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## Listvenite formation during mass transfer into the leading edge of the mantle wedge: Initial results from Oman Drilling Project Hole BT1B

Initial results from Oman Drilling Project Hole BT1B
 Peter B. Kelemen<sup>1</sup>, Juan Carlos de Obeso<sup>2</sup>, James A. Leong<sup>1</sup>, Marguerite Godard<sup>3</sup>, Keishi Okazaki<sup>4</sup>,
 Alissa J. Kotowski<sup>5</sup>, Craig E. Manning<sup>6</sup>, Eric T. Ellison<sup>7</sup>, Manuel D. Menzel<sup>8</sup>, Janos L. Urai<sup>8</sup>, Greg
 Hirth<sup>9</sup>, Matthew Rioux<sup>10</sup>, Daniel F. Stockli<sup>11</sup>, Romain Lafay<sup>3</sup>, Andreas M. Beinlich<sup>12</sup>, Jude A. Coggon<sup>13</sup>,
 Nehal H. Warsi<sup>14</sup>, Jürg M. Matter<sup>13</sup>, Damon A.H. Teagle<sup>13</sup>, Michelle Harris<sup>15</sup>, Katsuyoshi
 Michibayashi<sup>16</sup>, Eiichi Takazawa<sup>17</sup>, Zaher Al Sulaimani<sup>18</sup> and the Oman Drilling Project Science Team

Michibayashi<sup>16</sup>, Eiichi Takazawa<sup>17</sup>, Zaher Al Sulaimani<sup>18</sup> and the Oman Drilling Project Science Team
 11: Lamont Doherty Earth Observatory, Columbia University, Palisades NY 10964, <u>peterk@LDEO.columbia.edu</u> 2: Dept. of
 Geosciences, University of Calgary 3: Géosciences Montpellier, Université de Montpellier, CNRS 4: Kochi Institute for Core
 Sample Research, JAMSTEC 5: Dept. of Earth & Planetary Sciences, McGill University 6. Dept of Earth, Planetary & Space
 Sciences, University of California, Los Angeles 7. Dept. of Geological Sciences, University of Colorado, Boulder 8. Tectonics
 & Geodynamics, RWTH Aachen University 9. Dept. of Earth, Environmental and Planetary Sciences, Brown University 10.
 Dept. of Earth Science, University of California, Santa Barbara 11. Dept. of Geological Sciences, University of Texas, Austin
 Dept. of Earth Science, University of Bergen ... 13. Dept. of Ocean & Earth Science, National Oceanography Centre
 Southampton 14. Alara Resources Ltd., Oman 15. School of Geography, Earth and Environmental Sciences, University of
 Plymouth ...16. Dept. of Earth & Planetary Sciences, Nagoya University 17. Dept. of Geology, Niigata University & VERC IMG
 JAMSTEC 18. Oman Water Society & Middle East Desalination Research Center

#### Abstract

This paper provides an overview of research on core from Oman Drilling Project Hole BT1B and the 23 24 surrounding area, plus new data and calculations, constraining processes in the Tethyan subduction 25 zone beneath the Samail ophiolite. The area is underlain by gently dipping, broadly folded layers of allochthonous Hawasina pelagic sediments, the metamorphic sole of the Samail ophiolite, and 26 27 Banded Unit peridotites at the base of the Samail mantle section. Despite reactivation of some faults during uplift of the Jebel Akdar and Saih Hatat domes, the area preserves the tectonic "stratigraphy" 28 29 of the Cretaceous subduction zone. Gently dipping listvenite bands, parallel to peridotite banding and to contacts between the peridotite and the metamorphic sole, replace peridotite at and near the basal 30 31 thrust. Listvenites formed at less than 200°C and (poorly constrained) depths of 25 to 40 km by 32 reaction with CO2-rich, aqueous fluids migrating from greater depths, derived from devolatilization of subducting sediments analogous to clastic sediments in the Hawasina Formation, at 400-500°. Such 33 processes could form important reservoirs for subducted CO<sub>2</sub>. Listvenite formation was accompanied 34 by ductile deformation of serpentinites and listvenites - perhaps facilitated by fluid-rock reaction - in a 35 process that could lead to aseismic subduction in some regions. Addition of H<sub>2</sub>O and CO<sub>2</sub> to the 36 37 mantle wedge, forming serpentinites and listvenites, caused large increases in the solid mass and 38 volume of the rocks. This may have been accommodated by fractures formed as a result of volume 39 changes, mainly at a serpentinization front. 40 Plain language summary 41 42 This paper reports initial results from study of core from Oman Drilling Project Hole BT1B and the 43 surrounding area. It provides insights into subduction zone processes, including large fluxes of 44 recycled CO<sub>2</sub> from subducting sediments into the leading edge of the mantle wedge, and surprisingly low temperature ductile deformation at less than 200°C. Recycling of CO<sub>2</sub> via carbon mineralization in 45 the hanging wall of subduction zones may produce an important, lithospheric reservoir in the global 46 47 carbon cycle. Ductile deformation of serpentinite, and during or after transformation of peridotite to

48 listvenites (mixtures of carbonates and opal or quartz) could explain aseismic subduction atop some

49 subduction zones.

50	1. Introduction
51	
52	Oman Drilling Project (OmanDP) Hole BT1B at 23.364374°N, 58.182693°E, about 12 km southeast of
53	the town of Fanjah in the Sultanate of Oman, sampled serpentinized peridotites and listvenites (fully
54	carbonated peridotites, Halls & Zhao 1995) at the base of the Samail ophiolite, the basal fault of the
55	ophiolite, and the underlying metamorphic sole, with the intention of investigating mass transfer and
56	deformation in the "leading edge of the mantle wedge" overlying a Tethyan subduction zone. Study of
57	outcrops along the contact between the sole and the peridotite can illuminate processes in and above
58	a subduction zone that are generally inaccessible to direct observation. Of particular interest are (a)
59	the source of footwall fluids, (b) the nature and mechanism of fluid transport along the thrust fault and
60	into the mantle wedge, (c) chemical, mineralogical and rheological modification of the hanging wall by
61	reaction with footwall fluids, and (d) the mechanisms of subduction zone deformation.
62	
63	Hole BT1B was drilled using cylindrical diamond bits and wireline core retrieval, from March 7 to
64	March 23, 2017. Core recovery was ~ 100% throughout the Hole. Core was shipped to Japan and
65	loaded onto Drilling Vessel Chikyu, where the OmanDP Science Team performed analyses closely
66	following protocols established by the various incarnations of the Ocean Drilling Program (currently,
67	the International Ocean Discovery Program, IODP). Detailed core descriptions, together with drilling
68	history and some background information (Kelemen et al 2020b, Kelemen et al 2020c) are available
69	online at <a href="http://publications.iodp.org/other/Oman/VOLUME/CHAPTERS/113_BT1.PDF">http://publications.iodp.org/other/Oman/VOLUME/CHAPTERS/113_BT1.PDF</a>
70	
71	This paper reports new mapping and structural observations, mineral analyses and modeling,
72	together with a summary of prior observations. It is intended to provide interpretive context for more
73	detailed studies of core from Hole BT1B and the geology of the surrounding region. Such studies in
74	this Special Issue of the Journal of Geophysical Research include those focused on textural and
75	petrologic data for listvenites (Beinlich et al 2020) and the metamorphic sole (Kotowski et al 2021),
76	volume changes during serpentinization (Malvoisin et al 2020), major and trace element geochemistry
77	with relevance to subduction zone mass transfer (Godard et al 2021, Okazaki et al 2021), Mg-, Sr-
78	and C-isotope geochemistry constraining the source of CO2-bearing fluids and the fluid-rock reaction
79	process (de Obeso et al 2021a, de Obeso et al 2021b) and deformation of the listvenites (Menzel et al
80	2020, Menzel et al 2021).
81	
82	Section 2 provides geological context for observations of core and surrounding outcrops. Section 3
83	summarizes methods used to produce results presented for the first time in this paper. Section 4
84	reports new geological field observations and analytical data, and reviews key results from Kelemen
85	et al. (2020b) in context. Section 5 discusses the pressure, temperature and timing of listvenite
86	formation, the nature and source of the fluids that transformed mantle peridotite into serpentinite and
87	listvenite, the chemical and mechanical processes during these transformations, and deformation of

- 88 altered mantle peridotite immediately above a paleo-subduction zone beneath the "leading edge of
- the mantle wedge".

90	
91	It is hoped that this paper, and the other papers in this Special Issue, will provide a starting point for
92	future investigations of this unique and important site. In this context, readers should be aware that
93	the archive half of the core is currently stored at Petroleum Development Oman where it is available
94	for viewing, the working half of the core is stored at the American Museum of Natural History, where it
95	can be sampled upon request to the Museum, and a huge volume of data from shipboard visual core
96	observations and analytical data is available to anyone at
97	http://publications.iodp.org/other/Oman/OmanDP.html , https://www.icdp-
98	online.org/projects/world/asia/oman/, and other sites that can be accessed from there.
99	
100	2. Geological setting
101	
102	2.1 Regional geologic context
103	
104	The Samail ophiolite is composed of oceanic crust formed at a submarine spreading center above a
105	subduction zone. The crustal thickness and composition of the ophiolite is similar to the geophysically
106	and geologically constrained characteristics of fast-spreading, Pacific oceanic crust, with a few km of
107	submarine lavas and sheeted dikes overlying a thicker, gabbroic lower crust (e.g., Christensen &
108	Smewing 1981, Coleman & Hopson 1981, Nicolas et al 1996) However, the lavas have a trace
109	element "subduction signature" (Alabaster et al 1982, Pearce et al 1981, Pearce & Peate 1995), and
110	parental, mantle-derived magmas appear to have contained 0.2 to 2 wt% H <sub>2</sub> O, substantially more
111	than in primitive mid-ocean ridge basalts (MacLeod et al 2013). Beneath the crustal section of the
112	ophiolite, residual mantle peridotites and tabular dunites record polybaric decompression melting,
113	melt extraction, and focused transport of basaltic melt upward to form the crust (Braun & Kelemen
114	2002, Godard et al 2000, Kelemen et al 2000, Kelemen et al 1995, Monnier et al 2006).
115	
116	2.2 Lithologies at the base of the Samail ophiolite
117	
118	Here we describe the units above and below the basal fault of the Samail ophiolite. We begin with a
119	review of work on the metamorphic sole. Then, we describe the overlying Banded Unit at the base of
120	the ophiolite's mantle section. Finally, we briefly outline the nature of the sedimentary rocks beneath
121	the metamorphic sole.
122	
123	Beneath the mantle section of the ophiolite, and above allocthonous sediments of the Hawasina
124	Formation, are discontinuously exposed lenses of a "metamorphic sole". Peak pressures and
125	temperatures in the sole generally record hot subduction conditions at temperatures up to 700-900°C
126	(Cowan et al 2014, Ghent & Stout 1981, Hacker & Gnos 1997, Searle & Cox 1999, Searle & Cox
127	2002, Searle et al 1980, Searle & Malpas 1980, Searle & Malpas 1982, Soret et al 2017) though at
128	BT1B peak temperatures are 450-550°C (Kotowski et al 2021). Apparently – based on published data
129	and calculations - the sole records a broad range of peak pressures from a possible lower limit of 200

MPa (Ghent & Stout 1981) or 800 MPa (Soret et al 2017) to a possible upper limit of 1400 MPa
(Cowan et al 2014, Searle & Cox 2002). Kotowski et al. (2021) report that the sole in core from Hole
BT1B records a peak pressure in the range of 800 to 1200 MPa.

133

The sole contains metasediments and meta-volcanic rocks - including submarine pillow lavas - with 134 135 the major element compositions of mid-ocean ridge basalts (MORB) as well as alkali basalts and more evolved andesites and dacites (Alabaster et al 1982, Ernewein et al 1988, MacLeod et al 2013, 136 Pearce et al 1981, Searle et al 1980). In core from BT1B, alkaline metabasalt compositions in the sole 137 138 (Godard et al 2021, Kelemen et al 2020c) are unlike the magmas that formed the crust of the Samail ophiolite. In the ophiolite, the structurally lowest, "Geotimes" or "V1" lavas are similar to normal mid-139 ocean ridge basalts, though they probably were hydrous and they contain a hint of an arc trace 140 element signature (Alabaster et al 1982, Ernewein et al 1988, MacLeod et al 2013, Pearce et al 141 1981). Their composition, and that of dunite conduits for transport of primitive melts parental to V1 142 through the shallow mantle, is consistent with formation of the gabbroic lower crust in the Samail and 143 Wadi Tayin massifs of the ophiolite from primitive V1 magmas (Braun & Kelemen 2002, Kelemen et al 144 1997, Kelemen et al 1995). The overlying "Lasail" or "V2" lavas in the ophiolite are incompatible-145 146 element-depleted, with a stronger subduction signature (Alabaster et al 1982, Ernewein et al 1988, 147 MacLeod et al 2013, Pearce et al 1981) and include boninites (e.g., Ishikawa et al 2002) as well as tholeiitic basalts, andesites and dacites. Neither V1 nor V2 lavas are similar to the alkali basalt 148 compositions in the sole. The present day <sup>87</sup>Sr/<sup>86</sup>Sr ratios in the alkali basalts in core from BT1B range 149 from 0.704 to 0.706 (de Obeso et al 2021a), more radiogenic than MORB. Though their Sr isotope 150 ratio may have been modified by alteration, one possibility is that the alkali basalts are remnants of 151 152 subducted seamounts with elevated <sup>87</sup>Sr/<sup>86</sup>Sr, similar to accreted seamounts along the Cascadia 153 margin of North America (e.g., Duncan 1982).

154

In addition to metabasalts, regionally the sole contains metasediments, and "exotic limestones", all
incorporated by Searle and Malpas (1980) in the "Haybi Formation". However, in this paper we
informally group the Haybi Formation into an undifferentiated metamorphic sole unit. In the sole
sampled by drill core from Hole BT1B, metasediments are clearly distinguishable from the
metabasalts based on texture, but some of them are compositionally similar to the metabasalts,
perhaps reflecting a volcanoclastic origin, whereas others grade into somewhat odd, low-SiO<sub>2</sub>,
muscovite-bearing lithologies (Godard et al 2021, Kelemen et al 2020b, Kotowski et al 2021).

162

Above the sole – where it is present – and elsewhere at the base of the ophiolite, the lower few km of the mantle section contains easily visible, meter to 10-meter scale, parallel bands of dunite,

harzburgite and (rare) lherzolite, informally known as the Banded Unit (Godard et al 2000, Khedr et al

166 2013, Khedr et al 2014, Linckens et al 2011, Lippard et al 1986, Prigent et al 2018a, Prigent et al

167 2018b, Takazawa et al 2003, Yoshikawa et al 2015). The lithological contacts in this unit are sharp.

168 They have low angle dips with respect to the paleo-seafloor, the crust-mantle transition zone, and the

basal fault that juxtaposes mantle peridotite with the metamorphic sole. Mylonitic shear zones are

- present in the Banded Unit, with textures recording deformation at 700–1,000°C (Boudier et al 1988,
   Herwegh et al 2016, Linckens et al 2011, Prigent et al 2018a). Thus, there is evidence for high strain
- ductile deformation and transposition of layering at the base of the mantle section, which might have
- accommodated substantial thinning (Prigent et al 2018a, Soret et al 2017). Similarly, there is evidence
- 174 for thinning of the units below the ophiolite (Grobe et al 2018, Grobe et al 2019).
- 175

While some of the range in temperature and pressure estimates from the sole and the Banded Unit 176 may be due to analytical and methodological uncertainty, or incomplete preservation and sampling of 177 178 the highest-grade rocks, some may be due to temporal and spatial variability in peak metamorphic conditions. Moreover, the temperature record may be biased toward peak conditions, rather than later 179 180 cooling. At the initiation of subduction near a spreading ridge, hot metamorphic rocks from the footwall may be accreted to the hot base of the newly formed mantle wedge, whereas as subduction zones 181 grow colder and develop a steady-state thermal structure, cold dense lithologies in the footwall may 182 be subducted (Agard et al 2016, Soret et al 2017). If so, the metamorphic sole in Oman may record 183 the anomalously high temperatures of subduction initiation, and not the lower temperatures of 184 185 evolution toward a steady-state subduction zone geotherm. The relatively low temperatures recorded 186 by the sole in BT1B core could represent a point along this cooling path (Kotowski et al 2021). 187

A close correspondence between 96 to 95 Ma igneous ages in the crust, and both <sup>40</sup>Ar/<sup>39</sup>Ar and zircon U/Pb ages of metamorphic rocks along the basal thrust (ca. 96-94 Ma), indicates that thrusting of the Samail ophiolite over adjacent oceanic crust and nearby pelagic sedimentary units began during formation of igneous crust in the ophiolite (e.g. , Garber et al 2020, Hacker & Gnos 1997, Hacker & Mosenfelder 1996, Hacker et al 1996, Rioux et al 2012, Rioux et al 2013, Rioux et al 2016, Rioux et al 2021b, Stanger 1985, Styles et al 2006, Tilton et al 1981, Warren et al 2005) or perhaps even earlier (Guilmette et al 2018).

