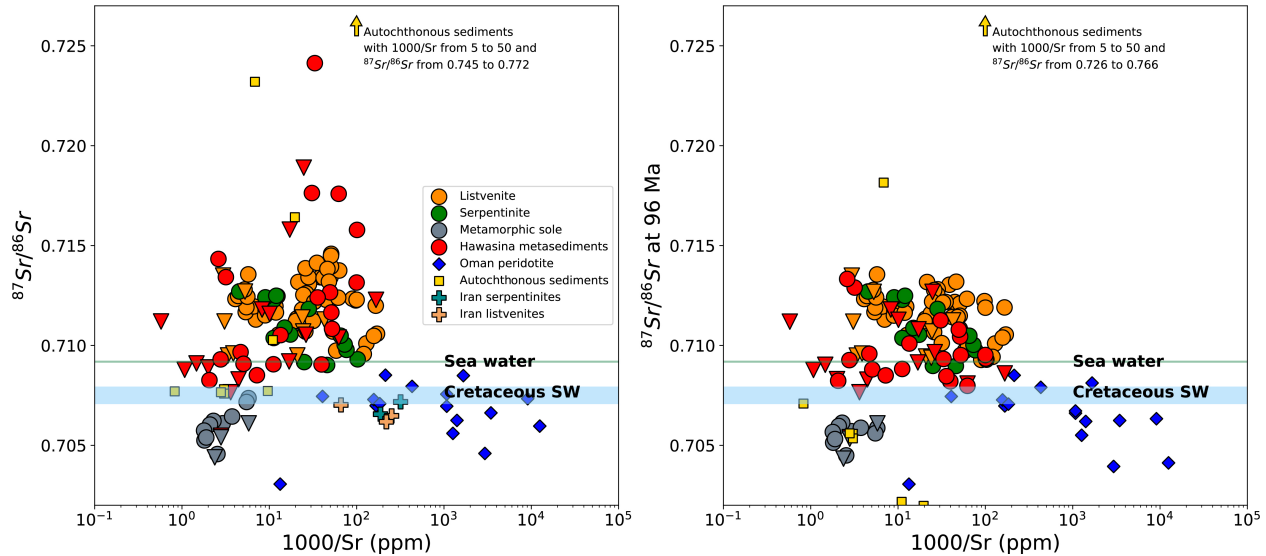
289  
290 Abbreviations SE= Standard error, TC= Total Carbon, T V-PDB= Vienna Pee Dee Belemnite standard,  
291 n.a.= not available, n.d.=not determined

292

### 293 *3.1 Strontium Isotopes*

294 Measured  $^{87}\text{Sr}/^{86}\text{Sr}$  values in Oman DP Hole BT1B lithologies show clear differences  
295 between (a) the metamorphic sole and (b) the listvenites and serpentinites. The listvenites and  
296 serpentinites vary from a minimum value of 0.709 in the upper serpentinite band to a maximum  
297 of 0.715 in some listvenites.  $^{87}\text{Sr}/^{86}\text{Sr}$  values in the listvenites increase with depth from the  
298 surface to 150 m while the serpentinite band between 80-100 m have lower values. Below 150  
299 m,  $^{87}\text{Sr}/^{86}\text{Sr}$  values are relatively constant with increasing depth, until the second serpentine band  
300 at 181-186 m. Below the basal thrust,  $^{87}\text{Sr}/^{86}\text{Sr}$  values are significantly lower in the metamorphic  
301 sole from 0.704 to 0.706, followed by lower  $^{87}\text{Sr}/^{86}\text{Sr}$  values with greater depths (Figure 2).  
302 Although the metamorphic sole contains meta-basalts (Godard et al., 2021; Kelemen et al., 2021;  
303 Kotowski et al., 2021; Searle & Malpas, 1980), all of the samples have higher  $^{87}\text{Sr}/^{86}\text{Sr}$  than  
304 typical mid-ocean ridge basalts (MORB) (Gale et al., 2013; Hofmann, 2013). The Sr isotope  
305 values of the listvenite and serpentinite samples from BT1B are similar to those of listvenites in  
306 nearby outcrops (Falk and Kelemen, 2015, Figure 2), which are significantly more radiogenic  
307 than  $^{87}\text{Sr}/^{86}\text{Sr}$  values of Oman peridotites (Gerbert-Gaillard, 2002; Lanphere et al., 1981;  
308 McCulloch et al., 1981) and Cretaceous to modern seawater (McArthur et al. 2001), but less  
309 radiogenic than some Hawasina and autochthonous sediments (Weyhenmeyer, 2000). Our  
310 serpentinites and listvenites are enriched in Sr and more radiogenic than similar lithologies in the  
311 Birjand ophiolite in eastern Iran (Boskabadi et al., 2020). Our samples of the Hawasina  
312 metasediments, and those collected from the same region by Falk and Kelemen (2015) have

