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

Juan Carlos de Obeso¹, Peter B Kelemen², James Andrew Leong³, Manuel D Menzel⁴, Craig Manning⁵, Marguerite Godard⁶, Yue Cai⁷, and Louise Bolge²

¹University of Calgary ²Columbia University ³Lamont Doherty Earth Observatory ⁴RWTH Aachen University ⁵University of California Los Angeles ⁶Universite de Montpellier ⁷Lamont-Doherty Earth Observatory

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Abstract

Completely carbonated peridotites represent a window to study reactions of carbon-rich fluids with mantle rocks. Here we present details on the carbonation history of listvenites close to the basal thrust in the Samail ophiolite. We use samples from Oman Drilling Project Hole BT1B, which provides a continuous record of lithologic transitions, as well as outcrop samples from listvenites, metasediments and metamafics below the basal thrust of the ophiolite. ⁸⁷Sr/⁸⁶Sr of listvenites and serpentinites, ranging from 0.7090 to 0.7145, are significantly more radiogenic than mantle values, Cretaceous seawater, and other peridotite hosted carbonates in Oman. δ^{13} C in the listvenites and serpentinites range from -10.6carbon component with δ^{13} C as low as -27reaffirms the presence of carbonaceous material in Hole BT1B. The source of the radiogenic Sr was probably similar to Hawasina metasediments that underlie the ophiolite, with values up to 0.7241 in clastic lithologies. These results indicate that decarbonation reactions in such clastic sediments, during subduction at temperatures above 500°C, form carbon rich fluids that could have migrated updip, supplying radiogenic ⁸⁷Sr/⁸⁶Sr and fractionated δ^{13} C to BT1B serpentinites and listvenites.

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4 Juan Carlos de Obeso^{1,2}, Peter B. Kelemen², James M. Leong², Manuel D. Menzel³, Craig

E. Manning⁴, Marguerite Godard⁵, Yue Cai², Louise Bolge² and Oman Drilling Project 5

- 6 **Phase 1 Science Party**
- 7 ¹Department of Geosciences, University of Calgary, Calgary, Canada
- 8 ² Lamont Doherty Earth Observatory Columbia University, Palisades, NY, USA.
- 9 ³ Institute of Tectonics and Geodynamics, RWTH Aachen University, Aachen, Germany.
- 10 ⁴ Dept. of Earth & Space Sciences, University of California, Los Angeles, CA, USA.
- ⁵ Géosciences Montpellier, Université de Montpellier, Montpellier, France. 11
- 12
- 13 Corresponding author: Juan Carlos de Obeso (juancarlos.deobeso@ucalgary.ca)
- 14
- 15 **Key Points:**
- 16 Strontium and Carbon were added to the peridotites during alteration of mantle peridotite • 17 with carbonated fluid derived from decarbonation reaction.
- 18

19 Abstract

- 20 Completely carbonated peridotites represent a window to study reactions of carbon-rich fluids
- 21 with mantle rocks. Here we present details on the carbonation history of listvenites close to the
- 22 basal thrust in the Samail ophiolite. We use samples from Oman Drilling Project Hole BT1B,
- which provides a continuous record of lithologic transitions, as well as outcrop samples from $\frac{24}{100}$
- listvenites, metasediments and metamafics below the basal thrust of the ophiolite. ⁸⁷Sr/⁸⁶Sr of
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 than mantle values, Cretaceous seawater, and other peridotite hosted carbonates in Oman. The
- Hawasina metasediments that underlie the ophiolite, on the other hand, show higher ⁸⁷Sr/⁸⁶Sr
- values of up to 0.7241. δ^{13} C values of total carbon in the listvenites and serpentinites range from
- 29 -10.6% to 1.92%. We also identified a small organic carbon component with δ^{13} C as low as -
- 30 27‰. Based on these results, we propose that during subduction at temperatures above 500°C,
- 31 carbon-rich fluids derived from decarbonation of the underlying sediments migrated up dip and
- 32 generated the radiogenic 87 Sr/ 86 Sr signature and the fractionated δ^{13} C values of the serpentinites
- 33 and listvenites in core BT1B.
- 34

35 Plain Language Summary

36 Samples from Oman Drilling Project Hole BT1B provide a record of interactions of fluids rich in

37 carbon dioxide with mantle rocks. This interactions lead to the formation of listvenites, rocks

38 composed mainly by magnesite and quartz. Here we describe the formation of listvenites in the

39 Oman ophiolite using Strontium and Carbon isotopes to characterize the source and nature of the

40 fluid that pervasively transform the mantle rocks that now store vast amounts of carbon dioxide.

41 **1 Introduction**

42 Hydration and carbonation of ultramafic rocks are important processes in the carbon and 43 water cycle of our planet (Alt et al., 2013; Fruh-Green et al., 2004). These alteration reactions are sinks of water and carbon where peridotites are exposed on the seafloor forming alteration 44 45 minerals like serpentine and carbonates (Alt et al., 2013; Klein et al., 2020; Macdonald & Fyfe, 46 1985; Paulick et al., 2006) which are carried back into the mantle in convergent margins. Fluids 47 derived from the subducted slab can migrate and interact with the mantle wedge in subduction 48 zones, so that the "leading edge of the mantle wedge" aka the "cold nose", can be partially 49 hydrated and carbonated (Blakely et al., 2005; Hyndman & Peacock, 2003; Peter B. Kelemen & 50 Manning, 2015). This is usually inferred from seismic data (e.g. DeShon and Schwartz, 2004; 51 Kamiya and Kobayashi, 2000; Tibi et al., 2008; Tsuji et al., 2008). Understanding the interaction 52 of carbon-bearing hydrous fluids with peridotites is important to supplement and constrain 53 geophysical observations.