195

The base of the metamorphic sole is truncated by a fault contact with autochthonous, low-grade 196 metasediments of the Hawasina Formation, composed of pelagic clastic units interlayered with 197 limestones (Béchennec et al 1990, Béchennec et al 1988). Relatively high, age corrected <sup>87</sup>Sr/<sup>86</sup>Sr 198 199 ratios in the clastic units suggest that they are distal sediments derived from erosion of continental crust (de Obeso et al 2021a, Weyhenmeyer 2000), while carbonate units record Sr isotope ratios 200 similar to those of Mesozoic seawater (Weyhenmeyer 2000, Wohlwend et al 2017). These 201 202 sedimentary units were thrust over Mesozoic to Proterozoic rocks of the Arabian continental margin, 203 forming a "rumpled rug" between the autochthon and the ophiolite throughout northern Oman and the eastern United Arab Emirates. 204

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- 206

207

### 2.4 History of the basal thrust of the Samail ophiolite

208 Where the metamorphic sole is preserved, its tectonic contact with the mantle section of the ophiolite 209 represents a paleo-subduction zone at the base of the ophiolite, which consumed several hundred

- kilometers of Tethyan basin and then continental crust before coming to rest on the Arabian
- continental margin (Béchennec et al 1990, Béchennec et al 1988, Breton et al 2004, Cooper 1988,
- Ninkabou et al 2021, Searle & Robertson 1990, van Hinsbergen et al 2019). During this time, the
- 213 overlying peridotite was "the leading edge of the mantle wedge".
- 214

215 During coeval metamorphism of the sole and igneous accretion of the crust in the ophiolite, the relative locations of these two units are uncertain. Primitive V1 magmas in the ophiolite, with 216 compositions almost indistinguishable from mid-ocean ridge basalts (MORB), crystallized to form 217 218 most of the crust, particularly in the southern Wadi Tayin and Samail massifs that were the site of of the OmanDP boreholes. Like MORB worldwide, these magmas probably formed via polybaric 219 decompression melting over a depth interval of 75 km or more (e.g., Allegre et al 1973, Asimow et al 220 2004, Bottinga & Allegre 1973, Klein & Langmuir 1987, McKenzie & Bickle 1988). Thus, the 221 222 metamorphic sole (recording peak depths less than 40 km) must not have been directly beneath the 223 spreading center during crustal formation and metamorphism. Instead, the ophiolite section and the

metamorphic sole were juxtaposed after crust formation and after peak metamorphism in the sole.

226 Moreover, the structural thickness of the Samail ophiolite measured perpendicular to the paleo-227 seafloor and/or the crust/mantle transition zone, and including both crust and mantle sections, never exceeds ~ 20 to 30 km (Boudier & Coleman 1981, Nicolas et al 2000), corresponding to a pressure 228 less than or equal to  $\sim$  800 MPa, beneath  $\sim$  7 km of crust (on average, Nicolas et al 1996) and up to  $\sim$ 229 230 20 km of fresh mantle peridotite. Thus, the structural thickness of the ophiolite appears to be at least 10 km less than the depth inferred from the high end of the pressure range recorded by the sole 231 232 (1300 or 1400 MPa, ~ 40 to 45 km). In addition, there is no evidence for widespread, tectonic thinning 233 of the crustal section of the ophiolite, either by faulting or ductile deformation.

234

How can these data be reconciled? One possibility is that the peak pressures inferred for the sole are 235 imprecise, and/or that they represent tectonic "overpressures" that don't correspond closely with 236 237 depth (Garber et al 2020). In this interpretation, the metamorphism of the sole could have occurred at  $\sim$  800 MPa, corresponding to the current structural thickness of the ophiolite. A second possibility, as 238 239 noted above, is that the base of the mantle section of the ophiolite underwent tectonic thinning, perhaps via simple shear (Prigent et al 2018a, Soret et al 2017). A third alternative is that some of the 240 241 lenses of the metamorphic sole - those recording the highest pressures - migrated updip to reach 242 their current structural position, ~ 25 km below the paleo seafloor. Indeed, upward transport of buoyant footwall lithologies during subduction is recorded in many preserved collisional orogens (e.g., 243 Chemenda et al 2000). If so, updip migration of the sole with respect to the overlying ophiolite must 244 245 have occurred after peak metamorphism but during Tethyan subduction, prior to emplacement of the ophiolite and the sole over the allochthonous Hawasina sediments. Finally, of course, it is possible 246 247 that the current juxtaposition of the sole and the base of the ophiolite may be explained as the result of combinations of these three alternatives. 248

250 Subsequent events have affected the wedge, the sole, the underlying sedimentary units, and the faults between them (Grobe et al 2018, Grobe et al 2019). The end of subduction and emplacement 251 of the ophiolite over allochthonous Hawasina sediments and the autochthonous, Arabian continental 252 margin was followed by subaerial erosion, and then by a marine transgression marked by deposition 253 of shallow water limestones at ~74 Ma (Al Khirbash 2015, Alsharan & Nasir 1996, Bailey 1981, 254 Hansman et al 2017a, Nolan et al 1990, Schlüter et al 2008, Wyns et al 1992). Then, large scale uplift 255 deformed all these units, forming the gigantic Jebel Akdar and Saih Hatat domes, cored by 256 Proterozoic continental crust (e.g., Glennie et al 1973, Glennie et al 1974b). Saih Hatat uplift and 257 258 cooling started at about 60 Ma, if not earlier (e.g., Grobe et al 2018, Grobe et al 2019, Hansman et al 2017b). Doming reactivated and cut the basal thrust of the ophiolite in normal faults and shear zones. 259 Some of these younger faults juxtaposed mantle peridotite from the ophiolite with the allochthonous 260 Hawasina sedimentary rocks, and even with the autochthonous platform carbonates of the Arabian 261 continental margin, along tectonic contacts where the metamorphic sole is no longer present. 262 263 However, despite these complexities, we reiterate that – where it is parallel to banding and foliation – 264

the contact between the metamorphic sole and the overlying Banded Unit at the base of the Samail ophiolite mantle section represents the basal thrust of the ophiolite. In this paper, when we refer to the "basal thrust", we are referring to contacts that preserve these characteristics. In turn, though of course there can have been imbrication and/or subduction erosion, the basal thrust of the ophiolite represents the locus of 100's of kilometers of subduction of oceanic crust, overlying sediments and, ultimately, the Arabian continental margin.

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### 2.5 Geology of MoD Mountain

Hole BT1B is on the north side of the wide Wadi Mansah, southeast of "Ministry of Defense
Mountain", (MoD Mountain), which is informally named for the military firing range on the south side of
Wadi Mansah. The MoD Mountain region is close to the saddle between the northeastward plunging
end of the Jebel Akdar massif, to the west, and the westward plunging Saih Hatat massif to the
northeast (Figure 1).

279

This region preserves a gently folded, tectonic "stratigraphy" (Figures 2, 3 and 4). The lowest 280 exposed units in this sequence, northeast of Hole BT1B, are diagenetically-altered sedimentary rocks, 281 282 mostly clastic shales and slates with a few meter to 10 meter scale intercalations of limestone and minor lenses of metavolcanic rocks. These are parts of the Hawasina Formation (Béchennec et al 283 1990, Béchennec et al 1988). Overlying the Hawasina Formation is one of the most aerially extensive 284 285 outcrops of the metamorphic sole in Oman and the UAE (e.g., Figure 1 and geologic maps in Soret et 286 al 2017, Wilde et al 2002). The outcrop area is unusually large in this region because the unit is 287 regionally flat lying, though broadly folded.

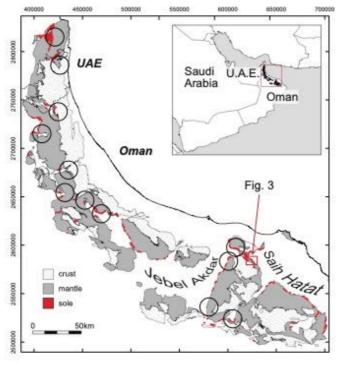


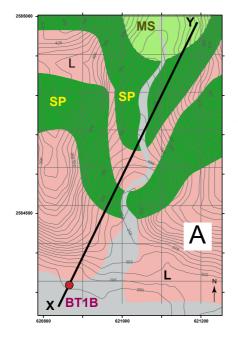
Figure 1: Outcrop area of the Samail ophiolite in Oman and the UAE, based on Nicolas et al. (2000). Metamorphic sole, in red. Black circles indicate approximate location of listvenites (Nasir et al 2007, Stanger 1985, Wilde et al 2002). Red rectangle indicates the approximate location of Figure 3, a geologic map of MoD Mtn and vicinity, which was the site of Oman DP Hole BT1B, and the focus of this paper.



290

Figure 2: Photograph of the west end of the MoD Mountain ridge, from the SW, looking NE, showing 291 bands of listvenite (rusty orange) parallel to contacts between partially serpentinized harzburgite 292 (brown) and dunite (tan) comprising the "Banded Unit" at the base of the Samail ophiolite mantle 293 294 section. Greenish metabasalts and metasediments of the metamorphic sole underlying the ophiolite are exposed along indistinct ridge in lower left. The listvenite band at top right is more than 100 m 295 thick and extends for 1.5 kilometers along the length of the summit ridge. The listvenite band in the 296 center of the photo is 15-20 m thick. Photo taken with a telephoto lens from ~ 23.35°N, 58.17°E, along 297 azimuth ~ 030°. 298

299



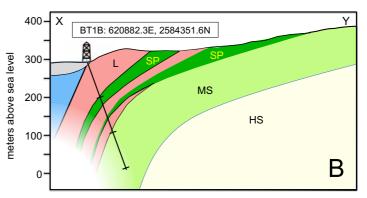


Figure 3: Map and cross section of the area NNE of Oman Drilling Project Site BT1. Yellow, HS: Hawasina sediments; light green, MS: metamorphic sole; rusty red, L: listvenite; dark green, SP: serpentinized peridotite; blue: inferred gabbro, based on outcrops south of Wadi Mansah; grey: gravels of Wadi Mansah. UTM coordinates in meters. Cross section has no vertical exaggeration. Map and cross-section modified from Figures F1 and F2 in Kelemen et al. (2020b)

302 Above the metamorphic sole in this gently folded "stratigraphy" is partially serpentinized peridotite, composed of distinctive, banded alternations of dunite, harzburgite and lherzolite on a scale of meters 303 304 to tens of meters, characteristic of the "Banded Unit" at the base of the ophiolite (Boudier et al 1988, 305 Boudier & Coleman 1981, Lippard et al 1986, Searle & Cox 2002). Indeed, partially serpentinized 306 mantle peridotites on MoD Mountain, northeast of Hole BT1B have compositions similar to the 307 Banded Unit elsewhere, and distinct from shallower, residual mantle peridotites of the Samail ophiolite emantle section, as discussed in Section 5.5, below. The presence of the Banded Unit overlying the 308 309 metamorphic sole is another indication that MoD Mountain area exposes the basal thrust of the 310 ophiolite - the paleo-subduction zone - together with the overlying mantle wedge.

311

In some localities, peridotites at the base of the Samail ophiolite mantle section have undergone 312 100% carbonation at ~ 100-200°C to form "listvenites" (Beinlich et al 2020, Falk & Kelemen 2015, 313 Glennie et al 1974a, Nasir et al 2007, Stanger 1985, Wilde et al 2002), in which all Mg and Ca are in 314 315 carbonate minerals, and silica derived from olivine and pyroxene has formed quartz or chalcedony. In 316 Oman, such listvenites are most abundant on and around MoD Mountain northeast of Hole BT1B, where they form part of the gently folded tectonic "stratigraphy" we are discussing here. Listvenites on 317 the flanks of MoD Mountain form discontinuous tabular lenses with low angle dips, 10 to 200 meters 318 319 thick, parallel to the basal thrust and to lithological banding in the peridotite. These lenses occur along 320 the basal thrust – between peridotite and the sole – and enclosed within the peridotite, up to 300 321 meters above the sole. Contacts in outcrop between listvenite and the surrounding, partially serpentinized peridotite are marked by strongly foliated, 100% serpentinized zones, 1 to > 20 m thick. 322 323 The lithological banding in the peridotite and the listvenite lenses dip gently south on the south side of 324 325 MoD Mountain, north on the north slopes of the mountain, and then south again along and NE of the

wadi bounding MoD Mountain to the north (Falk and Kelemen 2015 and this paper, Section 4.2).

- 327 Despite later faulting, these structures define a broad anticline with an axis approximately coincident 328 with the summit ridge of MoD Mountain, and a syncline with an axis roughly coincident with the wadi 329 northeast of MoD Mountain. Listvenites form erosion resistant dip slopes and the tops of small buttes
- 330 outlining the folded stratigraphy.
- 331

Listvenites elsewhere in Oman and the UAE are found along the basal thrust, commonly juxtaposed with, or within a few km of, the metamorphic sole and/or the Banded Unit at the base of the Samail mantle section, as at MoD Mountain. In some other outcrops, listvenites form lenses within broad serpentinite mélange zones at the base of the ophiolite (Nasir et al 2007, Stanger 1985). In contrast, listvenites are not found within the peridotite more than a few kilometers away from the basal thrust of the ophiolite.

338

339 A Rb/Sr isochron on listvenite mineral separates (97 Ma ± 29 Ma 2σ, Falk & Kelemen 2015) is similar to the better determined, 96 to 94 Ma U/Pb ages of zircon in the metamorphic sole and igneous crust 340 341 in the ophiolite. Based on the listvenite mineral isochron, there is a 67% probability that the listvenites 342 are older than 82.5 Ma (average  $-1\sigma$ ), and a 93% probability that they are older than 75 Ma (1.5 $\sigma$ ), 343 before the end of subduction. Thus, geological observations (summarized above and in Section 4.2) together with the isochron data indicate that the listvenites formed via transfer of CO2 and other 344 components from subducting material - probably sediments and/or altered lavas - into the leading 345 346 edge of the mantle wedge during Tethyan subduction and ophiolite emplacement. New geological data supporting this are presented in 4.2. Sections 5.6 and 5.7 provide thermodynamic modeling that 347 348 quantifies this hypothesis.

349

350 As noted in Section 2.4, Paleocene to Miocene uplift of the nearby Jebel Akdar and Saih Hatat massifs may have caused reactivation of some older faults, and definitely formed new, younger faults 351 352 in the MoD area. For example, shallow-level gabbros and sheeted dikes are juxtaposed with mantle 353 peridotite and the metamorphic sole on a steep fault parallel to Wadi Mansah, just south of Hole BT1B (Villey et al 1986, Wilde et al 2002). In another possible example, Scharf et al. (2020) report early 354 355 U/Pb formation or cooling ages ( $60 \pm 16$  and  $58 \pm 6$  Ma) of calcite veins that cut listvenite. cataclasite and fault contacts between listvenite and post-emplacement, Late Cretaceous conglomerates. 356 357 358 3. Methods 359 Here we describe methods used for new data presented for the first time in this paper. Supplementary 360 361 Section S2 provides methods for data illustrated in this paper, but previously reported elsewhere.