313  $^{87}\text{Sr}/^{86}\text{Sr}$  values ranging from 0.7082 to 0.7241. Six of these Hawasina samples are more  
 314 radiogenic than any of the samples from Hole BT1B (Figure 3).



315  
 316 **Figure 3.**  $^{87}\text{Sr}/^{86}\text{Sr}$  values, both measured (left) and age corrected to 96 Ma (right), plotted versus  
 317 inverse Sr concentration for core samples from OmanDP Hole BT1B and other lithologies  
 318 measured in Oman and Iran. Data from Falk and Kelemen (2015) for samples of the same  
 319 lithologies as those documented here are shown as inverted triangles, employing the same color  
 320 scheme as used for samples in this study. Data for Hawasina sediments are from this study and  
 321 Falk and Kelemen (2015), those for Oman peridotites are from Gerbert-Gaillard (2002, leached  
 322 samples only), those for autochthonous sediment samples are from Weyhenmeyer (2000) and  
 323 those for Iran samples are from Boskabadi et al. (2020). Rb concentrations were not reported for  
 324 the autochthonous clastic metasediments from Weyhenmeyer (2000), so we assume a Rb upper  
 325 bound concentration of 200 ppm (twice the maximum measured in Hawasina metasediments).  
 326 This Rb concentration was used with the measured  $^{87}\text{Sr}/^{86}\text{Sr}$  and Sr concentration, to estimate a  
 327 minimum Sr isotope ratio at 96 Ma for these samples. Seawater and Cretaceous seawater values  
 328 from McArthur et al. (2001).

### 329 3.2 Carbon concentrations and $\delta^{13}\text{C}$

#### 330 3.2.1 Total Carbon

331 The carbon contents of samples from Hole BT1B are highly dependent on lithology  
 332 (Figure 2). All samples of ultramafic origin contain some carbon, mainly as Mg-rich carbonates,  
 333 with serpentinites containing 0.2 to 5 wt% carbon, and listvenites containing 6 to 12 wt%.  
 334 Samples from the metamorphic sole below 200 m depth contain 0.02 to 0.27 wt% carbon. In  
 335 contrast, the Hawasina metasediments have highly variable carbon contents. Some are almost  
 336 pure metamorphosed limestones and dolomites with carbon contents of up to 11 wt%, while  
 337 some have less than 0.05 wt% carbon.

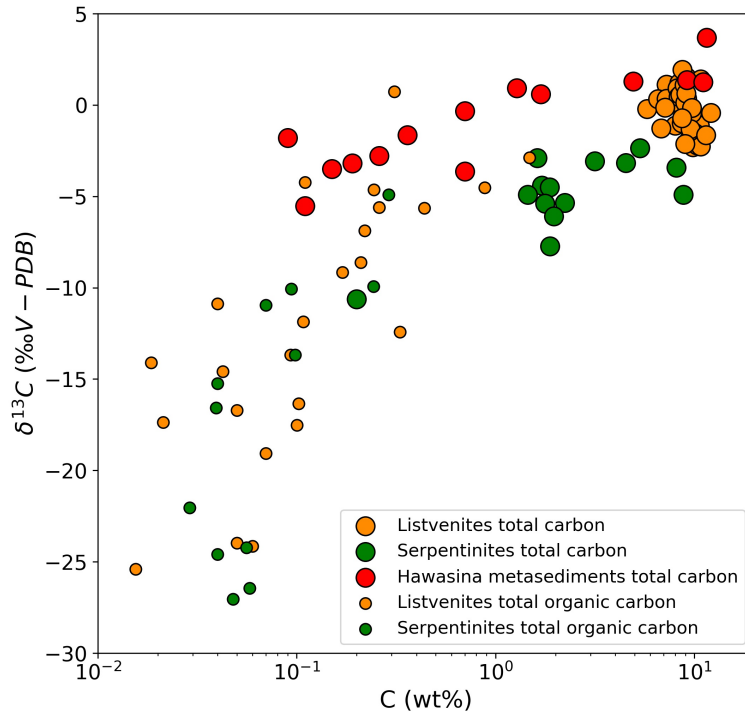
338  $\delta^{13}\text{C}$  in the upper 200m of Hole BT1B varies significantly between serpentinites and  
 339 listvenites (Figure 2). Serpentinites contain significantly lighter carbon than listvenites, as