54

Fully carbonated peridotites, also known as listvenites, provide a window into the alteration processes that occur in the "cold nose" of the mantle wedge above subduction zones, where mantle peridotite reacts with hydrous and carbonated fluids likely derived from the subducting slab at moderate temperatures and pressures (Beinlich et al., 2012; Boskabadi et al., 2017, 2020; Falk & Kelemen, 2015; Manuel D. Menzel et al., 2018). Formation of listvenites appears to be restricted to particular conditions requiring high carbon concentrations in the fluid 61 (Falk & Kelemen, 2015; Hansen et al., 2005; Peter B. Kelemen et al., 2021; Manuel D. Menzel

- 62 et al., 2018). For example, complete carbonation of peridotites to form listvenites is not observed
- near the surface in ophiolites (e.g. Clark and Fontes, 1990; de Obeso and Kelemen, 2020, 2018;
 Garcia del Real et al., 2016; Kelemen and Matter, 2008; Noël et al., 2018; Quesnel et al., 2016;
- 65 Schwarzenbach et al., 2016). It is also not observed near the seafloor, where serpentinization and
- 66 formation of carbonate veins are common (e.g Bach et al., 2011; Delacour et al., 2008;
- 67 Schwarzenbach et al., 2013). Fluid sources for complete carbonation have been associated with
- 68 subduction in different listvenite localities. Menzel at al. (2018) attributed carbonation of
- 69 harzburgite in the Advocate ophiolite (Canada) to fluxing by slab-derived, CO₂-rich fluids.
- 70 Isotopic data (¹³C, ¹⁸O and ⁸⁷Sr/⁸⁶Sr) of listvenites in the Late Cretaceous ophiolites of
- 71 eastern Iran point to carbon-bearing fluids derived from subducted sedimentary units as the
- 72 source of carbon (Boskabadi et al., 2020).

73 In Oman, listvenites occur along the basal thrust of the ophiolite (Nasir et al., 2007; 74 Wilde et al., 2002). Previous studies have investigated their formation conditions and the nature 75 of the carbonation fluids without reaching conclusive answers on the source of the fluids (Beinlich et al., 2020; Falk & Kelemen, 2015; Nasir et al., 2007; Stanger, 1985). In this paper, 76 77 we present ⁸⁷Sr/⁸⁶Sr and ¹³C data on samples from Oman Drilling Project Hole BT1B and the 78 underlying sediments of the Hawasina Formation. We show that devolatization of the subducting 79 sediments similar to the Hawasina Formation likely generated the carbonation fluids which 80 reacted with the mantle wedge in this fossil subduction zone to form listvenites. These processes probably operate in subduction zones worldwide, where fluids migrate updip along the slab 81 82 mantle interface, and then react with hanging wall peridotites. Our results have implications for the global carbon cycle, as significant amounts of carbon could be stored in the mantle during 83

- 84 mantle wedge carbonation.
- 85

86 2 Geological setting

87 2.1 The Samail ophiolite

88 The Samail ophiolite, along the northeast coast of Oman and the United Arab Emirates 89 (UAE), is the best-exposed block of oceanic crust and its underlying mantle in the world. It was 90 thrust over adjacent oceanic lithosphere soon after magmatic formation, and then onto the margin 91 of the Arabian subcontinent in the late Cretaceous. The mantle section of the ophiolite is mainly 92 composed of highly depleted, residual mantle peridotites (mostly harzburgites, e.g. (Godard et al., 2000; Hanghøj et al., 2010; Monnier et al., 2006), together with 5 to 15% dunite (Braun, 93 94 2004; Braun & Kelemen, 2002; Collier, 2012). Near the basal thrust, interlayered dunites and 95 refertilized harzburgites comprise the distinctive "Banded Unit" (Khedr et al., 2014). The peridotites are pervasively serpentinized, with serpentine (\pm brucite) making up ~ 30-100 wt% of 96 "fresh" rock (Godard et al., 2000; Hanghøj et al., 2010; Monnier et al., 2006) and/or completely 97 98 carbonated to form listvenites (Falk & Kelemen, 2015; Nasir et al., 2007; Stanger, 1985; Wilde 99 et al., 2002). The listvenites only occur within a few km of the basal thrust of the ophiolite, and