362 363

3.1 Geological mapping

Geological field work to produce the map and cross-section in Figure 4 spanned short visits over 364 365 about a decade. Contacts and structural measurements were located by GPS at the time of 366 measurement. The topographic profile for the cross section was constructed using Google Earth. 367 368 3.2 Raman spectroscopy 369 Analyses of minerals in thin section and rock slabs were conducted at the Raman Microspectroscopy 370 Laboratory, University of Colorado-Boulder with a Horiba Scientific LabRam HR Evolution Raman 371 372 microscope. Measurements used a 100 mW 532 nm laser, focused through a 50x (0.75 NA) 373 microscope objective onto a ~2 µm spot. The laser power was modulated with neutral density filters to about 15 mW at the sample surface. Multiple (2-10) accumulations were coadded in order to filter 374 spikes and improve signal to noise, and the acquisition time and accumulation number were adjusted 375 376 to yield appropriate data quality. Data processing was performed using LabSpec 6 software (Horiba Scientific), including correction for instrumental artifacts and polynomial baseline fitting/subtraction. 377 Raman mapping was performed using a motorized stage with 2 µm step size, and map datasets were 378 379 fit using classical least-squares fitting with endmember spectra isolated from regions within the map 380 using LabSpec 6 after data processing. 381 382 3.3 Thermodynamic calculations and modeling 383 The speciation and chemical mass transfer code EQ3/6 (Wolery 1992) was used to predict the 384 compositions of coexisting solid and aqueous phases that evolved during interaction between 385 386 representative lithologies from the MoD Mountain area and CO2-bearing fluids. Thermodynamic data 387 for minerals were mostly from Berman (1988). Data for pyrite and pyrrhotite were from Helgeson et al. (1978). For aqueous species, thermodynamic data used in the simulations were calculated using the 388 Deep Earth Water (DEW) model (Huang & Sverjensky 2019, Sverjensky et al 2014) which uses 389 recent experimental and theoretical advances (Facq et al 2016, Pan et al 2013) to expand the 390 391 extended Helgeson-Kirkham-Flowers (HKF) aqueous equation of state (Shock et al 1992, Shock et al 1997) to pressures up to 6.0 GPa. 392 393 The composition of 5 wt% aqueous fluid in equilibrium with a pelitic lithology from the Oman 394 Hawasina Formation at 400 - 600°C and 0.5 to 2.0 GPa at low water/rock ratios was used. 395 396 Specifically, a dilute fluid was equilibrated with the rock composition of sample OM20-17 (de Obeso et al 2021a), containing 0.06 wt% total carbon (Supplementary Table 2). The CaO content of OM20-17 397 was below detection. For the model calculations it was assumed to 0.1 wt%. In addition, the S content 398 399 of this sample has not been measured. For this calculation it was assumed 100 ppm. At these high 400 temperatures and low carbon contents, carbonate minerals are unstable and all carbon in the rock will

- 401 be mobilized into the fluid phase as dissolved CO<sub>2</sub>.
- 402

403 We calculated the outcome of cooling and decompression of the CO<sub>2</sub>-rich fluid from OM20-17, to 100

- 404 300 °C and 0.5 to 2.0 GPa. This had no significant effect on its composition. We then calculated the
- products of reaction between this fluid and average Oman harzburgite (Supplementary Table 2,
- 406 calculated from Godard et al 2000, Hanghoj et al 2010, Monnier et al 2006) at 100 300 °C and 0.5 –
- 407 1.0 GPa, at water:rock ratios ranging from 100 to 1.
- 408

409 As a result of data limitations, solid solutions of precipitating minerals were not considered, as the Berman database lacks properties for most Fe-endmembers of minerals commonly observed in 410 411 listvenites and associated rocks. Thus, for example, the model predicts co-precipitation of pure, 412 endmember magnesite, dolomite and siderite, whereas in listvenite samples we observe Fe-bearing 413 magnesite and dolomite. Among the serpentine polytypes, only chrysotile precipitation can be 414 modeled. We did not include goethite. There are no thermodynamic data for a chromian muscovite component, so this was not included, despite the fact that solid solutions ranging from fuchsite to 415 chromian muscovite are observed in MoD Mountain listvenites (e.g., Falk & Kelemen 2015). 416 417

418 Phase equilibrium calculations constraining the conditions for co-existing graphite (± amorphous 419 carbon compounds) and hematite, and updated calculations for co-existing antigorite and quartz, 420 used (1) Thermocalc (Powell et al 1998), (2) Perple X (https://www.perplex.ethz.ch/) (Connolly 1990, 421 Connolly 2005, Connolly 2009), with the Holland and Powell thermodynamic data for minerals (2003, 1998), and the default equations of state for  $H_2O$ -  $CO_2$  fluids (modified versions of Redlich-Kwong), 422 and (3) SUPCRT (Johnson et al 1992, Zimmer et al 2016) with thermodynamic data for minerals from 423 Helgeson et al. (Helgeson 1985, 1978) or Berman (1988, plus graphite from Helgeson et al.) and 424 425 various equations of state for H<sub>2</sub>O-CO<sub>2</sub> fluids (Shock et al 1992, Shock et al 1997) modified from 426 Helgeson et al. (1981). All of these different combinations provided consistent results, with antigorite + quartz stable with respect to talc at temperatures up to 80 to 120\*C. However, calculations using 427 the Deep Earth Water (DEW) model (Huang & Sverjensky 2019, Sverjensky et al 2014) and the 428 extended Helgeson-Kirkham-Flowers (HKF) aqueous equation of state (Shock et al 1992, Shock et al 429 430 1997) do not predict equilibrium coexistence of antigorite + quartz above 15°C.

- 431
- 432

3.4 (U,Th)/He ratio measurements and cooling age calculation

433

All (U-Th)/He analyses were completed at the UTChron facility at the University of Texas at Austin, 434 435 using aliquots of zircon separates from the metamorphic sole and lower crustal gabbros, previously analyzed for U/Pb ages by Rioux et al. (in prep.), following procedures of Wolf and Stockli (2010). 436 Individual zircon grains were morphometrically characterized to determine alpha ejection correction 437 438 (Ft, Farley et al., 1996; Cooperdock et al., 2020), equivalent spherical radius (ESR), and estimated mass assuming a tetragonal prism. Single-grain zircon sample aliquots were loaded into Pt tubes for 439 440 in-vacuo laser He heating for 10 min at ~1200°C by diode laser and 4He concentrations were measured by isotope dilution, using a <sup>3</sup>He tracer, on a Blazers Prisma QMS-200 guadrupole mass 441 spectrometer, after cryogenic purification. Blanks and <sup>4</sup>He gas standards were run between unknowns 442

- to monitor and quantify the procedural baseline during analytical runs. Aliquot laser reheating was
   repeated (2-5x) until 4He gas yields dropped <1% total extracted gas.</li>
- 445

After degassing, individual zircon grains were removed from the Pt packets and dissolved using a 446 two-step HF-HNO3 and HCI pressure vessel dissolution technique and measured on a Thermo 447 Element2 HR-ICP-MS following the procedure outlined in Wolf and Stockli (2010). U-Th-Sm 448 concentrations were calculated using isotope dilution with an isotopically enriched, mixed U-Th-Sm 449 spike calibrated against a 1 ppb U-Th-Sm gravimetric standard solution and blank-corrected using the 450 451 average of multiple procedural blanks. 452 Final (U-Th)/He ages were calculated using blank corrected U, Th, Sm and He measurements for 453 each aliquot. Reported concentrations were determined using the morphometrically determined mass 454 of each aliquot. The reported error for individual (U-Th)/He ages represents standard error (8%) 455 based on long-term intra-laboratory reproducibility of Fish Canyon tuff zircon standard, following the 456 approach of Farley et al. (2001). The reported mean sample ages reflect the arithmetic mean of the 457 458 aliguot ages and their standard deviations. 459

3.5 Calibration of XRF core scanner data

461

460

XRF core scanner data were collected onboard DV Chikyu, as described in Kelemen et al. (2020b). 462 463 We used the core scanner to analyze nine listvenite samples from BT1B core, and 14 gabbro samples from Hole GT1A core that had known bulk compositions based on XRF analysis at the 464 465 University of St. Andrews. While onboard DV Chikyu, we used the St. Andrews data to calibrate the 466 XRF data, as follows: wt% SiO<sub>2</sub> =  $0.89 \times (\text{scanner wt\% SiO}_2)$ ; wt% MgO =  $2.57 + 1.18 \times (\text{scanner wt\%})$ MgO); wt% FeO<sup>T</sup> = 1.048 + (scanner wt% FeO<sup>T</sup>)^0.848; and wt% CaO = 0.878 x (scanner wt% CaO), 467 where FeO<sup>T</sup> indicates all Fe is treated as FeO. Fits are illustrated in Supplementary Figure 2. Okazaki 468 et al. (2021) present a comprehensive analysis of the XRF scanner data, together with X-Ray 469 470 tomography data for BT1B core.

471 472

3.6 Calculation of mineral volume proportions

473

Volume proportions of quartz, magnesite and dolomite were estimated from bulk rock compositions 474 475 and XRF scanner data as follows. Weight fractions of SiO<sub>2</sub>, MgO, FeO<sup>T</sup> and CaO were converted to moles in 100 grams of rock using their molecular weights. (For all data reported in this paper, the sum 476 of wt% SiO<sub>2</sub>, MgO, FeO<sup>T</sup> and CaO was greater than 90% of the volatile free, bulk rock composition). 477 478 The number of moles of dolomite were taken to be equal to moles of CaO, moles of magnesite were 479 calculated as moles MgO – moles CaO, and moles of quartz were taken to be equal to moles of SiO<sub>2</sub>. 480 All Fe was inferred to be in Fe-oxides and hydroxides. If the small amounts of Fe in carbonate 481 minerals were included in such a calculation, this would slightly increase the proportions of magnesite

482 and dolomite, relative to quartz. Volumes of each mineral in 100 grams of rock were calculated using

483	their molar volumes. The data were "projected" from Fe oxy-hydroxides by normalizing the volumes of
484	quartz, magnesite and dolomite to 100%.
485	
486	4. Results
487	
488	4.1 Drilling operations
489	
490	Site BT1 is on the north side of the broad channel of Wadi Mansah, which drains mountainous
491	regions to the east and southeast. Hole BT1A penetrated 1.90 meters of gravels in Wadi Mansah,
492	south of listvenite outcrops flanking the Wadi. After we became concerned that a steep hole there
493	might intersect tens of meters of gravel before reaching bedrock, and/or that the gravel might overlie a
494	major, steep fault along Wadi Mansah that postdates ophiolite emplacement, we moved the drill and
495	inclined the Hole. Hole BT1B is three or four meters closer to the listvenite outcrop. The well head is
496	intact, marked and protected from floods by a concrete monument. Drilling of the fine-grained, silica-
497	rich listvenites was challenging, because this lithology has a hardness similar to fine-grained chert or
498	flint (~ 7 on the Mohs' scale), but with patience and expert drilling, we obtained ~ 100% recovery of all
499	lithologies intersected by the borehole. Details of drilling operations are in Kelemen et al. (2020b).
500	
501	4.2 Mapping and structural observations
502	
503	Geological mapping of the area around MoD Mountain, together with measurement of the orientation
504	of lithological contacts, began in 2009, and continued via brief visits through 2019. As noted in
505	Section 2.5, and reported by Falk and Kelemen (2015), the mountain is underlain by a southeast to
506	northwest striking anticline, with its faulted hinge approximately coincident with the ridge along the
507	summit. Exposed contacts between listvenite and serpentinized harzburgite consistently dip to the
508	southwest on the southwest side of the mountain, and to the east and northeast on the northeast side.
509	In the valley north of the mountain, listvenite-harzburgite, listvenite-metamorphic sole, and
510	harzburgite-sole contacts form a broad, shallow syncline. A new, detailed geological map and cross-
511	section, incorporating our lithological and structural observations, is presented in Figure 4.
512	
513	As shown on the map, in outcrops extending for more than 4 km north of Hole BT1B there is a regular
514	tectonic "stratigraphy", with variably altered peridotite overlying the metamorphic sole, in turn
515	overlying Hawasina Formation sediments, all with low angle fault contacts that have been deformed
516	by a series of gentle, broad open folds evident in the map and cross-section.
517	
518	We noted normal and reverse shear sense indicators on both sides of the anticline. As a result of the
519	folding, it is difficult to interpret the sense of shear recorded in various outcrops. Some shear sense
520	indicators may predate the folding, while others may record deformation during or after folding.
521	

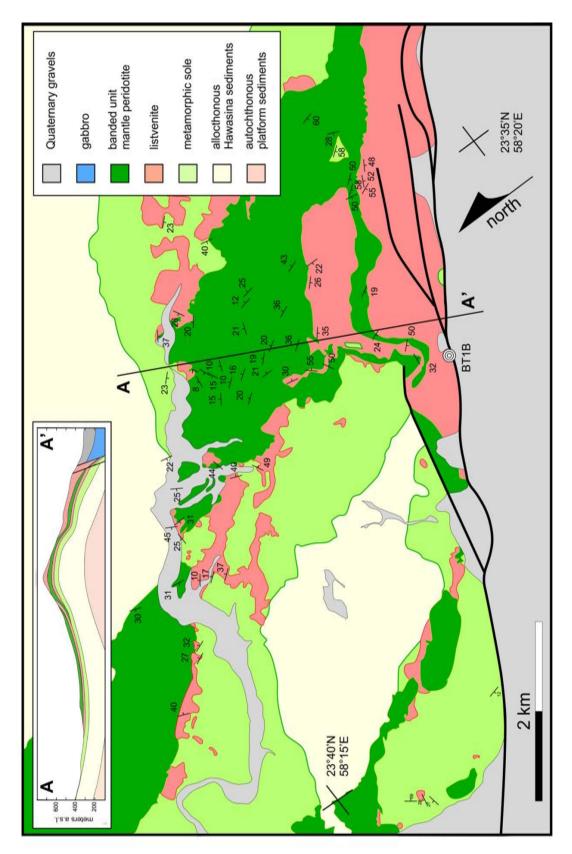
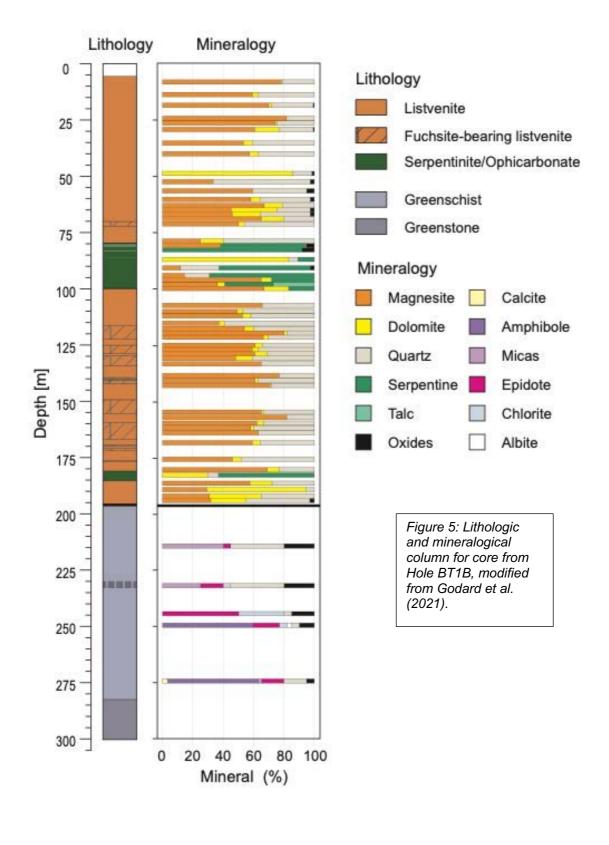


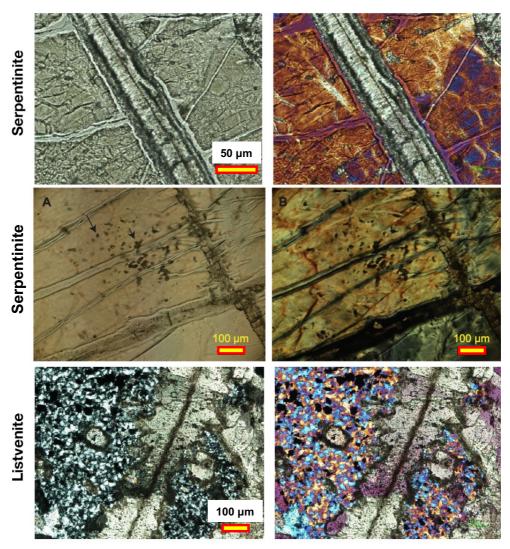
Figure 4: Geologic map and cross section of MoD Mountain and vicinity. Complex map pattern arises from intersection of topography with broadly folded, gently dipping units, as is more evident in the cross section (no vertical exaggeration) illustrating the antiform coinciding with MoD Mountain, and the syncline to its north. Dips are all measured on lithologic contacts, including dunite/harzburgite contacts within the Samail ophiolite mantle section.