340 observed in the depth profile (Figure 2). The lower serpentinite band contains two samples (74-2  
341 0.0-5.0 cm and 74-3 42.0-47.0 cm) that contain significantly lighter carbon than any serpentinite  
342 in the upper band. In general, carbon concentrations correlate positively with  $\delta^{13}\text{C}$  values in the  
343 upper 200 m (Figure 4). Metamorphic sole samples in the bottom 100 m have low carbon  
344 contents and did not yield enough  $\text{CO}_2$  for isotopic analysis. Total carbon  $\delta^{13}\text{C}$  values in the  
345 Hawasina nappes range from -5.52‰ to 3.69‰, which almost encompasses the entire range of  
346 variation observed in Hole BT1B and appears to decrease with lower carbon contents.

### 347 *3.2.2 Organic carbon*

348 Graphitic carbon was identified in the core both at the drill site and during core  
349 inspection aboard the D/V Chikyu (Kelemen et al, 2020, Kelemen et al. 2021). This organic  
350 component has a characteristic low  $\delta^{13}\text{C}$  signature in listvenites and serpentinites from Hole  
351 BT1B, which extends to a minimum of -27.04‰, representing 0.05wt% of total carbon in the  
352 sample from 74-1 56.0-64.0cm (Figure 4). In some cases, the intermediate values likely represent  
353 mixtures of organic and inorganic carbon, as some of the magnesite in the listvenites is very  
354 resistant to 3N HCl attack and remain undissolved even after 5 days of leaching. The observed  
355 light isotopic compositions are similar to those observed in other ultramafic localities such as  
356 Cerro de Almirez and Liguria (Alt et al., 2013). These data confirm the presence of organic

357 carbon in Hole BT1B, which was observed during drilling operations, core description aboard  
 358 D/V Chikyu and via Raman spectroscopy (Kelemen et al, 2020, Kelemen et al. 2021).



359

360 **Figure 4.** Total Carbon (large circles) and Total Organic Carbon (small circles) versus  $\delta^{13}\text{C}$  for  
 361 listvenites, serpentinites and Hawasina metasediments.

## 362 4 Discussion

### 363 4.1 Temperature and pressure of listvenite formation

364 Falk and Kelemen (2015) estimated the temperature range of listvenite formation based  
 365 on conventional and clumped stable isotope thermometry ( $90 \pm 15^\circ\text{C}$ ), phase equilibria ( $80-$   
 366  $130^\circ\text{C}$ ), and rock textures. New clumped isotope measurements on BT1B drill core samples by  
 367 Beinlich et al. (2020) widened the temperature range of listvenite formation and/or cooling from  
 368  $\sim 50 \pm 5^\circ\text{C}$  to  $250 \pm 50^\circ\text{C}$ . This range of values suggests that the infiltrating reactive fluids had  
 369 variable temperatures, and/or clumped isotope values were reset during cooling, as proposed for  
 370 fine-grained samples of peridotite-hosted carbonate veins (Garcia del Real et al., 2016). The  
 371 pressure of listvenite formation is very poorly constrained due to the lack of pressure-sensitive  
 372 assemblages and the small size of fluid inclusions. A minimum pressure of 0.3 GPa is provided  
 373 by the P-T conditions recorded by the Arabian carbonate platform during ophiolite obduction  
 374 (Grobe et al., 2019). For the upper limit, the listvenites must have formed at a pressure below the

375 maximum pressure reported for the metamorphic sole (~ 1.4 GPa) (Cowan et al., 2014; Searle  
376 and Cox, 2002).