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

102 *2.2 Lithologies below the Samail ophiolite nappe*

103 Beneath the mantle section of the Samail ophiolite is a locally preserved "metamorphic 104 sole". This sole is exposed discontinuously along the basal thrust, juxtaposed with the overlying 105 Banded Unit at the base of the Samail mantle section. It records peak metamorphic temperatures 106 of 700-900°C and imprecise peak pressures of 0.8 to 1.4 GPa (Cowan et al., 2014; Hacker & 107 Mosenfelder, 1996; M. P. Searle & Malpas, 1980; Michael P. Searle & Cox, 2002; Soret et al., 2017). A lower temperature unit (~450-550°C) with similar peak pressures (0.8 to 1.2 GPa) has 108 109 also been identified from the metamorphic section of Oman DP Hole BT1B (Kotowski et al., 110 2021). The base of the sole is in fault contact with low grade allochthonous sediments of the 111 Hawasina formation, which is composed of pelagic clastic units interlayered with limestones 112 (Bechennec et al., 1990; Béchennec et al., 1988) deposited from the late Permian to the 113 Cretaceous. The Hawasina sedimentary units were thrusted over autochthonous Mesozoic to 114 Proterozoic platform sediments of the Arabian continental margin, forming nappes between the 115 autochton and the ophiolite.

116 2.3 OmanDP Hole BT1B and Oman Listvenites

117 Hole BT1B was drilled in March 2017 in Wadi Mansah (23.364374°N, 58.182693°E), 118 which yielded a total length of 300.1 m with 100% recovery (Kelemen et al., 2020). The upper 6 119 meters are composed of alluvial gravels followed by an ultramafic sequence comprised of 120 listvenites (carbonated peridotites) interlayered with two serpentinite bands (80-100 m depth and 121 181 to 186 m depth). A thick (0.42 m) layer of grey-green fault gouge at 196.6 m-197.1 m depth 122 separates the ultramafic units from the metamorphic sole composed of fine-grained 123 metasediments and metabasalts (Kelemen et al., 2021; Kelemen et al., 2020). To the first order, 124 alteration of peridotite to form serpentinite and listvenite in Hole BT1B was nearly isochemical except for the addition of H₂O and CO₂. Average bulk rock Mg/Si, Fe/Si, Al/Si, Fe/Mg, and 125 126 Cr/Al ratios in serpentinite and listvenite are close to the average composition of the Samail 127 peridotite (Kelemen et al. 2020, Kelemen et al. 2021) and similar to the composition of 128 previously studied listvenites from the outcrops extending north and northeast from the drill site 129 (Falk & Kelemen, 2015). The core provides a unique record of the interaction between peridotite 130 in the leading edge of the mantle wedge and hydrous, CO₂-rich fluids derived from subducted 131 lithologies. For an expanded version of the geology of Oman DP Hole BT1B and MoD mountain 132 we refer the reader to the Proceedings of the Oman Drilling Project (Kelemen et al., 2020) and 133 Kelemen et al. (2021) in this Special Issue.

134 **3 Materials and Methods**

135 Samples analyzed for this study comprise a suite of drill core samples from OmanDP

136 Hole BT1B and hand samples of the Hawasina formation. Drill core samples encompass all the

137 identified lithologies from Hole BT1B. Major element compositions for Hole BT1B samples

138 were reported in the *Proceedings of the Oman Drilling Project* (Kelemen et al 2020) with the

139 exception of samples in the 181-186m depth interval which are reported by Godard et al. Trace

140 element compositions of Hole BT1B samples can be found in Godard et al. Trace element

- 141 compositions and loss on ignition for Hawasina formation outcrop samples were analyzed at
- 142 Lamont Doherty Earth Observatory (LDEO). Rb and Sr concentrations were analyzed using a
- VG PlasmaQuad ExCell quadrupole ICP-MS following HNO₃-HF digestion. Major element
- 144 compositions of the Hawasina samples are available in supplementary table 1. Additional Sr
 145 isotopes were measured on samples from outcrops northeast of Hole BT1B (Falk & Kelemen
- 145 isotopes were measured on samples from outcrops northeast of Hole BTIB (Falk & Kelemi 146 2015)
- 146 2015).

147For Sr isotope analysis, bulk rock powder was fully digested in a HNO3-HF mixture148overnight and redissolved in 3N HNO3 prior to column chemistry using the Eichrom® Sr resin.149Purified Sr were analyzed for isotopic compositions interspersed with US National Institute of150Standards and Technology (NIST) SRM 987 on a Thermo Scientific Neptune multi-collector151ICP-MS at LDEO. In-run mass fractionations were normalized to ${}^{86}Sr/{}^{88}Sr=0.1194$. Unknowns152were normalized to SRM 987 ${}^{87}Sr/{}^{86}Sr$ value of 0.701248. International standards BHVO-2153yielded ${}^{87}Sr/{}^{86}Sr$ value of 0.703509±41 (2σ , n = 3) and BCR-2 yielded 0.705046±34 (2σ , n = 3),

154 which agree with published values from Weis et al. (2006).

155 Total Carbon (TC) was measured from the same bulk rock powder splits as for Strontium 156 isotopes. Total Organic Carbon (TOC, or reduced carbon) was measured from the residual rock powder after the removal of Inorganic Carbon (carbonate carbon) through reaction with dilute (3 157 N) HCl for at least 3 days, followed by washing with Millipore[®] water. Concentrations and δ^{13} C 158 159 ratios of total carbon (TC) and total organic carbon (TOC), were determined using a Costech 160 element analyzer coupled with a Thermo Scientific Delta V plus mass spectrometer at LDEO. 161 Sample runs were calibrated using Acetanilide for carbon contents ($R^2=0.9998$). For $\delta^{13}C$ we used USGS40 ($\delta^{13}C = -26.77 \pm 0.16\%$ V-PDB, n=4), USGS41 ($\delta^{13}C = 37.63 \pm 0.12\%$ V-PDB, n=4) 162 and USGS24 ($\delta^{13}C = -16.04 \pm 0.13\%$ V-PDB, n=4). All measured values of $\delta^{13}C$ standards agree 163

- 164 with accepted values reported by the United States Geological Survey (USGS). Inorganic carbon
- 165 contents and δ^{13} C of TIC were estimated by mass balance between TC and TOC.