528	
529	In contrast, to the northwest and southeast, outside our map area, the outcrop patterns become much
530	more complex, the sole outcrop thins, and there are some vertical fault contacts where all of these
531	older units are juxtaposed with Late Cretaceous Al Khod conglomerates and younger, shallow marine
532	limestones (Stanger 1985, Villey et al 1986, Wilde et al 2002).
533	
534	4.3 Zircon (U,Th)/He cooling ages
535	
536	Zircon (U,Th)/He cooling ages on samples from the metamorphic sole southeast of Fanjah, near MoD
537	Mountain, are $38.7 \pm 7.7$ and $44.4 \pm 8.0$ Ma, cooling ages of zircons from the metamorphic sole at the
538	base of the Wadi Tayin massif to the east are $54.5 \pm 7.4$ and $61.8 \pm 2.6$ Ma, and the cooling age of
539	zircons from the lower crust of the Samail massif is 46.4 $\pm$ 3.9 Ma (Supplementary Table 1 and
540	Supplementary Figure 1).
541	
542	4.4 Lithology
543	
544	As illustrated in Figure 5, the top of the Hole sampled ~ 200 meters of listvenite interlayered with two
545	serpentinite bands from 80 to 100 m depth, and 181 to 185 m depth. Below 185 m, the listvenite is
546	ubiquitously deformed, with visual core descriptions indicating a mixture of brittle and ductile
547	deformation. At about 197 m, core was composed of a few tens of cm of soft, clay-rich fault gouge,
548	together with a few cm of hard, aphanitic, black ultracataclasite. Beneath these fault lithologies, the
549	core sampled ~ 102 meters of the metamorphic sole, grading from dominantly fine-grained, finely-
550	banded, muscovite-bearing metasediments at the top ("greenschists" in Figure 5) to coarser, more
551	massive-appearing, foliated "greenstones", interpreted as metavolcanic rocks, at the bottom.
552	
553	Serpentinite bands in the core have gradational contacts with host listvenites over 10's of centimeters
554	to ~ 1 m thick. Serpentinites contain antitaxial veins of magnesite with a median line composed of
555	hematite and other Fe-oxides. There are prismatic terminations of magnesite crystals away from vein
556	centers, toward the host serpentine (Figure 6). Antitaxial veins record growth of minerals outward
557	from the vein center. They are commonly interpreted to open due to the "pressure of crystallization"
558	(Durney & Ramsay 1973, Urai et al 1991). However, this interpretation is less clear in the
559	serpentinites and listvenites of Hole BT1B, where the Mg in the carbonate is derived from the host
560	rocks. To some degree these veins may replace, rather than displace, the host.
561	
562	Away from the veins, serpentinites also contain up to 10% magnesite ovoids 10 to 100 microns in
563	diameter, unevenly dispersed within a massive serpentine matrix. These magnesite vein and ovoid
564	textures are abundant in the listvenites as well. Thus, the Shipboard Scientific Party suggested that
565	they are indicative of incipient replacement of serpentinite by listvenite, grading from < 10% carbonate
566	(and no quartz) in veins and ovoids to 100% fine-grained carbonate + quartz across sharp reaction
567	fronts (Kelemen et al 2020b). Another notable feature is that some of the serpentinites are optically

568 isotropic in thin section, probably indicative of low temperature formation of poorly ordered or

amorphous material with serpentine stoichometry (e.g., Andreani et al 2004).





574 575 Figure 6: Antitaxial magnesite-hematite veins, and magnesite spheroids in serpentinites and listvenites in core from Hole BT1B. Top panels: plane light (left) and crossed-polarized images (right, 576 guartz plate (+1 $\lambda$ ) inserted) of magnesite-hematite veins near the lower contact of the upper 577 serpentinite band, TS\_BT1B\_44Z-3\_9-11.5, ~100 m depth, from Figure F47 in Kelemen et al. 578 (2020b). Middle panels: tiny magnesite spheroids in serpentinite, TS BT1B 44Z-3 9-11.5, ~100 m 579 depth, from Figure F29 in Kelemen et al. (2020b). F. Bottom panels: Cross-polarized images, right 580 581 one with quartz plate  $(+1\lambda)$  inserted, of texturally similar, "antitaxial" magnesite-hematite veins and 582 magnesite ovoids in quartz-rich, listvenite matrix, TS BT1B 47Z-3 15-19 at about 110 m depth, from Figure F35 in Kelemen et al. (2020b). 583 584

- 585 In turn, listvenites and serpentinites recovered in drill core are hosted by more typical, partially
- 586 serpentinized peridotites and dunites in outcrop north and northeast of Hole BT1B (Figure 7). Such
- 587 lithologies, typical of the Banded Unit at the base of the mantle section of the Samail ophiolite, are
- abundant on the flanks of MoD Mountain, and are particularly well exposed west of the summit
- 589 (Figure 2) and on the broad, north facing outcrop below the summit ridge.
- 590

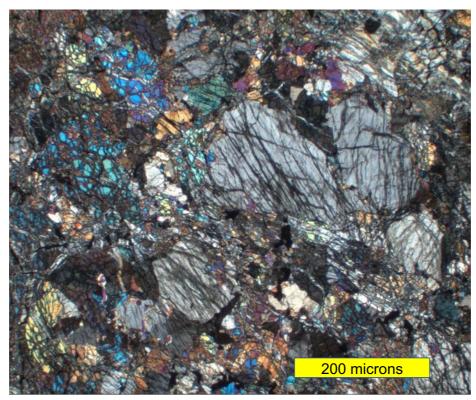


Figure 7: Cross-polarized image of partially-serpentinized harzburgite sample OM09-14 (Falk & Kelemen 2015) from ~ 10 m above lower listvenite band in Figure 2. Olivine: bright interference colors and irregular, serpentine-filled fractures. Orthopyroxene: grey interference colors and parallel to orthogonal fractures. Minor calcic-pyroxene and/or hornblende are barely visible in this image.

A sample transect on the ridge forming the drainage divide between Wadi Mansah (site of Hole BT1B) and the parallel wadi north of MoD Mountain documented a 5-meter scale progression from listvenite to serpentinite (with intergrown quartz and antigorite) to partially serpentinized peridotite containing relict olivine and orthopyroxene (Figure 5 in Falk & Kelemen 2015). Along that watershed transect, the presence of antigorite – rather than the serpentine polytypes lizardite and chrysotile – was attributed to high SiO<sub>2</sub>-activity produced by reaction of olivine and serpentine to produce carbonate and quartz, since antigorite is more SiO<sub>2</sub>-rich than the other polytypes.

604

605 However, the serpentinites in core from Hole BT1B are distinct from the serpentinized zone flanking

listvenite on the ridge transect, and from the surrounding, partially serpentinized Banded Horizon

- 607 harzburgites. Although quartz veins cut the serpentinite in the core, antigorite was not observed.
- Moreover, despite the presence of some orthopyroxene pseudomorphs ("bastites") in serpentinites,
- and a concerted effort to find relict mantle minerals, no olivine or pyroxene were detected in drill core.
- Taken together, field and core observations suggest that the contact between serpentinites and
- 611 partially serpentinized peridotites is gradational over a few meters at most, approximately as sharp as
- 612 the contact between listvenites and serpentinites. More on this in Sections 5.7 and 5.8, below.
- 613
- Essentially, two types of listvenite were recovered, magnesite + quartz + iron oxide lithologies, and volumetrically less abundant, dolomite + quartz + iron oxide rocks, previously termed magnesite-

- lisvenites and dolomite-listvenites (Falk & Kelemen 2015). Much of the core contains 0.5 to 3% relict
   chromian spinel, partially or fully altered to Fe-oxides. Instead or in addition, some samples contain
- 618 minor amounts of Cr-rich white mica (fuchsite-muscovite solid solutions, supplementary Figure 7 in
- 619 Falk & Kelemen 2015), in mm to cm scale, ovoid, microscopic intergrowths with quartz. These
- 620 intergrowths are macroscopically evident in outcrop and core as cm-scale green spots, though in fact
- 621 Cr-rich mica composes only a few percent of such spots, apparently has undergone alteration to clays
- 622 in some samples, and was significantly damaged or removed during thin section preparation. As a
- result, fuchsite crystals recovered from BT1B core are not large enough for <sup>40</sup>Ar/<sup>39</sup>Ar analyses. Figure
- 5 of Nasir et al. (2007) is a photomicrograph of crystalline fuchsite from another listvenite in Oman.
- 625
- Macroscopic listvenite textures are characterized by abundant veins (10 to 200 veins more than 1 mm thick per meter of core, typically ~ 1 per cm) in a fine-grained matrix. In massive listvenites, the finegrained matrix surrounding veins commonly contains ovoids of magnesite or quartz (Figure 8).
- Though they have similar textures, ovoids of the two different minerals are rarely adjacent to each
- other. Both commonly have Fe-oxides in their cores and/or in spherical zones. Microprobe analyses
- show that magnesite ovoids have low Fe cores, commonly rimmed with relatively Fe-rich magnesite
- 632 (Beinlich et al 2020). They have sizes and shapes similar to the quartz spherulites.
- 633

Carbonate ovoids and cross-cutting magnesite-hematite veins are also observed in serpentinite
bands in the core (Figure 6). Thus, the Shipboard Science Party considered it likely that many such
veins initially formed within serpentinite, followed by formation of ovoids within surrounding
serpentine, and then by replacement of the entire serpentinite matrix by carbonate + quartz (Kelemen
et al 2020b). If so, in contrast to conventional interpretations of veins as relatively young features,
"cross-cutting" their matrices, in this case the fine-grained listvenite matrix may postdate the earliest
veins found within the matrix. This hypothesis is discussed further in Section 5.9

641

The quartz ovoids have the texture of "spherulites", with radiating microscopic crystals producing a 642 643 false, "uniaxial interference pattern" in cross-polarized light. Spherulites form during devitrification of amorphous opal as well as rhyolite glass, so Falk & Kelemen (2015) and the Shipboard Science Party 644 645 (Kelemen et al 2020b) interpreted these as replacing opal, which would have been among the earliest SiO<sub>2</sub> minerals to form in many of the listvenites. Importantly, opal is commonly found in other 646 listvenites and serpentine-magnesite associations worldwide (Abu-Jaber & Kimberley 1992, Aftabi & 647 648 Zarrinkoub 2013, Akbulut et al 2006, Arisi Rota et al 1971, Barnes et al 1973, Beinlich et al 2010, Borojević Šoštarić et al 2014, Boschi et al 2009, Ece et al 2005, Jurković et al 2012, Lacinska & 649 Styles 2013, Lapham 1961, Oskierski et al 2013a, Oskierski et al 2013b, Posukhova et al 2013, 650 651 Quesnel et al 2016, Searston 1998, Ulrich et al 2014, Zarrinkoub et al 2005).

- 652
- Vein types cutting this fine-grained matrix generally record a progression from texturally early,
- 654 antitaxial magnesite veins some with cores of hematite + other Fe-oxides (Figure 6) and related,
- 655 early Fe-oxide veins, to syntaxial dolomite veins and carbonate-quartz veins, and lastly to syntaxial

- 656 calcite veins. Some of the late, syntaxial veins contain vugs lined with prismatic calcite and/or
- 657 dolomite. Syntaxial vein textures record inward crystallization of crystals into fractures that opened
- due to external, tensional stresses (Durney & Ramsay 1973, Urai et al 1991).
- 659
- 660 (A poorly exposed, weathered, fuchsite vein has been observed in outcrop 200 meters north of Site
- 661 BT1B, but no such veins were sampled in BT1B core. We mention this simply to emphasize that Cr
- and AI were mobile at some stage during listvenite formation or later alteration.)
- 663

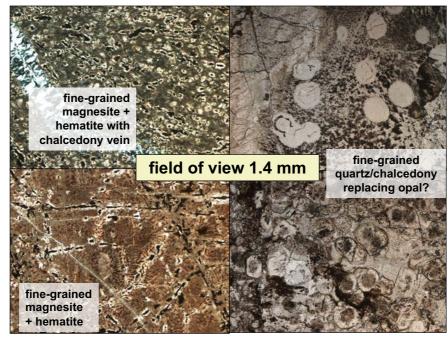
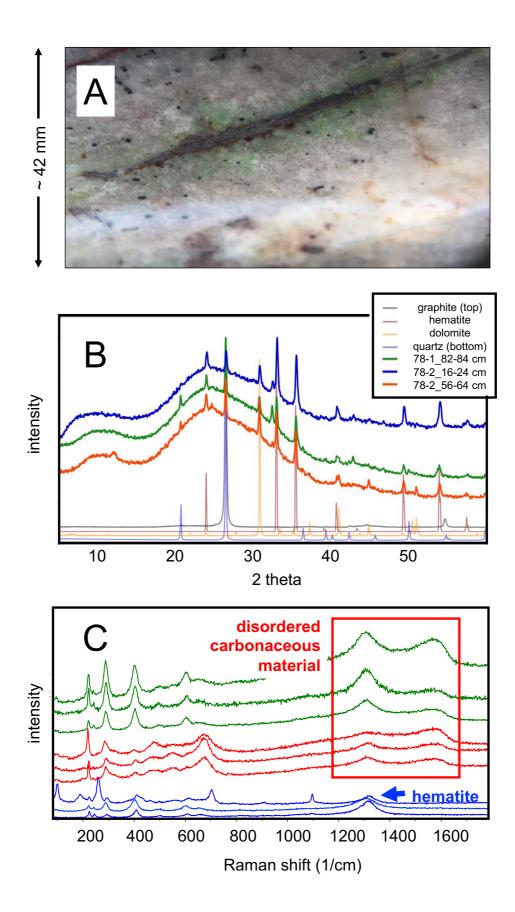
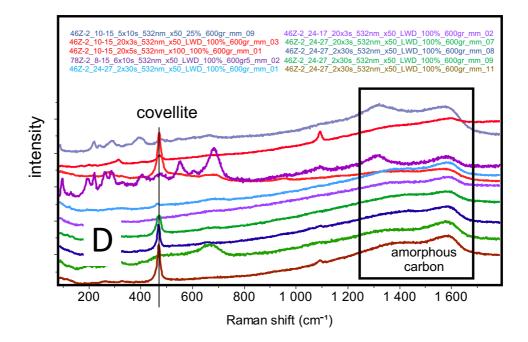
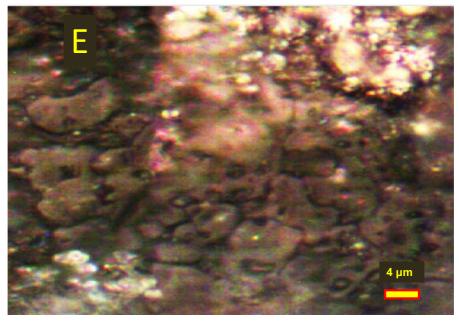


Figure 8: Plane light photomicrographs illustrating magnesite ovoids in a matrix composed of
magnesite, quartz, and subordinate hematite and Fe-oxyhydroxides, TS\_BT1B\_20Z-1\_42-46, ~ 40 m
depth (left top) and TS\_BT1B\_27Z-2\_6-8.5, ~ 59 m depth (left bottom), and quartz spherulites with
carbonate and hematite inclusions, in matrix of fine-grained quartz and hematite with subordinate,
microscopic carbonates, TS\_BT1B\_60Z-1\_12-17, ~140 m depth (right).