#### 377 *4.2 Timing of listvenite formation*

378 Falk and Kelemen (2015) used Rb/Sr and  $^{87}\text{Sr}/^{86}\text{Sr}$  data on mineral separates to produce  
379 an imprecise isochron age of  $97 \pm 29$  Ma for a listvenite sample. This age is broadly consistent  
380 with the ~96 Ma age of formation of igneous crust in the ophiolite, along with the same age of  
381 metamorphism for the underlying metamorphic sole just beneath the basal thrust of the ophiolite  
382 (Hacker, 1994; Hacker et al., 1996; Rioux et al., 2013, 2016). Moreover, listvenites are found in  
383 and near the basal thrust, from the UAE near the northwestern end of the ophiolite outcrop to the  
384 area around Hole BT1B, near the southeastern end of the ophiolite outcrop in Oman (Nasir et al.,  
385 2007; Stanger, 1985). The extensive outcrop NE of Hole BT1B, known informally as MoD  
386 Mountain, exposes the Banded Horizon, a peridotite unit found at the base of the ophiolite  
387 mantle section composed of alternating 1- to 10-meter scale bands of dunite, harzburgite, and  
388 minor lherzolite. This unit has distinct geochemical characteristics, with higher Al and middle  
389 rare-earth-elements, compared to the residual mantle harzburgites that comprise most of the  
390 mantle section of the ophiolite (Boudier et al., 1988; Godard et al., 2021; Khedr et al., 2014;  
391 Prigent et al., 2018; Yoshikawa et al., 2015). The BT1B listvenites have compositions suggesting  
392 that they were formed after basal peridotites from the Banded Horizon (Godard et al., 2021).

393 Throughout this region, the contacts of listvenite bands within and at the base of the  
394 Banded Unit are broadly parallel to banding in the peridotite, and to the contacts between  
395 listvenite, peridotite, metamorphic sole, and the Hawasina nappe. These data are consistent with  
396 our hypothesis that most of the listvenites formed by alteration of mantle peridotite during the  
397 subduction of the underlying sediments via intra-oceanic thrusting and/or later emplacement of  
398 the ophiolite onto the Arabian continental margin.

399 On the basis of steep, fault-bounded contacts of listvenite with young, post-emplacement  
400 conglomerates, several workers have inferred that the listvenites formed during later uplift and  
401 extension (Nasir et al., 2007; Stanger, 1985; Wilde et al., 2002). Recently, Scharf et al. (2020)  
402 reported  $60 \pm 16$  Ma and  $58 \pm 6$  Ma U/Pb ages for two carbonate veins that cut listvenite, and  
403 structural observations indicating a top-down sense of shear along some faulted listvenite-  
404 peridotite and listvenite-sole contacts. Following prior interpretations, they interpret these ages  
405 as formation ages of the listvenites after ophiolite emplacement during uplift of the nearby Jebel  
406 Akdar and Saih Hatat anticlinoria. We find that their interpretation is inconsistent with the field  
407 observations reported above and evidence of a multistage tectonic overprint after peridotite  
408 carbonation and listvenite formation as reported by Menzel et al. (2020) and Kelemen et al.  
409 (2020).

410 We have age-corrected our  $^{87}\text{Sr}/^{86}\text{Sr}$  to the 96 Ma age reported by Falk and Kelemen  
411 (2015). This correction gives the lowest possible  $^{87}\text{Sr}/^{86}\text{Sr}$  for all the samples based on reported  
412 ages. For the corrections, we used Sr and Rb concentrations reported by Godard et al. (2021) for  
413 Hole BT1B and our analyses of Hawasina metasediments. While Rb/Sr is low in most BT1B  
414 samples and thus age corrections are small, this correction removes some of the apparent trends  
415 observed in measured  $^{87}\text{Sr}/^{86}\text{Sr}$  versus depth. The age corrections particularly affect listvenites  
416 with relatively abundant chromian mica (fuchsite-muscovite solid solutions, Falk & Kelemen

417 2015, supplement) in the 115-163 m depth interval, as these micas host abundant Rb (Godard et  
418 al. 2021). The age correction also affects the estimated  $^{87}\text{Sr}/^{86}\text{Sr}$  values of the metamorphic sole  
419 of BT1B and some of the Hawasina metasediments as their Sr and Rb concentrations are  
420 heterogeneous, ranging from 9 to 638 ppm for Sr and 0.4 to 97.7 ppm for Rb. Regardless, the  
421 age-corrected  $^{87}\text{Sr}/^{86}\text{Sr}$  values of the listvenites are much higher than those of the mantle and  
422 Cretaceous seawater.