166 **3 Results**

- 167 Depth profiles of ⁸⁷Sr/⁸⁶Sr, Sr and Rb concentrations, carbon concentrations, Total
- 168 Carbon and Total carbon δ^{13} C are shown in Figure 1 and Table 1 and 2. The Hawasina sediments
- 169 are plotted with respect to map distance to the closest metamorphic sole/ultramafic contact.



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Figure 1. Depth profiles of OmanDP Hole BT1B and distance from basal thrust for Hawasina 172 sediments. From left to right ⁸⁷Sr/⁸⁶Sr (measured), Sr concentrations, Rb concentrations, total 173 carbon (TC) concentration, total carbon δ^{13} C. Green bands for reference from left to right are 174 average MORB⁸⁷Sr/⁸⁶Sr (measured) (Gale et al., 2013; Hofmann, 2013), Oman harzburgite 175 average Sr (ppm) and Rb (ppb) (Godard et al., 2000; Hanghøj et al., 2010; Monnier et al., 2006), 176 177 Oman harzburgite average total carbon (TC) (Peter B. Kelemen & Manning, 2015), average 178 mantle δ^{13} C (Deines, 2002). The distance from the basal thrust for the Hawasina sediments 179 (bottom panels) is measured as the horizontal distance to the closest mapped metamorphic 180 sole/ultramafic contact.

181

								TC		
								d13C	OC	TOC
	Depth	Litholog	87Sr/8	2SE total	Rb	Sr	TC	(‰PD	(wt%	(d13C
Sample	CCD (m)	у	6Sr	error	(ppb)	(ppb)	(wt%)	B))	‰PDB)
C5704B_7Z4_1			0.7101							
4-19	8.64	listvenite	029	0.000023	10	7733	9.9	-1.72	NA	NA
C5704B_9Z3_2			0.7097							
3-28	13.67	listvenite	414	0.000022	61	17156	8.01	-1.12	NA	NA
C5704B_12Z1_			0.7105							
84-89	18.72	listvenite	864	0.000023	64	5844	9.32	-1.98	NA	NA
C5704B_14Z2_	24.06	1	0.7119	0.000000	1.4.4	(0.12	10.16	0.70	0.04	14.50
25-30	24.86	listvenite	908	0.000023	144	6042	10.16	-0./9	0.04	-14.58
C5/04B_14Z3_	25.02	listvonito	0./104	0.000010	1/2	6251	10.09	1 42	1 49	200
/1-/0	23.95	iistvenite	/9	0.000019	145	0551	10.08	-1.42	1.46	-2.00
$C_{3}/04B_{10}L_{5}$	28.9	listvenite	0.7107	0.000021	368	38660	9.42	-1.88	NA	NA
C5704B 1873	20.7	instvenite	0 7095	0.000021	500	50000	7.42	-1.00	INA	11A
25-31	35.34	listvenite	716	0.000023	319	8284	8.42	0.44	1.49	NA
C5704B 20Z1		notrenite	0.7113	01000020	517	0201	0112	0	,	1.1.1
78-83	40.01	listvenite	509	0.000022	152	48281	8.46	0.18	NA	NA
C5704B 23Z1			0.7112							
37-42	48.75	listvenite	99	0.000019	80	142571	10.63	-0.82	0.07	-19.07
C5704B 25Z2			0.7116							
55-60	52.45	listvenite	712	0.000020	78	186366	5.78	-0.21	0.02	-17.36
C5704B_26Z2_			0.7122							
77-82	56.11	listvenite	875	0.000019	682	22385	8.76	-0.23	NA	NA
C5704B_28Z1_			0.7106							
69-74	60.47	listvenite	974	0.000020	272	18688	8.86	0.02	0.09	-13.68
C5704B_30Z2_	<i>(</i> 1))		0.7105							
53-58	63.07	listvenite	413	0.000021	318	78656	9.82	-2.31	NA	NA
C5704B_31Z3_	(5.71	1	0.7119	0.000010	170	151222	10.2	2.20	0.22	6.07
31-3/ C5704D 2071	65./1	listvenite	56/	0.000019	1/8	151322	10.3	-2.20	0.22	-6.8/
C5/04B_52ZI_	66.97	listvonito	625	0.000020	296	85105	8 02	1.02	0.00	4.52
C5704B 3272	00.87	listvenite	0.7135	0.000020	380	65105	0.92	-1.03	0.00	-4.52
C3704B_32Z2_ 7_12	67 55	listvenite	589	0.000019	115	174106	10.61	-1.13	2 4 3	NA
C5704B 35Z1	07.55	nstvenite	0.7122	0.000017	115	1/4100	10.01	1.15	2.45	1471
6-11	71.54	listvenite	553	0.000022	98	10237	7.23	1.11	0.89	NA
C5704B 38Z3		serpentin	0.7108							
86-91	80.9	ite	931	0.000022	310	66812	5.33	-2.37	0.09	-10.07
C5704B_39Z1_		serpentin	0.7093							
25-30	82.18	ite	07	0.000022	48	9733	2.23	-5.35	0.10	-13.67
C5704B_39Z3_		serpentin	0.7104							
13-8	83.81	ite	894	0.000021	19	15271	1.96	-6.08	0.03	-22.04
C5704B_40Z3_		serpentin	0.7103							
3-8	86.98	ite	74	0.000019	6	89171	8.81	-4.91	0.06	-24.23
C5704B_42Z2_		serpentin	0.7118		10					
26-31	91.85	ıte .	308	0.000021	18	35313	1.71	-4.40	0.07	NA
C5704B_43Z2_	04.01	serpentin	0.7122	0.000010	50	011717	1.07	4.51	0.07	214
3-8 05704D 4472	94.96	ite	88	0.000019	59	211715	1.87	-4.51	0.07	NA
C5/04B_44Z2_	08 17	serpentin	0.7124	0.000010	21	110087	4.52	2 17	0.04	15.24
27-34	90.17	ne	092	0.000019	∠1	11090/	4.32	-3.17	0.04	-13.24