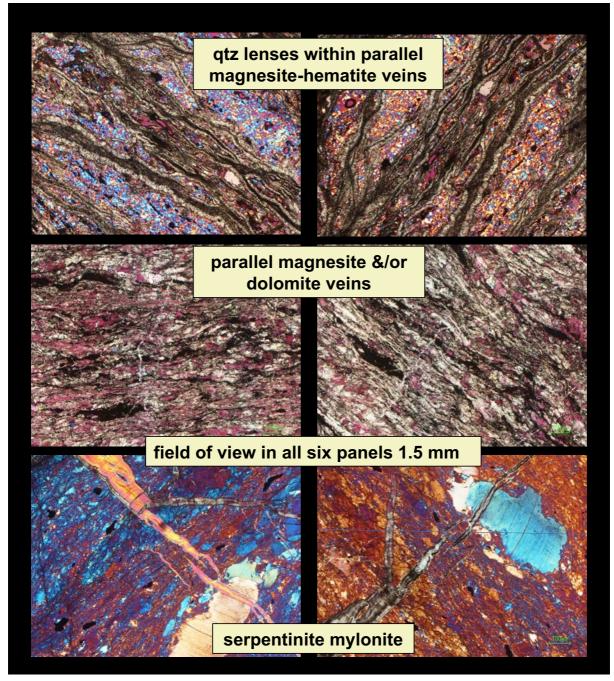
- 670
- 671 Among the Fe-oxide veins, some contain minor sulfide generally not detected during core
- 672 description and amorphous, organic carbon compounds. The carbon compounds were first
- 673 identified in core at the drill site, in the lowest listvenite band, as elongate lenses within transposed
- hematite veins parallel to the penetrative foliation, where they were described as "graphite". Soft
- organic carbon compounds in these features appear to have been largely lost from sample surfaces
- 676 during washing and handling of the core prior to shipboard observations and analyses, and again
- during fabrication of thin sections. However, Raman spectroscopy of small, armored relicts, in oxide
- veins and also in isolated, dark red spots that resemble relict spinel on the core face and rock slabs,
- reveals the presence of disordered, thermally immature carbonaceous material (Figure 9), some of
- 680 which may retain a more ordered organic molecular structure. The carbonaceous materials we can
- still find, on freshly cut surfaces from the core interior, are commonly on the margins of microscopic
- 682 chalcocite and covellite crystals, in one case also associated with copper sulfate (chalcanthite)







685 Figure 9: Carbonaceous material in drill core. A: Drill site photo of 1 to 2 mm vein of shiny, grey, soft material described as graphite on the drill site, in a vein rimmed with hematite. Field of view 5 cm 686 wide, core 74Z-01, ~ 195 m depth. B: Shipboard XRD spectra of soft "graphite" powder extracted from 687 veins with "graphite" + hematite, replacing Figure F43 in Kelemen et al. (2020b). Interpretation of 688 689 these data is complicated by the similarity of the quartz and graphite peaks at ~ 26° 2 $\Theta$ , but quartz also has a prominent peak at ~ 21°C which is absent from the blue spectrum for 78Z-02 16-24. C: 690 Raman spectra of black material in samples BT1B 77Z-03 30-38 (blue), 78Z-02 8-15 (red) and 78Z-691 02 50-56 (green, ~ 192-198 m depth. Broad double peaks at wavenumbers of ~1350/cm and 692 693 1600/cm are indicative of disordered carbon compounds; no Raman spectra diagnostic of graphite 694 were obtained. Many microscopic, soft, black domains contained hematite, with a single broad peak 695 at ~1350/cm. instead of, or in addition to, disordered carbon compounds, D: Raman spectra of black material in samples BT1\_44Z-02\_10-15, BT1\_78Z-02\_8-15, and BT1\_44Z-02\_24-27, showing broad, 696 double peaks indicative of disordered carbon compounds, some associated with covellite. Core 44Z ~ 697 98 m depth. E: dark grey copper sulfate "cow pies", spatially associated with brightly reflecting 698 covellite, near amorphous carbon material, field of view ~ 100 microns, sample BT1\_44Z-02\_24-27. 699 Data and images from core 44Z are from a 1 mm diameter black spot with a red rim in the interior of 700 701 the core, exposed by the rock saw during sample preparation.



703 Figure 10: Examples of crystallographic preferred orientation (CPO) in localized zones in core from Hole BT1B. All images in cross-polarized light with quartz plate inserted. Images on right are of the 704 same area as on left, but rotated 90° with respect to the polarizers. Areas showing optical continuity 705 have a crystallographic preferred orientation. Top, TS BT1B 31Z-4 12-14, ~ 65 m depth, quartz 706 lenses within sub-parallel, anastomosing magnesite-hematite veins. Middle, TS\_BT1B\_78Z-2\_34-38, 707 ~ 195 m, thin section composed almost entirely of parallel magnesite and/or dolomite veins, with a 708 CPO in the carbonates. Bottom, TS BT1B 74Z-1 59-62, ~ 183 m, serpentinite mylonite, with a 709 strong CPO in the fine-grained matrix, and visible, internal deformation in serpentine porphyroclasts. 710

711 712

4.5 Crystallographic preferred orientation of minerals

- 713
- An overall preferred orientation of veins is not evident from structural data on the core scale, possibly
- 715 due to differential rotation of core pieces. And, systematic measurements have not been made on the

716	outcrop scale. However, at the meter scale many core intervals contain textures indicative of ductile
717	deformation (Figure 10). Some samples show a strong macroscopic foliation, defined by parallel
718	(possibly transposed) veins enclosing elongate lenses of the fine-grained matrix, in textures
719	commonly interpreted to form via boudinage during ductile deformation. In samples with a strong
720	foliation defined by dozens of subparallel, early magnesite veins per 10 mm <sup>2</sup> , intervening patches of
721	fine-grained quartz commonly have a crystallographic preferred orientation (CPO). Some samples
722	with a strong foliation defined by abundant, subparallel (tectonically transposed?) carbonate veins
723	also have an optically evident, crystallographic preferred orientation in magnesite and/or dolomite.
724	Similarly, some shear zones in serpentinite have an optically evident, strong shape- and
725	crystallographic-preferred orientation of lizardite crystals, and contain deformed serpentine
726	porphyroclasts.
727	
728	4.6 Brittle deformation textures
729	
730	A broad range of different breccias and cataclasites are observed in listvenites and in the
731	metamorphic sole, in outcrop and in core. In turn these are cut by sharp faults - some associated with
732	planar bands and branching veins of aphanitic ultracataclasite and/or pseudotachylite - and by late
733	calcite veins. The nature and interpretation of cataclasites and faults observed in core from Hole
734	BT1B are discussed in detail by Menzel et al. (2020).
735	
736	4.7 Geochemical data
737	
738	The bulk composition of core samples was measured in five different ways: (1) Major and minor
739	element compositions of nine samples were measured by XRF at St. Andrews University. (2) Major
740	and minor element compositions of 74 samples, including those previously analyzed at St. Andrews,
741	were measured via XRF (both fusion and pressed pellets) onboard the Drilling Vessel Chikyu. (3)
742	Major element compositions of the cut face of selected core sections were analyzed onboard using an
743	XRF core scanner. (4) Trace element compositions of a few samples were analyzed onboard via ICP-
744	MS. (5) Trace element compositions of 61 samples were analyzed via ICP-MS at the Université de
745	Montpellier. These data, and subsequent analyses, are tabulated and described in Kelemen et al.
746	(2020b) and Godard et al. (2021), so for brevity we simply refer readers to those other publications.
747	
748	5. Discussion
749	
750	5.1 Subduction zone setting of listvenite formation
751	
752	On the basis of their structural observations west and northwest of the MoD Mountain area, and U/Pb
753	formation or cooling ages from cross-cutting calcite veins, (60 $\pm$ 16 and 58 $\pm$ 6 Ma) Scharf et al.
754	(2020) infer that listvenites in the Fanjah area postdate ophiolite emplacement, echoing the qualitative
755	ideas of Stanger (1985) and Wilde et al. (2002). As noted in <mark>Sections 2.5 and 4.2</mark> , our

geochronological and field observations are inconsistent with this interpretation. The listvenite mineral

- isochron age of 97 ± 29 Ma ( $2\sigma$ ) (Falk & Kelemen 2015) yields a 93% chance (1.5 $\sigma$ ) that the
- listvenites formed before 75 Ma, while subduction beneath the ophiolite was still active, and a 97%
- chance that the listvenites formed before 68 Ma. Thus, the isochron data support listvenite formation
- during subduction, and show that the calcite veins cutting listvenite, dated by Scharf et al., postdate
- 761 listvenite formation.
- 762

763 In this paper, our mapping and structural observations indicate an intact tectonic "stratigraphy" around 764 the basal thrust of the ophiolite, encompassing the allocthanous Hawasina metasediments, overlain 765 by the metamorphic sole, in turn overlain by Banded Unit dunites and harzburgites of the Samail ophiolite mantle section, with essentially horizontal lithologic contacts extending for kilometers. This is 766 the stratigraphy created during subduction beneath the ophiolite, and - together with the isochron - it 767 768 indicates that these units were juxtaposed during low angle thrusting along the subduction zone. In 769 turn, all the listvenites in the MoD Mountain area are less than a kilometer away from the basal thrust, and have contacts with surrounding mantle peridotites and the metamorphic sole that are parallel to 770 the basal thrust. All these data, taken together, indicate that the listvenites formed along and above 771 772 the basal thrust of the ophiolite during subduction.

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- 774 775

5.2 Faulting, deformation and veining after listvenite formation

(U,Th)/He cooling ages (Section 4.3) indicate that the region around MoD Mountain remained above 776 the closure temperature for He diffusion in zircon, ~ 180°C (Reiners et al 2004), or were reheated 777 778 above this temperature, during 30 to 60 million years after formation of the igneous crust in the 779 ophiolite and peak metamorphism of the sole. Based on textural observations, it is clear that the breccias and cataclasites sampled in BT1B drill core postdate listvenite formation (Menzel et al 2020). 780 Some may postdate ophiolite emplacement, and may be broadly related to Paleocene to Miocene 781 uplift and extension during formation of the nearby Jebel Akdar and Saih Hatat domes. On the other 782 783 hand, some could be related to deformation during subduction beneath the ophiolite. The long term persistence of temperatures above 180°C in this region renders it difficult to distinguish formation from 784 785 cooling ages using (U,Th)/He data.

- 786
- 787 788

#### 5.3 Temperature of listvenite formation

The temperatures of listvenite formation have been previously constrained using metamorphic phase equilibria, conventional oxygen isotope thermometry, and clumped isotope analyses. Falk & Kelemen (2015) noted the presence of intergrown antigorite (serpentine) + quartz. in the reaction zone between listvenite and serpentinite on the west ridge of MoD Mountain (Figure 2). In some cases, these samples also contain talc. They used Thermocalc (Powell et al 1998), with thermodynamic data from Holland and Powell (2003, 1998), and similar methods using the thermodynamic data of Gottschalk (1997), to estimate that equilibrium coexistence of antigorite, quartz and talc occurs at 80 to 120°C,

- 796 depending on the choice and uncertainty of thermodynamic parameters for the minerals, and the (poorly known) pressure at which these assemblages crystallized. For this paper, we obtained similar 797 using Perple X (https://www.perplex.ethz.ch/) (Connolly 1990, Connolly 2005, Connolly 2009) and 798 mineral properties from Holland and Powell. 799
- 800

801 In contrast, calculations using the Deep Earth Water (DEW) model (Huang & Sverjensky 2019,

Sverjensky et al 2014) and the extended Helgeson-Kirkham-Flowers (HKF) aqueous equation of state 802

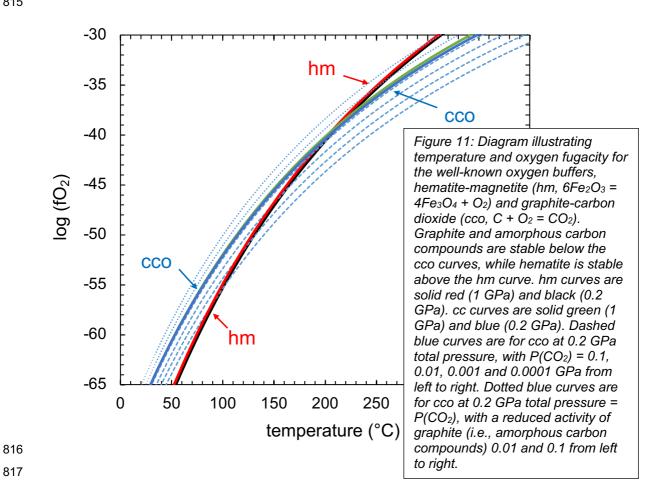
(Shock et al 1992, Shock et al 1997) do not predict equilibrium coexistence of antigorite + quartz 803

804 above 15°C. Since antigorite-guartz intergrowths indicate coeval crystallization of both phases, in

805 rocks that formed at > 80°C, we presume that these latter results stem from incorrect extrapolation of 806 thermodynamic data to low temperature.

807

Leaving the DEW results aside, the other temperature estimates based on co-existing antigorite + 808 guartz are broadly consistent with temperature estimates based on  $\delta^{18}$ O in guartz and carbonate 809 minerals in the listvenite, assuming that fluid  $\delta^{18}$ O was similar to seawater, and with clumped isotope 810 analyses of magnesite and dolomite yielding temperatures of 80 to 130°C for listvenite samples (Falk 811 812 & Kelemen 2015). Observation of quartz spherulites in listvenite, possibly reflecting devitrification of 813 opal, also suggests that listvenite formation occurred in this "low temperature" regime, at ~ 150°C or 814 less, in the presence of aqueous fluid.



819 Here we show that veins of intergrown amorphous carbon compounds and hematite sampled in the core also must have formed below ~ 200°C (Figure 11). The observation of "reduced" hydrocarbon 820 species intergrown with oxidized iron minerals in veins may seem surprising at first, but they are 821 mutually stable at low temperature due to the stronger temperature dependence of the iron oxidation 822 state, compared to the carbon oxidation state. Above ~ 200°C, reduced, organic carbon compounds 823 and oxidized iron minerals such as hematite cannot coexist at the same oxygen fugacity, but they can 824 crystallize together at moderate oxygen fugacities below ~ 200°C. We've found that this result is 825 826 robust across all available sets of internally consistent thermodynamic data, and in the pressure range from 0.2 to 1.0 GPa. 827

828

Fourteen more recent clumped isotope analyses for listvenites and carbonate-bearing serpentinites 829 from BT1B core yield an average temperature of 147±58°C (1o). Ten of the 14 temperature estimates 830 lie within the 1σ range, whereas two are lower (45±5 and 52±8°C) and the two least precise estimates 831 are higher (227±52 and 247±52°C) (Beinlich et al 2020). Though a few MoD Mtn listvenites might 832 have formed at temperatures greater than 200°C, such temperatures are too high for crystallization of 833 834 intergrown amorphous carbon + hematite, they are too high for crystallization of intergrown antigorite 835 + quartz, and they are too high for crystallization of opal. Also, carbonation of peridotite above ~150°C would form abundant talc + magnesite, whereas talc is absent in most MoD Mountain listvenites, and 836 rare in serpentinite-listvenite contact zones that are gradational over a few meters (Falk & Kelemen 837 838 2015, Kelemen et al 2020b).

839

840 The range in temperature estimates based on phase equilibrium and clumped isotope ratios from the 841 MoD Mountain listvenites may indicate that mineral assemblages in the listvenites and surrounding 842 serpentinites formed gradually over a range of times and temperatures. In addition, some of the clumped isotope data may record closure temperatures during cooling, rather than the peak 843 temperature at which the MoD Mountain listvenites first crystallized, as proposed for clumped isotope 844 data from fine-grained 10-meter scale magnesite veins in California (Garcia del Real et al 2016). 845 Alternatively, since the highest clumped isotope temperatures from Beinlich et al. are also the most 846 847 imprecise estimates, perhaps they result from analytical uncertainties or disequilibrium effects. When the 14 clumped isotope temperatures from Beinlich et al. are combined with the 31 older clumped 848 isotope temperatures from Falk and Kelemen, the full data set yields an average of  $100 \pm 46^{\circ}$ C (1 $\sigma$ ), 849 850 with the highest temperatures from Beinlich et al. falling outside the  $2.5\sigma$  (99% probability) range.