#### 423 *4.3 Fluid source for carbonation of peridotites in Oman DP Hole BT1B*

424  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $\text{d}^{13}\text{C}$  data on MoD Mountain listvenites and Hole BT1B samples suggest  
425 that replacement of peridotite by serpentinite and listvenite resulted from reaction with a single  
426 fluid along a reaction path (Kelemen et al., 2021). The initial fluid was far from equilibrium with  
427 peridotite, which converted olivine and serpentine in the protolith to carbonates + quartz and  
428 approached equilibrium with serpentinite at higher extents of reaction progress and lower  
429 integrated water/rock ratios (Beinlich et al., 2020, Kelemen et al., 2021). Mg isotope data from a  
430 set of samples studied by Falk and Kelemen (2015) show significant differences between  
431 dolomite and magnesite listvenites. Dolomite listvenites (average  $\delta^{26}\text{Mg} \sim -1.33\text{‰}$ ) are lighter in  
432 Mg isotopes than magnesite listvenites (average  $\delta^{26}\text{Mg} \sim -0.33\text{‰}$ ) (de Obeso et al., 2021), which  
433 suggests magnesite dissolution and dolomite formation. This is consistent with the modelled  
434 evolution of listvenites during fluid-rock reaction, with dolomite replacing magnesite at  
435 increasing water/rock ratios (Kelemen et al., 2021). The process of formation of dolomite veins  
436 at the expense of magnesite has been documented in other Cretaceous ophiolites (Boskabadi et  
437 al., 2020).

438 As noted above, after the magnesite and dolomite listvenites formed they were  
439 cataclastically deformed, and then cut by late Ca-rich carbonate veins (Menzel et al. 2020). Thus,  
440 one might expect the veins to have formed from a later, geochemically distinctive fluid as  
441 suggested by variable clumped isotope ( $\Delta 47$ ) derived temperatures (Beinlich et al., 2020) and  
442 changes in geochemical signatures in dolomite-dominated intervals (Godard et al. 2021). This  
443 can be further addressed in future studies via careful sampling of the post-cataclastic veins and  
444 dolomite-rich domains.

445 Returning attention to the source of the fluid that formed the bulk of the listvenites,  
446 assuming that Sr and  $\text{CO}_2$  were derived from the same fluid, Sr isotopes can be used to constrain  
447 the source of the carbonating fluids that formed the serpentinites and listvenites. Falk and  
448 Kelemen (2015) proposed three possible sources of fluids for carbonation: (1) compaction of  
449 pore waters from underlying Hawasina metasediments, (2) low temperature dehydration of opal  
450 and clay minerals in calcite-bearing Hawasina metasediments, and (3) higher-grade metamorphic  
451 devolatilization reactions involving subducted sediments similar to the Hawasina metasediments  
452 coupled with fluid that migrated up the subduction zone.

453 Unlike younger, mantle-peridotite-hosted carbonates, the listvenites do not contain a  
454 significant fraction of seawater- or groundwater-derived Sr (Gerbert-Gaillard, 2002; Kelemen et  
455 al., 2011; de Obeso & Kelemen, 2018; Weyhenmeyer, 2000). The listvenites have  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios  
456 that are distinct from recent, low-temperature carbonate veins and travertine in the mantle section  
457 of the ophiolite (Kelemen et al., 2011; de Obeso & Kelemen, 2018; Weyhenmeyer, 2000).  
458 Almost all of the listvenites and associated serpentinites from BT1B core and MoD Mountain

459 outcrops have  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios at 96 Ma higher than Cretaceous seawater (Falk & Kelemen 2015,  
460 and this paper). In contrast, young, peridotite-hosted carbonate veins and travertines in the  
461 Samail ophiolite consistently have  $^{87}\text{Sr}/^{86}\text{Sr}$  lower than 0.709 and appear to contain mixtures of  
462 Sr derived from seawater or groundwater and the mantle (Gerbert-Gaillard, 2002; Kelemen et al.,  
463 2011; de Obeso & Kelemen, 2018; Weyhenmeyer, 2000).

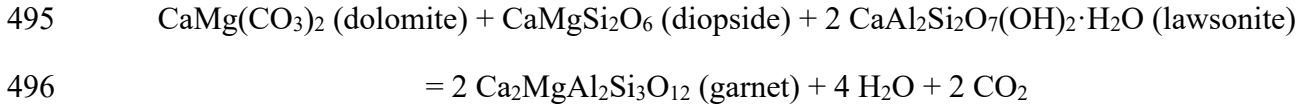
464 Core samples of the sole have  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios that are similar to Indian Ocean MORB and  
465 near-ridge Pacific seamounts (Hofmann, 2013), which are systematically lower than the Sr  
466 isotope ratios of the listvenites. Perhaps the metamorphic sole metabasalts are remnants of a  
467 subducted seamount, similar to accreted seamounts along the Cascadia margin of North America  
468 (e.g., Duncan, 1982), which derived from the enriched mantle source of some Indian Ocean  
469 MORB. Alternatively, the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios may have increased during alteration. The stark  
470 difference in  $^{87}\text{Sr}/^{86}\text{Sr}$  between the metamorphic sole and the listvenites indicates that fluids  
471 derived from the sole are not responsible of the  $^{87}\text{Sr}/^{86}\text{Sr}$  enrichment observed in the ultramafic  
472 lithologies in Wadi Mansah.