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C5704B_44Z3_	00.21	serpentin	0.7124	0.000000	27	02210	216	2.00	0.00	26.45
57-62 C5704B 44Z4	99.31	serpentin	888 0.7127	0.000023	37	83218	3.16	-3.08	0.06	-26.45
50-55	99.94	ite	018	0.000020	47	221049	8.11	-3.42	0.29	-4.90
C5704B_47Z3_ 18-23	107.88	listvenite	0.7134 127	0.000020	2612	25806	9.26	0.10	0.05	-23.97
C5704B_48Z2_	110.50	1	0.7130	0.000000	2.420	25120	214	274	0.02	25.40
C5704B 48Z3	110.52	listvenite	0.7131	0.000022	2429	35139	NA	NA	0.02	-25.40
90-95	111.5	listvenite	781	0.000023	33	46533	8.74	0.66	NA	NA
C5704B_50Z4_ 65-70	115.53	listvenite	0.7134 198	0.000023	8287	35574	6.55	0.32	NA	NA
C5704B_52Z1_	110.12	listuonito	0.7138	0.000010	11406	29542	Q 1	0.40	0.44	5 61
C5704B_52Z3_	119.15	listvenite	0.7145	0.000019	11400	36342	0.1	0.49	0.44	-5.04
0-5 C5704B 5273	119.67	listvenite	867	0.000022	13213	19572	9.72	-0.44	NA	NA
61-66	120.28	listvenite	184	0.000019	14220	19930	8.64	0.10	NA	NA
C5704B_54Z2_ 47-52	125.92	listvenite	0.7137 525	0.000022	16514	15830	9.15	0.36	NA	NA
C5704B_55Z1_	107.7(1	0.7139	0.000022	1(50)	10(74	0.27	1.00	0.07	
8-13 C5704B 55Z2	127.76	listvenite	0.7118	0.000022	16596	19674	8.37	1.20	0.07	NA
59-64	129.16	listvenite	074	0.000020	3417	39477	8.84	-0.37	0.11	-4.23
C5704B_55Z3_ 15-20	129.65	listvenite	0.7112 276	0.000022	9522	31563	7.87	0.16	NA	NA
C5704B_56Z4_	133 29	listvenite	0.7124	0.000022	11464	80713	911	1.52	NΔ	NA
C5704B_60Z1_	155.27	iistvenite	0.7123	0.000022	11404	00715	9.11	1.52	1111	1171
12-17 C5704B 60Z3	140	listvenite	404	0.000022	9275	11158	8.69	0.61	NA	NA
40-45	141.79	listvenite	358	0.000019	15152	28514	7.22	0.35	0.31	0.74
C5704B_60Z4_ 24-29H	142.43	listvenite	0.7144 926	0.000020	16037	19667	8.21	0.93	NA	NA
C5704B_64Z4_	154.05	listuonito	0.7133	0.000020	12775	22008	9 69	1.02	NA	NA
C5704B_65Z3_	134.93	listvenite	0.7122	0.000020	12775	23908	8.08	1.95	INA	INA
33-38 C5704B 6673	156.91	listvenite	888	0.000020	1079	10005	10.76	1.41	0.10	-16.35
66-71	160.62	listvenite	0.7120	0.000022	11964	37852	8.31	0.45	0.06	-24.15
C5704B_67Z2_ 26-31	162.03	listvenite	0.7138 291	0.000022	3747	21596	8.48	0.55	NA	NA
C5704B_67Z4_			0.7131							
39-44 C5704B 69Z2	163.67	listvenite	952 0.7117	0.000019	10001	20778	8.89	1.10	0.04	-10.87
55-63	168.38	listvenite	951	0.000023	2341	42165	8.98	0.13	NA	NA
0-8	175.63	listvenite	0.7125 344	0.000018	3745	40763	7.12	-0.13	0.05	-16.71
73-1-71.0- 75.0cm	180.01	listvenite	0.7123	0.000022	254	16269	9.1	0.63	0.24	-4 63
C5704B_73Z2_	100.01	nstvenite	0.7121	0.000022	234	10207	9.1	0.05	0.24	-4.05
0-5	180.3	listvenite	8	0.000020	292	30626	10.04	-1.53	0.21	-8.61
13.0cm	181.1	listvenite	166	0.000022	1021	182886	6.79	-1.29	0.02	-14.10
73-4-11.0- 16.0cm	181.85	serpentin ite	0.7105 479	0.000022	182	57427	1.62	-2.89	0.04	-16.58
C5704B_74Z1_	192.05	serpentin	0.7091	0.000022	2072	20007	1.45	4.00	0.05	27.04
56-64	182.95	serpentin	0.7097	0.000023	2073	39906	1.45	-4.90	0.05	-27.04
74-2-0.0-5.0cm	183.07	ite	785	0.000017	61	13067	1.88	-7.72	0.07	-10.96
47.0cm	184.24	ite	228	0.000019	294	21788	0.2	-10.62	0.04	-24.59
74-3-67.0- 73.0cm	184 49	serpentin	0.7100 467	0.000023	49	13717	1 77	-5 40	0.24	_9 94
75-1-12.0-	107.79		0.7112	0.000023		13/1/	1.//	0.70	0.27	7.77
16.0cm 75-1-70.0-	185.52	listvenite	295 0.7117	0.000022	134	49603	12.12	-0.43	0.10	-17.53
75.0cm	186.1	listvenite	597	0.000020	247	110258	9.7	-0.16	0.16	NA