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853

### 5.4 Depth of listvenite formation

The depth of listvenite formation is difficult to constrain. As noted in Section 2.4, published work on the metamorphic sole beneath the Samail ophiolite reports peak temperatures up to 700 to 900°C at pressures ranging from 200 to 1400 MPa, indicative of anomalously hot subduction zone conditions. Common peak pressure estimates, ~ 1200 MPa, indicate depths exceeding the structural thickness of 858 the ophiolite. Thus, either (1) the published range of pressure estimates for the metamorphic sole is the result of uncertainty rather than true variation in depth, and/or (2) the lower few km of the ophiolite 859 860 mantle section has undergone thinning in some places, and/or (3) the lenses of the metamorphic sole recording the highest pressures migrated upward with respect to the overlying peridotite, and/or (4) 861 some mineral equilibria record tectonic overpressures rather than lithostatic loads. Also, we infer from 862 863 Sr isotope data that the fluids that formed the listvenites were not derived from the metamorphic sole (Section 5.6, below, and de Obeso et al 2021a). For all these reasons, it is unclear whether the 864 metamorphic pressures inferred for the sole constrain the depth of listvenite formation. However, 865 866 temperatures and pressures recorded by the sole do provide a window into the thermal evolution of subduction beneath the ophiolite before and during listvenite formation. 867

868

Initiation of subduction, during formation of the metamorphic sole, involved thrusting of a hot mantle 869 wedge over newly formed, hot basaltic crust. Over time, subduction of progressively older oceanic 870 871 crust and - eventually - the pelagic sediments of the Hawasina Formation would have caused cooling of the subduction zone as it evolved toward a steady state geotherm. A few clumped isotope analyses 872 873 on calcite in sediments beneath the ophiolite and the metamorphic sole on MoD Mountain yield 874 temperatures of 150 to 200°C (Falk & Kelemen 2015). It is possible that these were peak 875 temperatures during diagenesis of the sediments along a late, steady state subduction geotherm at 876 the pressure and depth recorded by the metamorphic sole in core from Hole BT1B, 800 -1200 MPa, or about 25 to 40 km. 877

878

In turn, even lower temperatures for most listvenite samples may record continued, isobaric cooling of 879 880 rocks flanking the subduction zone. Temperatures of 100 to 200°C at depths of 25 to 40 km are 881 inferred for fore-arc regions above subduction zones from heat flow data (reviewed in Peacock 1996) 882 predicted for steady state, oceanic subduction geotherms in numerical models (e.g., Peacock 1996, Peacock et al 2005, Syracuse et al 2010), including those recently modeled by Van Keken et al. 883 (2019), and lie within the cold end of the range of PT conditions recorded by subduction-related 884 885 metamorphic rocks (Hacker 1996, Hacker 2006, Penniston-Dorland et al 2015). Such low temperatures at 25 to 40 km are rare or absent in other tectonic environments. 886 887

To summarize, it is possible that the sediments and the overlying mantle peridotites at the base of the 888 889 ophiolite were juxtaposed by subduction at the leading edge of the mantle wedge, at a depth of 25 to 890 40 km, and that the MoD Mountain listvenites formed at these depths along a cold, steady state subduction geotherm. Alternatively, if the metamorphic sole has migrated updip with respect to the 891 overlying peridotites, then the listvenites could have formed at lower pressures and shallower depths. 892 893 894

895

### 5.5 Composition of listvenite protolith and geochemical fluid additions

Given the abundance and variety of mineralogically simple veins, many of which are monomineralic, 896 there is substantial compositional variation in listvenites at the millimeter to meter scale. This 897

- variability extends to larger scales in some parts of the core. Nevertheless, remarkably enough,
- average MgO/SiO<sub>2</sub>/FeO\* (all Fe as FeO) ratios in the listvenites are very similar to those in average
- 900 residual peridotites from the ophiolite (Figure 12), as discussed further by Okazaki et al. (Okazaki et al
- 2021). These oxides comprise more than 90% of the volatile-free bulk composition of the rock. These
- 902 data suggest that there was little dissolution and export of major elements from the rocks during
- transformation of peridotite to serpentinite and then to listvenite as discussed further in Section 5.9.
- On the other hand, serpentinites and listvenites record addition of tens of weight percent H<sub>2</sub>O and
   CO<sub>2</sub> to the original bulk composition of mantle peridotite protoliths. In addition to CO<sub>2</sub>, dolomite
   listvenites clearly record substantial addition of CaO, and since CaO and Sr concentrations are
   strongly correlated of Sr as well.
- 909

Although the shipboard data do not reveal systematic variation in the abundance of magnesite vs
dolomite listvenites downhole, there is a clear change in the abundance of both Al and K, together
with many other highly incompatible trace elements. Concentrations of these elements are relatively
low above the serpentinite band at 80-100 m depth, and much higher below that band (Godard et al
2021, Kelemen et al 2020b).

915

Understanding the source for enrichment in these elements is complicated by uncertainty about their 916 concentration in the peridotite protolith. As noted in Sections 2.3, 2.5 and 4.4, a "Banded Unit" of 917 918 alternating dunite, harzburgite and lherzolite characterizes the base of the ophiolite mantle section in many regions. Some Banded Unit peridotites record high temperature geochemical "refertilization" by 919 920 reaction of residual mantle peridotites with infiltrating melt or fluid at > 800°C, with addition of calcic 921 pyroxene and Mg-rich hornblende. This produced enrichment in CaO and Al<sub>2</sub>O<sub>3</sub> to levels well above 922 those in typical residual mantle peridotites in the Samail ophiolite (Godard et al 2000, Hanghoj et al 2010, Monnier et al 2006). Indeed, four out of six harzburgite samples from the Banded Unit on MoD 923 Mountain have Ca and AI contents outside the 1o range of variability in residual mantle peridotite in 924 925 the ophiolite (Falk & Kelemen 2015, Godard et al 2021, Kelemen et al 2020c).

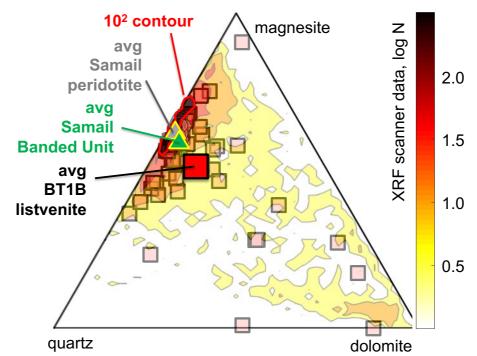
926

The listvenites below 100 meters depth in core from Hole BT1B have trace element ratios that are distinct from typical peridotites, but characteristic of the Banded Unit (Figure 12 and Godard et al 2021), as shown in Figure 13. In particular, middle to heavy rare earth element ratios in the Banded Unit, and in the listvenites, are high compared to typical Samail peridotites. Such enrichment in middle rare earth elements is commonly associated with the presence of igneous hornblende in peridotites. Thus, the listvenites probably inherited these characteristics from the enriched, Banded Unit protolith.

On the other hand, it is clear that most listvenites have higher Sr concentrations than typical Samail
ophiolite peridotites *and* the Banded Unit (de Obeso et al 2021a). Instead, Sr and Ca were added

936 during low temperature alteration, along with H<sub>2</sub>O and CO<sub>2</sub>. These topics are discussed further by

937 Godard et al. (2021).



938 Figure 12: Ternary diagram illustrating volume proportions of guartz, magnesite and dolomite. 939 projected from hematite, calculated from whole rock compositions, for Samail mantle peridotites 940 941 (average in grey triangle, data as large, grey open circles, barely visible beneath the averages and 942 the 10<sup>2</sup> contour area, Godard et al. 2000;Monnier et al. 2006;Hanghoj et al. 2010}, Banded Unit peridotites near the base of the Samail ophiolite mantle section (small, green open circles. barely 943 944 visible, Falk & Kelemen 2015, Khedr et al 2013, Khedr et al 2014, Takazawa et al 2003), and listvenites from Hole BT1B and MoD Mtn (open squares, Falk & Kelemen 2015, Kelemen et al 945 2020b), superimposed on contoured histogram of mineral proportions from shipboard XRF scanner 946 data. Contour interval 10<sup>1/2</sup>. Okazaki et al. (2021) provide more thorough interpretation of shipboard 947 XRF scanner data plus X-Ray Computed Tomography data on the whole core. Average listvenite, 948 Banded Unit and Samail mantle peridotite compositions are provided in Supplementary Table 2. 949

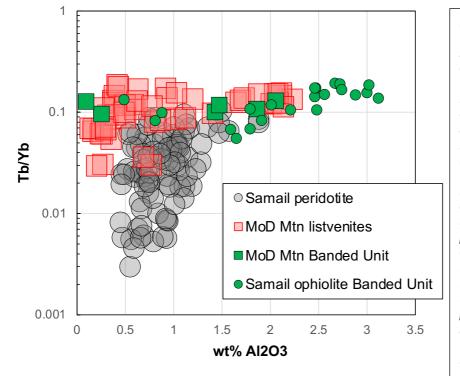


Figure 13: Wt% Al<sub>2</sub>O<sub>3</sub> versus Tb/Yb ratios in bulk rock compositions of MoD Mtn listvenites (open squares, Falk & Kelemen 2015, Godard et al 2021, Kelemen et al 2020b), and MoD Mountain peridotites that host listvenites (green squares, Falk & Kelemen 2015, Godard et al 2021), compared to typical Samail mantle peridotites (grey open circles, Godard et al 2000, Hanghoj et al 2010, Monnier et al 2006), and Banded Unit peridotites from the base of the ophiolite (Khedr et al 2013, Khedr et al 2014. Takazawa et al 2003).

Present day Sr isotope ratios in listvenites and serpentinites from MoD Mtn range from ~ 0.708 to 953 0.715 (de Obeso et al 2021a, Falk & Kelemen 2015). Using current Rb/Sr contents in these samples, 954 age corrected <sup>87</sup>Sr/<sup>86</sup>Sr ratios at 96 Ma range from ~ 0.708 to 0.714, much higher than the range of Sr 955 ratios in Samail ophiolite peridotites (0.703 to 0.707, Benoit et al 1999, Gerbert-Gaillard 2002, 956 Gregory & Taylor Jr 1981, Lanphere et al 1981, McCulloch et al 1980, McCulloch et al 1981), the 957 range in Late Cretaceous to modern seawater (~ 0.7075 to 0.7081), or the range in peridotite-hosted 958 ground water in the Samail ophiolite (~0.7065 to 0.7092, Weyhenmeyer 2000). Thus, the Sr-, Ca- and 959 CO<sub>2</sub>-rich fluid(s) that modified the mantle overlying the basal thrust of the ophiolite had relatively high 960 <sup>87</sup>Sr/<sup>86</sup>Sr ratios compared to fresh, residual mantle peridotites. 961

962

Initially, we expected that the metamorphic sole, as sampled by core from Hole BT1B, might be the
source of fluids that formed listvenites in overlying peridotites, or at least might be analogous to the
source of these fluids. However, this is not consistent with the Sr isotope data on the sole in core from
Hole BT1B. Measured and age-corrected Sr isotope ratios in the metamorphic sole are consistently
lower than corresponding ratios in the listvenites (de Obeso et al 2021a).

968

Instead, pelagic, clastic units of the underlying Hawasina sedimentary rocks have measured and age-969 970 corrected Sr isotope ratios that span the same range as those in the listvenites (de Obeso et al 971 2021a). Thus, these sedimentary units in the Hawasina may be analogous to subducted sedimentary rocks that produced CO<sub>2</sub>-bearing fluids which, in turn, formed the MoD Mountain listvenites. This is 972 973 likely, despite the presence of C- and Sr-rich limestone and dolomite units with lower Sr isotope ratios 974 in the Hawasina Formation, because devolatilization of clay and mica bearing, clastic metasediments produces abundant, CO<sub>2</sub>-rich aqueous fluids, while limestone and marble remain relatively refractory 975 at low to moderate temperature, subduction zone conditions (e.g., Kerrick & Connolly 2001, Stewart & 976 Ague 2020). 977

978

Indeed, there is evidence for a deeply subducted component with terrigenous isotope characteristics
– like those of the Hawasina clastic sedimentary rocks – elsewhere in the Samail ophiolite. A series of
felsic intrusions in the sole, mantle and lower crust along the length of the ophiolite have low, agecorrected <sup>143</sup>Nd/<sup>144</sup>Nd (and thus, presumably, high <sup>87</sup>Sr/<sup>86</sup>Sr(t)), attributed to melting of high-grade
metasediment in the subduction zone below the ophiolite (Amri et al 2007, Briqueu et al 1991, Cox et
al 1999, Haase et al 2016, Haase et al 2015, Lippard et al 1986, Rioux et al 2021a, Rioux et al 2013,
Rioux et al 2021b, Rollinson 2015, Spencer et al 2017).

986

987 A complication is that the clastic units in the Hawasina formation have  $\delta^{13}$ C less than -4 per mil,

988 whereas listvenites have  $\delta^{13}$ C > -3 per mil. These differences in carbon isotope ratios can be

989 understood as the result of temperature dependent carbon isotope fractionation. As discussed in

990 more detail in de Obeso et al. (2021a), at temperatures greater than ~ 300°C, dissolved CO<sub>2</sub> in

- aqueous fluids has  $\delta^{13}$ C higher than co-existing calcite and dolomite (Deines 2002, Horita 2014). At
- lower temperatures, calcite and dolomite have  $\delta^{13}$ C higher than co-existing fluids. Dolomite and
- magnesite crystallized at relatively low temperature, from aqueous fluids that acquired their carbon
- 994 isotope ratios during higher temperature devolatilization of Hawasina clastic sediment compositions,
- would have  $\delta^{13}$ C in the range of 1.0 to -3.0‰, as observed in the MoD Mountain listvenites. In
- addition, low carbon solubilities in low temperature, low pressure aqueous fluids saturated in
   carbonate minerals in mineral assemblages similar to those in the Hawasina clastic sedimentary
- carbonate minerals in mineral assemblages similar to those in the Hawasina clastic sedimentary
   rocks (Figure 22 and associated text in Chapter 4, Falk 2014, Kelemen & Manning 2015) may be
- insufficient to produce the MoD Mountain listvenites (Falk & Kelemen 2015)de Obeso, 2021 #300}.
- 1000
- Based on the data and reasoning described in the previous paragraph, we hypothesize that 1001 1002 subduction zone devolatilization at 400 to 600°C produced CO2-rich aqueous fluids that then cooled 1003 and decompressed by flow up the subduction zone, and reacted with peridotite at less than 200°C to produce the MoD Mountain listvenites. To quantify this hypothesis, we made thermodynamic 1004 calculations with the compositions of solid reactants given in Supplementary Table 2, methods 1005 described in Section 3, and results outlined in Figures 14 and 15. As the source of fluid, we chose 1006 1007 sample OM20-17, a pelitic end-member from among the Hawasina clastic sedimentary rocks 1008 analyzed by Falk & Kelemen (2015) and de Obeso et al. (2021a). As the peridotite reactant, we used 1009 an average Samail harzburgite composition calculated from published studies (Godard et al 2000,
- 1010 Hanghoj et al 2010, Monnier et al 2006).
- 1011

Devolatilization of clastic Hawasina sediments, including OM20-17, is predicted to produce fluids with
~ 20,000 ppm dissolved C at ~ 400°C to ~ 600°C along subduction zone geotherms (Figure 14).
Closed system decompression and cooling of fluid produced from OM20-17 to 100 to 300°C, and 0.5
to 1 GPa produced no significant change in the composition of this modeled fluid. (Modeling open
system, reactive transport of this fluid, updip along a subduction zone geotherm, is beyond the scope
of this paper, as is modeling the fate of subduction zone fluids that have more than 10 wt% CO2 –

1018 predicted for mixed siliciclastic-carbonate rocks (Figure 14) – as they cool and decompress).