473 In contrast, age-corrected Sr isotope ratios for Hawasina metasediments underlying the  
474 ophiolite and the metamorphic sole, north and northeast of Hole BT1B, have  $^{87}\text{Sr}/^{86}\text{Sr}$  up to  
475 0.7134 at 96 Ma (Figure 2). The samples with the most radiogenic  $^{87}\text{Sr}/^{86}\text{Sr}$  are clastic sediments  
476 containing minor amounts of carbonates. Hawasina limestones, on the other hand, have lower  
477  $^{87}\text{Sr}/^{86}\text{Sr}$  values, which is consistent with calcite precipitated from seawater that incorporated a  
478 minor, radiogenic clastic component. Based on these observations, the most likely source of the  
479 fluids that formed the listvenites are derived from the Hawasina metasediments.

480 Thermodynamic modeling of fluid-rock reactions (Kelemen et al. 2021) shows that the  
481 characteristic listvenite mineral assemblages – magnesite + quartz – are attained from fluids with  
482 ~20,000 ppm dissolved C for listvenite formation at 100-300 °C and 0.5 to 1 GPa, similar to the  
483 assemblages modeled by Klein and Garrido (2011) at lower pressures. Such high dissolved  
484 carbon contents are impossible to attain by congruent dissolution of pure calcite in aqueous  
485 fluids at these P-T conditions (Kelemen & Manning, 2015), which rules out silicate-poor  
486 limestones such as those from the continental margin as a carbon source. On the other hand,  
487 metamorphic devolatilization of rocks composed of silicate-carbonate mixtures can produce C-  
488 rich fluids at temperatures above 400 °C and low to moderate pressures, depending on the rock  
489 composition. Thus, we infer that the fluids that formed the listvenites derived from  
490 devolatilization of subducting metasediments. During prograde subduction metamorphism, calc-  
491 silicate rocks, containing both clastic and carbonate components, undergo extensive  
492 devolatilization at 2-3 GPa and 500 to 700°C (Gorman et al., 2006; Stewart & Ague, 2020) and



493 lose significant amounts of their CO<sub>2</sub> in this PT range due to reactions similar to the simplified  
494 reaction:



497 In contrast, carbonate-rich compositions (limestone, dolomite, marble) are predicted to  
498 retain most of their CO<sub>2</sub> during subduction (e.g, Kerrick & Connolly, 2001; Stewart & Ague,  
499 2020).