75-2-27.0-			0.7115							
32.0cm	186.46	listvenite	092	0.000022	373	111620	8.61	-1.00	0.07	NA
C5704B_75Z2_			0.7116							
31-40	186.54	listvenite	533	0.000021	272	84029	9.54	-1.32	0.11	-11.87
75-2-76.0-			0.7123							
82.0cm	186.95	listvenite	277	0.000019	76	244293	10.73	-2.27	0.69	NA
C5704B_76Z2_			0.7124							
38-42	189.58	listvenite	782	0.000019	111	227322	11.43	-1.64	0.33	-12.42
C5704B_77Z3_			0.7124							
40-48	193.71	listvenite	411	0.000019	1696	178429	8.98	-2.12	0.26	-5.61
C5704B_77Z4_			0.7127							
43-48	194.57	listvenite	999	0.000020	1072	192155	8.66	-0.71	0.17	-9.16
C5704B_82Z1_		Metamor	0.7073							
50-55	201.18	phic	636	0.000020	70152	173001	0.02	NA	NA	NA
C5704B_88Z2_		Metamor	0.7071							
76-82	214.59	phic	713	0.000018	77502	182069	0.05	NA	NA	NA
C5704B_90Z3_		Metamor	0.7061							
8-13	220.79	phic	036	0.000019	36796	341585	0.03	NA	NA	NA
C5704B_92Z3_		Metamor	0.7064							
0-8	223.58	phic	436	0.000020	40839	266694	0.02	NA	NA	NA
C5704B_96Z2_		Metamor	0.7062							
0-8	232.1	phic	253	0.000019	11143	436010	NA	NA	0.09	-19.25
C5704B_99Z1_		Metamor								
0-5	240.33	phic	NA	NA	12063	378913	0.02	-0.42	NA	NA
C5704B_100Z2		Metamor	0.7060							
_13-21	244.47	phic	383	0.000018	9065	477887	0.25	NA	NA	NA
C5704B_105Z4		Metamor	0.7057							
_31-36	261.44	phic	39	0.000019	10901	558612	0.04	NA	NA	NA
C5704B_109Z4		Metamor	0.7052							
_24-32	270.5	phic	469	0.000020	17054	553129	0.11	NA	NA	NA
C5704B_116Z1		Metamor	0.7045							
_31-39	283.35	phic	642	0.000019	7182	392644	0.04	NA	NA	NA
C5704B_124Z1		Metamor	0.7053							
53-61	294.32	phic	928	0.000018	11092	527027	0.07	NA	NA	NA

Table 1. OmanDP Hole BT1B Strontium and Carbon concentrations and isotope rations

182 183

					Sr				С	
	Latitut	Longit	Elevatio	Distance from Basal	(ppm	Rb	87Sr/8	2SE total	(wt%	d13C (‰
Sample	e	ude	n (m)	thrust (m))	(ppm)	6Sr	error)	PDB)
OM20-	23.372	58.189			360.9		0.7093			
01	7	73	556	NA	2	3.6	033	0.000008	0.99	-0.61
OM20-	23.358	58.231					0.7116			
03	72	22	364	420	19.47	6.38	514	0.00001	0.26	-2.79
OM20-	23.373	58.516					0.7090			
05	26	63	468	470	25.23	5.8	506	0.000007	0.36	-1.64
OM20-	23.376	58.214			314.2		0.7134			
06	3	93	493	640	6	44.7	211	0.000007	0.04	NA
OM20-	23.376	58.215			382.4	103.9	0.7143			
07	28	08	488	640	6	6	305	0.000007	0.05	NA
OM20-	23.357	58.218					0.7126			
08	56	43	418	120	20.15	10.2	578	0.000007	0.09	-1.8
OM20-	23.357	58.218					0.7176			
09	44	29	432	100	32.54	56.28	305	0.000006	0.04	NA
OM20-	23.357	58.218					0.7175			
11	89	56	454	140	16.06	41.9	916	0.00001	0.11	-5.52
OM20-	23.386	58.145			213.3		0.7096			
12	09	61	262	780	9	4.94	678	0.000009	9.18	1.36
OM20-	23.397	58.182					0.7105			
13	92	25	278	150	74.29	8.52	193	0.000006	1.69	0.59
OM20-	23.407	58.154					0.7157			
14	35	96	269	700	9.9	17.3	845	0.000008	0.19	-3.19
OM20-	23.407	58.154					0.7090			
15	58	7	265	700	89.69	5.73	595	0.000009	4.93	1.3