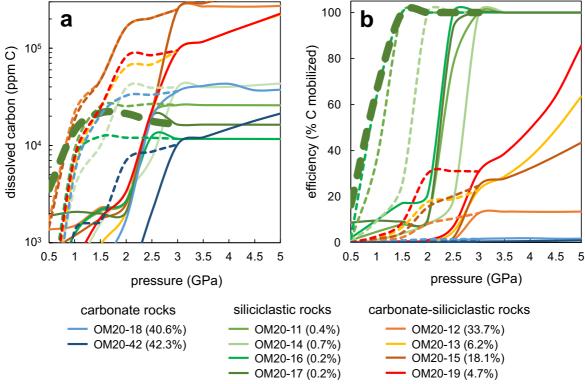




Figure 14: Calculated dissolved carbon concentrations at a water/rock ratio of 5% (a) and extent of 1020 carbon removal (b) for samples of Hawasina sediments in the MoD Mountain area (Supplementary 1021 Table 2 in de Obeso et al 2021a), evolving along cold (550°C at 2.5 GPa) and hot (500°C at 1.5 GPa) 1022 subduction geotherms. Solid curves are results for the average ("cold") subduction zone geotherm 1023 1024 from Syracuse et al. (2010). Dashed curves follow a hot geotherm suggested by Penniston-Dorland et al. (2015). The initial carbon contents of the rocks (wt%, all carbon as CO<sub>2</sub>) are given in the legend. 1025 1026

1027 1028 5.7 Listvenite formation

Reaction of the model fluid from OM20-17 with peridotite at 100-300°C and 0.5 to 1.0 GPa is 1029 1030 predicted to produce mineral assemblages similar to those modeled by Klein and Garrido (2011): (1) Small masses of "birbirite" (silicified peridotite) at high water/rock ratios and/or low temperatures, 1031 1032 through (2) moderate masses of listvenite and soapstone (talc-carbonate rocks) at moderate water/rock and temperature, to (3) relatively large masses of carbonate-bearing serpentinite at low 1033 1034 water/rock ratios and/or high temperature (Figure 15). Predicted magnesite and quartz proportions correspond closely to observed proportions in MoD Mtn listvenites (Figure 12). Most of the PT 1035 1036 conditions we modeled produced small amounts of hematite coexisting with magnesite and quartz, as observed. Predicted magnesite and siderite proportions correspond to a solid solution with ~ 8 wt% 1037 FeO, which is a few wt% higher than observed in MoD Mtn listvenites. Most modeled conditions 1038 1039 produced dolomite in listvenite assemblages at water/rock ratios at water/rock ratios less than 10 (log water/rock = 1), consistent with the presence of relatively late, cross-cutting dolomite-bearing veins in 1040 1041 the listvenites. Most model runs produce small amounts of kaolinite, and very limited proportions of 1042 muscovite, in listvenite mineral assemblages, consistent with the widespread presence of minor 1043 amounts of chromian white mica in MoD Mtn listvenites, and in listvenites worldwide. It is possible that addition of a thermodynamic model for fuchsite would yield stable white mica, rather than kaolinite, 1044

- over a wider range of temperature and water/rock ratios. Alternatively, perhaps some of the green
   sheet silicates in listvenites are chrome-bearing clays rather than true micas.
- 1047

Thus, thermodynamic modeling suggests that the CO<sub>2</sub>-bearing aqueous fluids that formed the MoD Mountain listvenites formed by metamorphic devolatilization in a subduction zone at > 400°C. These fluids then migrated updip to react with peridotite at the leading edge of the mantle wedge, probably at a depth less than 40 km. However, this is just a forward model, and there may be other possibilities.

- 1053 Subhorizontal lenses of listvenite at MoD Mountain contain a cumulative mass of about 2 billion tons 1054 of CO<sub>2</sub> over a strike length of 2 km NS, and 5 km EW, corresponding to 1 million to 400,000 tons of CO<sub>2</sub> per m along strike. The allochthonous sedimentary units below the ophiolite are about 3 km thick. 1055 1056 Within these, clastic units comprise at least half the section, and contain about 2300 ppm C, or 0.84 1057 wt% CO<sub>2</sub> on average (de Obeso et al 2021a), yielding a total of about 35 tons CO<sub>2</sub> per m along strike, per m subducted. (As noted above, rocks composed mainly of calcite and/or dolomite in the 1058 subducting sedimentary section would be unlikely to contribute significant amounts of CO<sub>2</sub> to 1059 subduction fluids at temperatures less than 800°C). At subduction velocities of 0.05 to 0.1 m/year, 1060 1061 90% decarbonation of the clastic units in the Hawasina Formations with a density of 2.75 tons/m<sup>3</sup>, at  $\sim$ 1062 400-500°C would produce at least 1.7 to 3.5 tons of CO<sub>2</sub> per year per m of strike length. If most of this 1063 CO<sub>2</sub> reacted with peridotite at the depth and temperature of MoD Mtn listvenite formation, this could supply the observed mass of CO<sub>2</sub> at MoD Mountain in less than 600,000 years. 1064
- 1065

Most of the model results at different conditions predict precipitation of a few weight percent magnesite (and some dolomite) in the serpentinization domain. This is consistent with observation of carbonate veins in serpentinites from Hole BT1B, and with the hypothesis that formation of carbonate veins in the serpentinite zone preceded transformation of the serpentinite host rocks to listvenite (also see Figure 6 and associated text in Section 4.4,

1071

1072 The fluid temperatures, compositions and fluxes used in these calculations are different from the constraints used in some calculations by Falk & Kelemen (2015). In that previous work, we explored 1073 1074 the possibility that CO<sub>2</sub> to form the listvenites was supplied over tens of millions of years, carried in 1075 100 to 200°C fluids containing a few hundred ppm dissolved carbon, derived from pore fluids and/or dewatering of opal and clay minerals, from the Hawasina sedimentary rocks immediately below the 1076 1077 site of listvenite formation at MoD Mountain. However, one of us (Falk!) insisted on mentioning the possibility that the CO<sub>2</sub> to form listvenites was derived from fluids formed deeper in the subduction 1078 1079 zone, that migrated updip. We now prefer this latter hypothesis, for the reasons outlined above. 1080

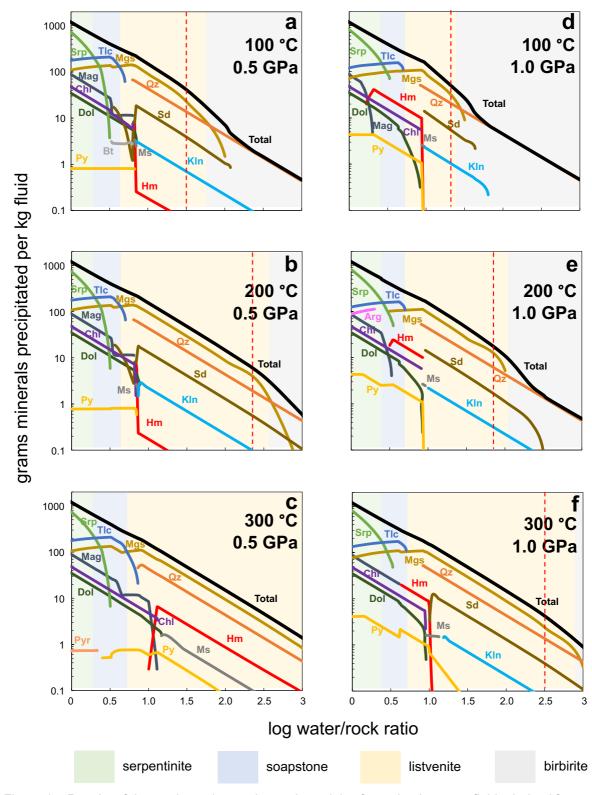


Figure 15: Results of thermodynamic reaction path models of reaction between fluids derived from devolatilization of Hawasina pelitic sedimentary rock sample OM20-17 (Section 5.6) and average Oman harzburgite. Mineral end-member abbreviations Qz quartz, Sd siderite, Mgs magnesite, Kln kaolinite, Ms muscovite, Dol dolomite, Chl chlorite, Py pyrite, Tlc talc, Srp serpentine (chrysotile), Pyr pyrrhotite. Red, vertical dashed line indicates where magnesite/quartz molar and volume proportions are ~ 2:1, as observed in magnesite listvenites from MoD Mtn (e.g., Figure 12).

Updip migration of fluids in a subduction zone has been predicted in some simplified dynamic models
 of fluid flow in a viscously deforming subduction zone with high permeability (Wilson et al 2014).
 However, the tendency of fluid buoyancy to drive vertical fluid flow may often dominate subduction

zone fluid fluxes. Thus, formation of relatively shallow listvenites, like those at MoD Mountain, may be
 localized and unusual. Elsewhere, CO<sub>2</sub>-bearing fluids may migrate vertically into overlying mantle
 peridotite at greater depth (e.g., Kelemen & Manning 2015). It is interesting to ponder how subduction
 zone CO<sub>2</sub> fluxes may be partitioned between these different transport and mineralization processes.

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## 5.8 Multiple reaction fronts

The thermodynamic models presented in Sections 5.6 and 5.7 provide a starting point for 1099 1100 understanding "chromatographic effects" during transformation of peridotite to serpentinite. As can be 1101 seen in Figure 15, our reaction path models predict sharp fronts where serpentinite is replaced by soapstone, and then by listvenite. Talc-bearing, soapstone assemblages are predicted to crystallize in 1102 1103 a limited range of conditions, consistent with the fact that talc is rare in core from Hole BT1B, and in hand specimens from MoD Mtn, where it is almost entirely restricted to narrow (~1 m scale) transition 1104 1105 zones between listvenite and serpentinite. If we were to use different thermodynamic data, talc might 1106 be even less abundant, or absent in models at 100°C, because talc is predicted to be unstable below 1107 ~ 100°C with respect to antigorite + quartz when using mineral data from Holland and Powell or Gottschalk, together with the Redlich-Kwong equation of state for H<sub>2</sub>O- CO<sub>2</sub> fluids as modified by 1108 Kerrick and Jacobs (1981) and by Holland and Powell (2003). 1109

1110

1111 In the model results presented in Figure 15, with reaction progress increasing from high fluid/rock 1112 ratios on the right to low fluid/rock ratios on the left, magnesite + quartz and magnesite + talc become 1113 unstable with respect to serpentine as dissolved carbon in the fluid is exhausted. Because carbon is a 1114 minor constituent of aqueous fluid, but a major component of the listvenite assemblage, exhaustion of 1115 dissolved carbon is predicted to occur at a fluid/rock ratio much greater than 1 (log fluid/rock > 0). Of 1116 course, because aqueous fluid is composed mainly of H<sub>2</sub>O, the potential for serpentinization of 1117 peridotite continues to much lower fluid/rock ratios.

1118

Although we cannot model it, we can also predict that – in the presence of pervasive fluid flow on the grain scale – there could also be a sharp front where serpentine replaces olivine at water/rock ratios

1121 less than one. Thermodynamic calculations for simplified olivine serpentinization by Kelemen et al.

1122 (2020a) indicate that olivine +  $H_2O$  would be stable with respect to serpentine at 100°C and a partial

1123 pressure of ~  $10^{-2}$  bars, and at 200°C and P(H<sub>2</sub>O) ~ 1 bar. While these conditions cannot be modeled

using EQ3/6, we can anticipate that low partial pressures of  $H_2O$  – much lower than lithostatic

- 1125 pressures are produced along grain boundaries and near the tips of incipient fractures and veins,
- especially where fluid has been almost completely consumed by peridotite hydration reactions. Under
- these conditions, there could be a sharp front where serpentine (at higher fluid pressures) becomes
- 1128 stable relative to olivine (at lower fluid pressure).

- Throughout the mantle section of the Samail ophiolite, residual mantle peridotites commonly contain 1130 1131 about 50 to 80% serpentine, as inferred from the fact that bulk rock analyses yield loss on ignition 1132 (mostly, H<sub>2</sub>O) of 8 to 10 wt% as compared to 13 to 16 wt% H<sub>2</sub>O in completely serpentinized. Mg end-1133 member harzburgite and dunites. These partially serpentinized peridotites commonly show a "mesh texture", with relict olivine and pyroxene "cores" transected by a "mesh" of cross-cutting serpentine 1134 veins - typically 10 to 100 microns apart (Francis 1956, Green 1961, Green 1964, Raleigh & Paterson 1135 1965). In some regions within the Samail ophiolite, particularly areas of relatively subdued topography 1136 that have undergone extensive, penetrative weathering, relict mantle minerals in the mesh cores are 1137 1138 completely replaced by serpentine (e.g., OmanDP Sites BA1, BA2, BA3 and BA4, Kelemen et al 2021, Kelemen et al 2020c). However, along the steep canyons and narrow ridges that are typical of 1139 outcrops in the mantle section of the ophiolite, subject to relatively rapid erosion, the pervasive 1140 1141 presence of the serpentine vein mesh surrounding relict mantle minerals attests to relatively rapid fluid transport in fractures and veins, compared to slow transport of  $H_2O$  into the mesh cores by 1142 1143 diffusion and/or imbibition. 1144
- 1145 In contrast, as noted above, the serpentinites sampled in core from Hole BT1B contain no relict olivine 1146 or orthopyroxene, though pyroxene pseudomorphs ("bastites") are evident. A zone of 100% 1147 serpentinized peridotites a few meters thick was sampled by Falk & Kelemen (2015) in a transect across a listvenite-peridotite contact on the watershed ridge east of the summit MoD Mountain. 1148 Outside this zone, samples of peridotite had compositions and textures typical of partially 1149 serpentinized residual mantle peridotites throughout the ophiolite. Based on these observations, we 1150 1151 infer that there is a 100% serpentinite zone a few meters thick, with sharp fronts – less than a meter 1152 thick, between serpentinte and listvenite on one side, and between partially and completely serpentinized peridotite on the other side. 1153
- 1154
- 1155 1156

## 5.9 Volume change during listvenite formation

The likely volume change during fluid-rock reactions has long been debated. Whereas Coleman & 1157 1158 Keith (1971) proposed that serpentinization involved simple addition of  $H_2O$  to peridotite, with no significant change in the volatile-free solid composition, Carmichael (1987), Nahon & Merino (1987), 1159 and Fletcher & Merino (2001) argued that such reactions take place at nearly constant volume, with 1160 1161 addition of some components balanced by dissolution and export of others. Fletcher & Merino provided quantitative calculations to support this hypothesis. Where a fluid that is super-saturated in a 1162 new mineral phase, A, starts to crystallize A within a host mineral B that is initially in equilibrium with 1163 1164 fluid, expansion of B around A leads to an increase in local effective stress. In turn, because chemical 1165 potential is proportional to the mean stress, this reduces supersaturation in A and leads to 1166 undersaturation in B. This process continues, with very small volume changes, until the rate of crystallization of A becomes equal to the rate of dissolution of B, at a steady state stress. 1167 1168

Using the methodology of Fletcher and Merino (2001) and a saturation index of 2 (as they did), 1169 Kelemen and Hirth (2012) calculated that the steady state, effective stress during replacement of 1170 1171 olivine with serpentine is ~ 40 MPa. However, because olivine is very far from equilibrium with water 1172 at low temperature, the saturation index for water reacting with olivine to form serpentine at 50 to  $250^{\circ}$ C and P(H<sub>2</sub>O) > 1 bar is close to  $10^{7}$ , and the steady state, effective stress estimated using the 1173 method of Fletcher and Merino is ~ 800 MPa (Kelemen & Hirth 2012). Clearly, such a large differential 1174 stress cannot be sustained within most rocks, which will deform at a lower stress, before the steady 1175 state can be reached, either via ductile mechanisms (if reactions are slow and temperatures are high) 1176 or via fracture and frictional deformation (if reactions are fast and temperatures are low). The latter 1177 1178 outcome is sometimes termed "reaction-induced cracking" (Jamtveit et al 2009, Rudge et al 2010) or "reaction-driven cracking" (Kelemen & Hirth 2012). Thus, while some workers still disagree over the 1179 1180 extent of volume change in specific replacement processes, the approach of Fletcher and Merino 1181 nicely explains a continuum between nearly constant volume replacement of one mineral by another at high temperature, close-to-equilibrium conditions, and large volume changes accommodated by 1182 fractures and frictional sliding at low temperature, far-from-equilibrium conditions. 1183