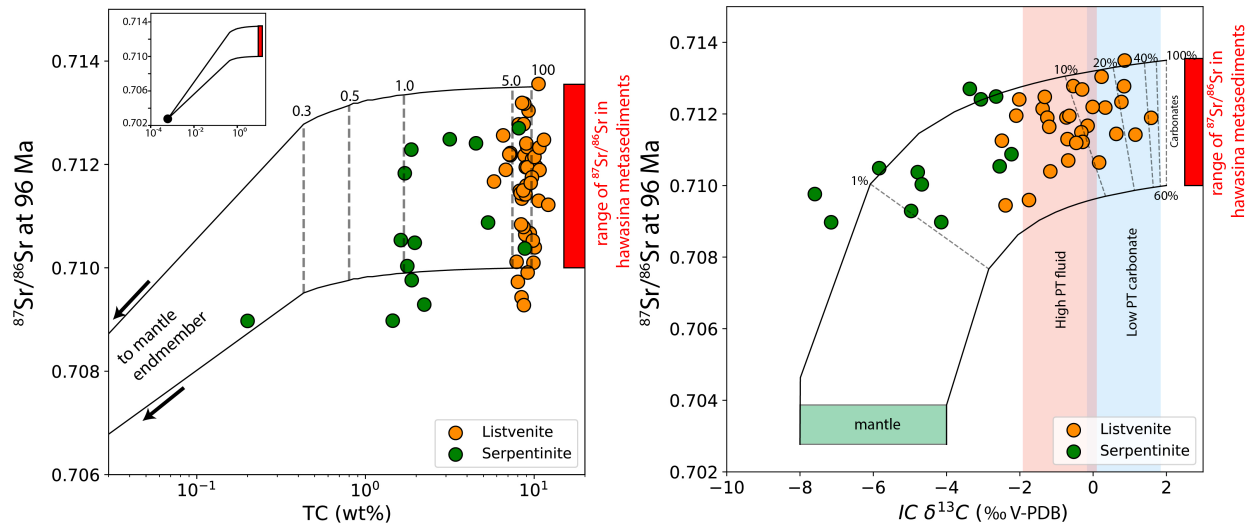
500 These decarbonation reactions are possible for both the metamorphic sole and Hawasina  
501 sediments. The modelling approach of Kelemen et al. (2021) suggests that fluids derived from  
502 the sole and clastic Hawasina metasediments will have enough dissolved carbon to react with  
503 peridotite and result in magnesite-quartz rocks, the characteristic listvenite assemblage.  
504 However, if fluids derived from the sole are the source of the carbon, admixing of a different  
505 fluid derived from Hawasina metasediments is required to explain the <sup>87</sup>Sr/<sup>86</sup>Sr measured in the  
506 listvenites. On the other hand, Kelemen et al. show that fluids derived from a rock with  
507 composition analogous to Hawasina OM20-17 contain enough dissolved carbon for  
508 serpentinization and carbonation of peridotite with increasing water/rock (W/R) ratios. This,  
509 combined with the radiogenic <sup>87</sup>Sr/<sup>86</sup>Sr composition of OM20-17, suggests that a single fluid  
510 could be responsible for carbonation of the peridotites. We test this scenario using the reaction  
511 path proposed by Kelemen et al. (2021). Their model generates a fluid with ~14,000 ppm carbon  
512 at 400°C and 1GPa from decarbonation of a rock with composition such as OM20-17. Under  
513 these modelled conditions (400 °C, 1 GPa) a lithology like OM20-17 will lose ~65% of its  
514 carbon, other Hawasina samples require less decarbonation to produce fluids capable of  
515 carbonating the peridotites. If this fluid is then cooled to 200°C and depressurized to 0.5GPa  
516 without losing much of its carbon before reacting with peridotites, listvenites are predicted to  
517 form at water/rock (W/R)~100.

518 At temperatures greater than ~ 300°C, dissolved CO<sub>2</sub> in aqueous fluids have δ<sup>13</sup>C values  
519 that are higher than those of co-existing calcite and dolomite (Chacko et al., 1991; Deines, 2004;  
520 Horita, 2014). Thus, for example, fluid in equilibrium with calcite with δ<sup>13</sup>C between -5.5‰ to -  
521 3.5‰ (as in Hawasina clastic metasediments) would contain dissolved CO<sub>2</sub> with δ<sup>13</sup>C of -1.9‰  
522 to 0.1‰ at 400°C. At lower temperatures, like those estimated for listvenite formation in the  
523 Samail ophiolite, calcite and dolomite have δ<sup>13</sup>C higher than co-existing fluids. Thus, dolomite  
524 and magnesite in equilibrium with fluids with δ<sup>13</sup>C of -1.9‰ to 0.1‰ would have δ<sup>13</sup>C in the  
525 range of -0.2‰ to 1.8‰, similar to the δ<sup>13</sup>C observed in the listvenites from Hole BT1B and the  
526 surrounding outcrops.

527 Figure 5a illustrates that the listvenites and serpentinites from Hole BT1B lie along the  
528 reaction path used by Kelemen et al., (2021). In our calculations, we assume that reacting fluids  
529 enriched in Sr (250 ppm), with <sup>87</sup>Sr/<sup>86</sup>Sr values at 96 Ma that are similar to those of clastic  
530 Hawasina metasediments (0.7110 to 0.7135), reacted with the peridotite with mantle-like  
531 <sup>87</sup>Sr/<sup>86</sup>Sr (0.7027±0.0011). In the model serpentinites plot at low W/R between 1 to 5 while  
532 listvenites require significantly more fluid at W/R between 5-100. The listvenites and  
533 serpentinites follow a mixing trend (Figure 5b) between mantle peridotite like compositions