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OM20-	23.393	58.178					0.7147			
16	77	62	302	560	34.61	27.25	076	0.000006	0.05	NA
OM20-	23.407	58.154					0.7241			
17	54	6	273	700	30.13	121.1	322	0.000009	0.06	NA
OM20-	23.357	58.218					0.7124			
10	86	54	458	140	27.97	29.92	03	0.000007	0.7	-3.64
OM20-	23.407	58.154			484.9		0.7082			
18	63	68	275	700	7	4.07	638	0.000009	11.06	1.26
OM20-	23.396	58.049			197.3		0.7090			
19	55	98	310	300	5	15.03	828	0.000007	1.28	0.93
OM20-	23.414	58.161			138.7		0.7085			
42	996	405	253	1200	1	0.32	112	0.00001	11.53	3.69
OM20-	23.362	58.234					0.7108			
04a	14	78	384	970	19.13	6.84	331	0.000009	0.7	-0.33
OM20-	23.362	58.234					0.7131			
04B	14	78	384	970	10.01	9.86	5	0.000008	0.15	-3.5

184 **Table 2.** Hawasina metasediments Strontium and Carbon concentrations and isotope rations

185 *3.1 Strontium Isotopes*

Measured ⁸⁷Sr/⁸⁶Sr values in Oman DP Hole BT1B lithologies show clear differences 186 187 between (a) the metamorphic sole and (b) the listvenites and serpentinites. The listvenites and 188 serpentinites vary from a minimum value of 0.709 in the upper serpentinite band to a maximum of 0.715 in some listvenites. ⁸⁷Sr/⁸⁶Sr values in the listvenites increase with depth from the 189 190 surface to 150 m while the serpentinite band between 80-100 m have lower values. Below 150 191 m, ⁸⁷Sr/⁸⁶Sr values are relatively constant with increasing depth, until the second serpentine band 192 at 181-186 m. Below the basal thrust, ⁸⁷Sr/⁸⁶Sr values are significantly lower in the metamorphic sole from 0.704 to 0.706, followed by lower ⁸⁷Sr/⁸⁶Sr values with greater depths. Although the 193 194 metamorphic sole contains meta-basalts and what appear to be volcanoclastic sediments 195 (Boudier et al., 1988; Kotowski et al., 2021; M. P. Searle & Malpas, 1980, 1982), all of the 196 samples have higher ⁸⁷Sr/⁸⁶Sr than typical mid-ocean ridge basalts (MORB) (Gale et al., 2013; 197 Hofmann, 2013). The Sr isotope values of the listvenite and serpentinite samples from BT1B are 198 similar to those of listvenites in nearby outcrops (Falk and Kelemen, 2015, Figure 2), which are 199 significantly more radiogenic than ⁸⁷Sr/⁸⁶Sr values of Oman peridotites (Gerbert-Gaillard, 2002; 200 Lanphere et al., 1981; McCulloch et al., 1981) and Cretaceous to modern seawater (McArthur et 201 al. 2001), but less radiogenic than some Hawasina and autochthonous sediments (Weyhenmeyer, 202 2000). Our samples of the Hawasina sediments, and those collected from the same region by



Hawasina samples are more radiogenic than any of the samples from Hole BT1B (Figure 2).

205



206

Figure 2. Measured (left) and age corrected at 96Ma (right) Sr isotope ratios and Sr concentrations for core samples from Oman DP Hole BT1B (large circles) compared to Sr isotope ratios in other lithologies measured in Oman. Hawasina sediments from this study and Falk and Kelemen (2015), leached peridotites from Gerbert-Gaillard (2002) and autochthonous sediment samples from Weyhenmeyer (2000). Rb concentrations were not reported for the autochthonous clastic metasediments, so an estimated upper bound Rb concentration of 200 ppm was used.

213

214 **3.2** Carbon concentrations and $\delta^{13}C$

215 **3.2.1** Total Carbon

The carbon contents of samples from Hole BT1B are highly dependent on lithology (Figure 3). All samples of ultramafic origin contain some carbon, mainly as Mg-rich carbonates, with serpentinites containing 0.2 to 5 wt% carbon, and listvenites containing 6 to 12 wt%. Samples from the metamorphic sole below 200 m depth contain 0.02 to 0.27 wt% carbon. In contrast, the Hawasina sediments have highly variable carbon contents. Some are almost pure metamorphosed limestones and dolomites with carbon contents of up to 11 wt%, while some have less than 0.05 wt% carbon.