1184

1185 As noted in Section 5.5, despite dramatic, local variability, the average major element composition of 1186 listvenites from MoD Mountain - in drill core and surface samples - is strikingly similar to the average 1187 composition of residual mantle harzburgites in the Samail ophiolite. Together, the concentrations of MgO, SiO<sub>2</sub> and FeO\* (all Fe as FeO) account for more than 90% of the volatile-free bulk composition 1188 of listvenite samples. Thus, the fact that average Mg/Si/Fe ratios in the listvenites are almost identical 1189 to those in Samail peridotites suggests that either (1) large scale dissolution of the peridotite protoliths 1190 1191 was nearly congruent, exporting dissolved, major elements in approximately their initial proportions, or 1192 (2) there was little dissolution and export of major elements in the protolith during addition of  $CO_2$  to form listvenites, and addition of H<sub>2</sub>O to form serpentinites. 1193

1194

Thermodynamic models, experimental data, and observations of natural rock samples strongly favor 1195 1196 the second of these hypotheses. The solubility of silicate and Fe-oxide minerals in rock-buffered aqueous fluids at low temperature is too low to allow for large scale dissolution and export of major 1197 elements in open system, fluid-rock reactions. In turn, models of addition of CO<sub>2</sub> and/or H<sub>2</sub>O to 1198 1199 peridotites, predict very little removal of other species, togther with large increases in the mass and volume of the solid products of reaction. Birbirite formation may involve net volume and mass loss 1200 1201 due to extensive dissolution of peridotite reactants at water/rock ratios greater than ~ 100. However, the models of listvenite formation illustrated in Figure 15 yield predicted mass and volume increases 1202 of 10 to 55 percent relative to an anhydrous, peridotite reactant, as illustrated in Figure 16. Similar 1203 1204 results have been produced by thermodynamic models of serpentinization (volume increase of 40-1205 60%, de Obeso & Kelemen 2018, Malvoisin 2015), experimental observation of closed system 1206 serpentinization (30-60%, Klein & Le Roux 2020) and analysis of microstructures in partially serpentinized peridotites formed in an open system (59±30 to 74±36%, Malvoisin et al 2020). 1207 1208

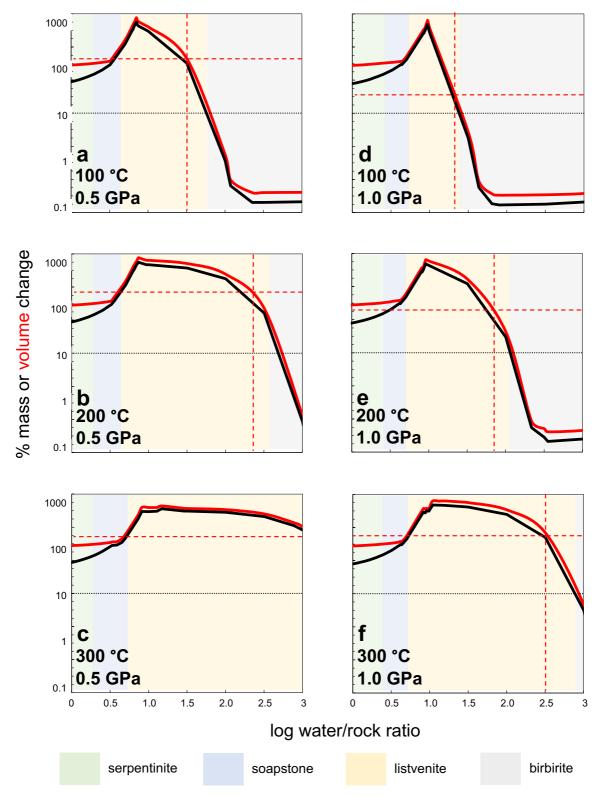


Figure 16: Calculated mass and volume change relative to an anhydrous peridotite, for the reaction path models illustrated in Figure 15. Dashed, vertical red lines indicate where molar and volume 1211 proportions of magnesite/quartz reach 2:1, as onserved in MoD Mtn listvenites (e.g., Figure 12). 1212

Dashed, horizontal line highlights the minimum increase in solid volume calculated for listvenite 1213 formation. 1214

1216	These modeled and observed volume changes approximate those resulting from simplified, Fe-fre	e,
1217	stoichiometric reactions. Thus, hydration of olivine to form serpentine and brucite,	
1218	$2Mg_2SiO_4 + 3H_2O = Mg_3Si_2O_5(OH)_4 + Mg(OH)_2$	(1)
1219	and hydration of olivine + orthopyroxene to form serpentine,	
1220	$Mg_2SiO_4 + MgSiO_3 + 2H_2O = Mg_3Si_2O_5(OH)_4$ ,	(2)
1221	can produce 52 and 63% increases in the solid volume, respectively. (Volume change calculated a	as
1222	100% (product volume -reactant volume)/(reactant volume)). Direct carbonation of olivine	
1223	$Mg_2SiO_4 + 2CO_2 = 2MgCO_3 + SiO_2,$	(3)
1224	and olivine + orthopyroxene,	
1225	$Mg_2SiO_4 + MgSiO_3 + 3CO_2 = 3MgCO_3 + 2SiO_2,$	(4)
1226	can lead to 85% and 74% increases in the solid volume, respectively. And, carbonation of serpent	ine
1227	plus brucite	
1228	$Mg_3Si_2O_5(OH)_4 + Mg(OH)_2 + 4CO_2 = 4MgCO_3 + 2SiO_2 + 3H_2O_3$	(5)
1229	and serpentine alone	
1230	$Mg_3Si_2O_5(OH)_4 + 3CO_2 = 3MgCO_3 + 2SiO_2 + 2H_2O_3$	(6)
1231	both produce solid volume increases ~ 22%.	
1232		

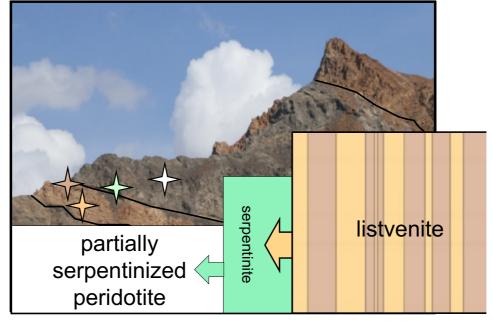


Figure 17: Schematic illustration of sequential volume changes during replacement of partially serpentinized peridotite by serpentinites and then replacement of serpentinites by listvenites. The height of the rectangles corresponds to the relative volumes, produced by reaction of CO2-bearing fluid with an initial, anhydrous peridotite. The tan and brown stripes in the "listvenite" represent alternations of different listvenite compositional bands. Stars on photo of the SW side of MoD Mtn illustrate position of listvenite, serpentinite and partially serpentinized peridotite samples (Falk & Kelemen 2015).

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1242 Because porosities in peridotite, serpentinite and listvenite rarely if ever exceed 5%, all of these solid

- 1243 volume changes must be accommodated mainly by expansion of the entire rock volume. It is
- 1244 interesting to speculate how much uplift and/or lateral expansion in forearc regions is caused by

- hydration and carbonation of the mantle wedge. However, at the plate tectonic scale, the rates of
  reactions similar to those outlined in equations (1-6) are unknown. It is likely that the strains due to
  reaction are comparable to, or smaller than, other rates of deformation at convergent plate
- boundaries, rendering the effect of solid volume expansion difficult to detect at the regional scale.
- 1249

Based on the considerations outlined in this section, it is likely that large increases in the solid volume occurred during formation of the MoD Mountain listvenites, and were accommodated mainly by reaction-driven cracking, frictional sliding along existing fractures, and/or reaction-assisted granular flow (Menzel et al 2021). The presence of abundant, antitaxial magnesite, magnesite-hematite, and Fe-oxide veins in both serpentinites and listvenites is qualitatively consistent with a reaction-driven cracking process. However, we have not identified any obvious strain markers that would allow a quantitative evaluation of this hypothesis using rock textures.

1257

Building upon an idea from Hansen et al. (2005), the Shipboard Scientific Party developed the
hypothesis that large volume increases due to hydration of olivine and pyroxene (reactions 1 & 2) may
have initially formed fractures at (and beyond) a serpentinization front – not observed in drill core, but
traversed by Falk & Kelemen (2015) – and these fractures were conduits for fluid flow and sites of
localized deformation during the smaller volume changes due to carbonation of serpentinites
(reactions 5 and 6) as schematically illustrated in Figure 17.

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# 5.10 Low temperature ductile deformation in subduction zones

As noted above, the listvenites at MoD Mountain may have formed at depths of 25 to 40 km, at temperatures less 200°C, yielding low temperature "geotherms" of 5 to 8°C/km depth. Such small increases of temperature with depth are characteristic of the forearc above subduction zones (Peacock 1996). High fluid pressures at these depths may account for the somewhat surprising indications of low temperature, ductile deformation in core samples from Hole BT1B, based on the observed crystallographic preferred orientation (CPO) in quartz, magnesite and serpentine in core samples (Section 4.4, Figure 10).

1274

Some of the fabrics illustrated in Figure 10 could be inherited. For example, in early formed, magnesite-hematite veins, magnesite forms parallel crystals, perpendicular to vein margins, with small misorientations between adjacent crystals. When these veins are parallel (because they form that way, or after they are transposed), this imparts a CPO to the sample, and may also give the impression that sub-grain boundaries are present, even if there was no deformation of magnesite crystals via dislocation creep. Similarly, quartz replacing opal may inherit a CPO, or a CPO may arise due to anisotropic stress during recrystallization, without substantial strain.

1282

1283 On the other hand, the serpentine mylonite illustrated in Figure 10 almost certainly records large 1284 strains due to distributed deformation along grain boundaries. Moreover, clear examples of ductile deformation and shear zones with classical indictors of substantial strain indicate that ductile
deformation was active during the initial stages of listvenite formation, perhaps assisted by positive
feedback between weakening, due to reaction-induced recrystallization, and porosity enhancement
due to deformation, as discussed by Menzel et al. (2021). An important role for ductile deformation in
reaction zones at the top of subducting oceanic crust may help to explain why, in the relatively hot
Cascadia and SW Japan subduction zones, there are very few earthquakes at the top of the
subducting crust (Abers et al 2009, Abers et al 2013, Hirose et al 2008).

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#### 6. Conclusions

Observations of drill core of listvenite (completely carbonated peridotite), serpentinite, and 1295 subduction-related metamorphic rocks from OmanDP Hole BT1B provide constraints on temperature, 1296 depth, and deformation during mass transfer of H<sub>2</sub>O and CO<sub>2</sub> from subducted sediments into 1297 overlying mantle peridotites at the leading edge of the mantle wedge. Listvenites, and a surrounding 1298 zone of serpentinite, formed at temperatures less than ~ 200°C and poorly constrained depths of 25 1299 1300 to 40 km. Serpentinization and carbonation involved reaction of partially serpentinized, residual 1301 mantle peridotite with CO2-rich, aqueous fluids produced by devolatilization of subducting, clastic 1302 sediments analogous to the Hawasina formation, probably at 400 to 600°C. These fluids cooled as 1303 they were transported up the subduction zone to the site of listvenite formation.

1304

Such processes could play a significant role in the global carbon cycle. Not all listvenites form above
subduction zones. However, listvenites are found at and near the basal thrust in other ophiolites,
worldwide (Akbulut et al 2006, Borojević Šoštarić et al 2014, Escayola et al 2009, Menzel et al 2018,
Quesnel et al 2016, Quesnel et al 2013, Scarsi et al 2018, Sofiya et al 2017, Ulrich et al 2014). The
leading edge of the mantle wedge – and subduction modified mantle that has later been incorporated
into the continental mantle lithosphere – may be a globally important reservoir for carbon (Foley &
Fischer 2017, Kelemen & Manning 2015, Li et al 2017, Scambelluri et al 2016)

1312

During BT1B listvenite formation, total solid volume increased by tens of percent due to hydration followed by carbonation. While core and surface samples provide few direct constraints on the mechanism that accommodated this expansion, one hypothesis is that large volume changes during hydration of olivine and pyroxene along a serpentinization front caused large stresses and fractures that accommodated expansion via frictional sliding, and provided secondary porosity for the CO<sub>2</sub>-rich fluids that transformed serpentinites to listvenites.

1319

1320 Based on observed crystallographic preferred orientation in quartz, magnesite and serpentine in

1321 macroscopically identified shear zones, it is inferred that ductile deformation of listvenite and

1322 serpentinite occurred under low temperature conditions at the base of the mantle wedge during

1323 subduction. Low temperature ductile deformation, coeval with serpentinization and listvenite

1324 formation, may have been facilitated by recrystallization associated with the hydration and carbon

mineralization processes, as discussed in more detail by Menzel et al. (Menzel et al 2021). Such a
 process could be active in subduction zones where the interface between subducting oceanic crust,
 sediments, and hanging wall peridotites is aseismic.

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- 1329 1330

## 6. Author contributions

Kelemen and de Obeso conducted field mapping at MoD Mountain. de Obeso made new Sr and C 1331 isotopic analyses of listvenites, the metamorphic sole and the underlying sedimentary rocks, updating 1332 1333 work by Falk & Kelemen (2015). Stockli carried out the reconnaissance (U,Th)/He analyses of zircons 1334 from the metamorphic sole and lower curst, using zircon separates samples provided by Rioux. Godard expertly led the BT1B geochemistry team onboard Drilling Vessel Chikyu, and then heroically 1335 continued with analyses at the Université de Montpellier on behalf of the Shipboard Scientific Party. 1336 1337 Okazaki labored with Kelemen on the triangular histogram of shipboard XRF scanner data. Leong and de Obeso conducted the EQ3/6 thermodynamic modeling, in consultation with Kelemen. Working with 1338 1339 his computer in 46°C weather on the drill site, Manning provided key insights into conditions for coexisting hematite and organic carbon compounds. Ellison supervised Kelemen's Raman 1340 1341 spectroscopy analyses at the University of Colorado, Boulder, and offered essential advice and data 1342 interpretation. Kotowski led analysis of core from the metamorphic sole and shared her results. Urai 1343 led the structural geology team during core description onboard DV Chikyu, and is now advising Menzel, who is leading analysis and interpretation of microstructures. Hirth offered insights into 1344 potential mechanisms of low temperature, ductile deformation in subduction zones. Lafay and Beinlich 1345 provided valuable input on the drill site and as members of the Shipboard Scientific Party. Coggon 1346 1347 (Project Manager) supervised drilling, core curation and logistics for this and all other OmanDP 1348 boreholes, together with Nehal Warsi (Site Geologist). Matter (Project Director), Teagle (ICDP Lead Investigator) and Sulaimani (Country Manager) worked tirelessly to ensure the success of the Oman 1349 Drilling Project. Harris, Kelemen, Michibayashi, and Takazawa served as Co-Chief Scientists onboard 1350 DV Chikyu during description of core from Hole BT1B. Kelemen (Chief Scientist) helped lead the 1351 1352 project, had a few ideas, and took the lead in writing this manuscript.

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- 1355

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1390

1391 Data Availability: Samples in Supplementary Table 1 have IGSN numbers and locations. Data in

1392 Supplementary Table 1 are in the process of being archived in the Geochron database

1393 (www.geochron.org). Figures 4, and 6 are compilations of images that are published at

1394 <u>http://publications.iodp.org/other/Oman/OmanDP.html</u>, where they are freely available, with more

1395 detailed references provided in the figure captions. Figures 1, 2, 5, and 7-17, Supplementary Figures

1396 **1** and 2, and Supplementary Tables 1 and 2 constitute data that are original with this paper. When

1397 this manuscript is accepted, the tabulated data will uploaded to the Zenodo repository

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