534 (Sr=1.5 ppm,  $^{87}\text{Sr}/^{86}\text{Sr}=0.7027\pm 0.0011$ , C=680ppm and  $\delta^{13}\text{C}=-6.0\pm 2.0\%$ ) and low temperature  
 535 carbonates that crystallized from a fluid produced by high-pressure, high-temperature  
 536 devolatilization with Sr isotope ratios in the range of the Hawasina clastic metasediments  
 537 (Sr=250ppm,  $^{87}\text{Sr}/^{86}\text{Sr}\sim 0.7100$  to 0.7135) and fractionate carbon isotopes as described above.  
 538 These trends together suggest that the  $\text{CO}_2$ -bearing aqueous fluids that formed the listvenites  
 539 from Hole BT1B and surrounding outcrops were derived by devolatilization of calc-silicate  
 540 metasediments, with  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $\delta^{13}\text{C}$  similar to clastic sediments in the Hawasina Formation  
 541 along a subduction zone geotherm. These fluids migrated updip to lower pressures and  
 542 temperatures to form the listvenites.

543



544

545 **Figure 5.** (left) Total carbon (wt%) vs.  $^{87}\text{Sr}/^{86}\text{Sr}$  at 96 Ma. Black lines are reaction paths of  
 546 carbon rich fluid reacting with peridotite at 200°C and 0.5GPa at variable water/rock (dashed tie  
 547 lines) from Kelemen et al. (2021). The reacting fluid is assumed to have  $^{87}\text{Sr}/^{86}\text{Sr}$  values of  
 548 0.7100 to 0.7135 like those of clastic Hawasina metasediments. (right) Inorganic carbon  $\delta^{13}\text{C}$  vs.  
 549  $^{87}\text{Sr}/^{86}\text{Sr}$  at 96 Ma. Black lines are mixing lines between mantle and carbonate minerals  
 550 precipitated from a metamorphic fluid. The fluid has the same carbon concentration as the  
 551 reaction path on the figure 5a and the  $\delta^{13}\text{C}$  fractionation between the carbonate and the fluid are  
 552 described in the body of the text, with values for high and low PT shown as red and blue boxes  
 553 respectively. We assume that the fluid has the Sr isotope ratios of Hawasina clastic  
 554 metasediments.  $^{87}\text{Sr}/^{86}\text{Sr}$  values for the depleted mantle are from mid-ocean-ridge-basalt  
 555 (MORB) from Hofmann (2013) . Mantle  $\delta^{13}\text{C}$  is from Deines (2002).

## 556 5 Conclusions

557 Listvenites and spatially associated serpentinites from Hole BT1B and surrounding  
 558 outcrops that replace residual mantle peridotites from the base of the Samail ophiolite have Sr  
 559 isotope ratios that are more radiogenic than their peridotite protoliths, Cretaceous seawater,  
 560 modern seawater, groundwater in the ophiolite, and the underlying metamorphic sole. We  
 561 suggest that the radiogenic Sr isotope component was transported via carbon-rich aqueous fluid  
 562 that reacted with the peridotite to form the listvenites and serpentinites. The  $^{87}\text{Sr}/^{86}\text{Sr}$  values of

563 this component resemble those of calcite-bearing clastic sediments in the Hawasina Formation  
564 underlying the ophiolite. However, the fluid must have contained higher dissolved carbon  
565 contents than feasible for congruent dissolution of pure calcite at < 2 GPa and/or < 550°C. Thus,  
566 we hypothesize that this fluid was derived by devolatilization of carbonate- and silicate bearing  
567 meta-sediments akin to the Hawasina clastic metasediments at 0.5 to 2.3 GPa and 400 to 700°C  
568 in the subduction zone (Searle et al., 1994). This fluid then migrated updip to react with hanging  
569 wall peridotite at <1 GPa and <250°C, forming the listvenites and serpentinites. Carbon isotope  
570 fractionation during high temperature devolatilization followed by low temperature carbonate  
571 precipitation during the reaction with peridotite likely controlled the isotopic characteristics of  
572 the listvenites and the serpentinites with  $\delta^{13}\text{C}$  from -10.6‰ to 1.92‰ and  $^{87}\text{Sr}/^{86}\text{Sr}$  from 0.7090  
573 to 0.7145.

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594 Authors declare no conflict of interest.

595 All data is in the process of being approved by Pangea repository:  
596 <https://issues.pangaea.de/browse/PDI-30281> and <https://issues.pangaea.de/browse/PDI-30282>

597

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