 $\begin{array}{ll} 223 & \delta^{13}\text{C} \text{ in the upper 200m of Hole BT1B varies significantly between serpentinites and} \\ 224 & \text{listvenites (Figure 1). Serpentinites contain significantly lighter carbon than listvenites, as} \\ 225 & \text{observed in the depth profile (Figure 1). The lower serpentinite band contains two samples (74-2 \\ 226 & 0.0-5.0 \text{ cm and } 74-3 \ 42.0-47.0 \text{ cm}) \text{ that contain significantly lighter carbon than any serpentinite} \\ 227 & \text{in the upper band. In general, carbon concentrations correlate positively with } \delta^{13}\text{C} \text{ values in the} \\ 228 & \text{upper 200 m (Figure 3). Total carbon } \delta^{13}\text{C} \text{ values in the Hawasina nappes range from -5.52\% to} \\ \end{array}$

3.69‰, which almost encompasses the entire range of variation observed in Hole BT1B andappears to decrease with lower carbon contents.

3.2.2 Organic carbon

232 Graphitic carbon was identified in the core both at the drill site and during core 233 inspection aboard the D/V Chikyu (Kelemen et al, 2020, Kelemen et al. 2021). This organic component has a characteristic low δ^{13} C signature in listvenites and serpentinites from Hole 234 BT1B, which extends to a minimum of -27.04‰, representing 0.05wt% of total carbon in the 235 236 sample from 74-1 56.0-64.0cm (Figure 3). In some cases, the intermediate values likely represent 237 mixtures of organic and inorganic carbon, as some of the magnesite in the listvenites is very 238 resistant to 3N HCl attack and remain undissolved even after 5 days of leaching. The observed 239 light isotopic compositions are similar to those observed in other ultramafic localities such as 240 Cerro de Almirez and Liguria (Alt et al. 2013). These data confirm the presence of organic

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



243



246 4 Discussion

247 *4.1 Temperature and pressure of listvenite formation*

248 Falk and Kelemen (2015) estimated the temperature range of listvenite formation based 249 on conventional and clumped stable isotope thermometry (90±15°C), phase equilibria (80-250 130°C), and rock textures. New clumped isotope measurements on BT1B drill core samples by 251 Beinlich et al. (2020) widened the temperature range of listvenite formation and/or cooling from 252 $\sim 50\pm5$ °C to 250 ± 50 °C. This range of values suggests that the infiltrating reactive fluids had 253 variable temperatures, and/or clumped isotope values were reset during cooling, as proposed for 254 fine-grained samples of peridotite-hosted carbonate veins (Garcia del Real et al., 2016). The 255 pressure of listvenite formation is very poorly constrained due to the lack of pressure-sensitive 256 assemblages and the small size of fluid inclusions. A minimum pressure of 0.3 GPa is provided by the P-T conditions recorded by the carbonate platform during ophiolite obduction (Grobe et 257

al., 2019). For the upper limit, the listvenites must have formed at a pressure below the

- 259 maximum pressure reported for the metamorphic sole (~ 1.4 GPa) (Cowan et al., 2014; Searle 260 and Cox. 2002)
- and Cox, 2002).
- 261 *4.2 Timing of listvenite formation*

Falk and Kelemen (2015) used Rb/Sr and ⁸⁷Sr/⁸⁶Sr data on mineral separates to produce 262 an imprecise isochron age of 97±29Ma for a listvenite sample. This age is broadly consistent 263 264 with the \sim 96 Ma age of formation of igneous crust in the ophiolite, along with the same age of 265 metamorphism for the underlying metamorphic sole just beneath the basal thrust of the ophiolite (Hacker, 1994; Hacker et al., 1996; Rioux et al., 2013, 2016). Moreover, listvenites are found in 266 267 and near the basal thrust, from the UAE near the northwestern end of the ophiolite outcrop to the 268 area around Hole BT1B, near the southeastern end of the ophiolite outcrop in Oman (Nasir et al., 269 2007; Stanger, 1985). The extensive outcrop NE of Hole BT1B, known informally as MoD 270 Mountain, exposes the Banded Horizon, a peridotite unit found at the base of the ophiolite 271 mantle section composed of alternating 1- to 10-meter scale bands of dunite, harzburgite, and 272 minor lherzolite. This unit has distinct geochemical characteristics, with higher Al and middle 273 rare-earth-elements, compared to the residual mantle harzburgites that comprise most of the 274 mantle section of the ophiolite (Boudier et al., 1988; Khedr et al., 2014; Prigent et al., 2018; 275 Yoshikawa et al., 2015).

276 Tabular listvenites parallel to the dunite-harzburgite banding replace the Banded Unit, at 277 and above the basal thrust. In this region, there is a consistent "stratigraphy" with peridotite (\pm 278 listvenite) overlying the metamorphic sole, which in turn overlies the allochthonous Hawasina 279 nappe. This stratigraphy is nearly flat-lying, forming a broad anticline with a NW trending hinge 280 near the summit ridge of MoD Mountain, and a broad syncline that is coincident with the valley 281 north of MoD Mountain. Throughout this region, the contacts of listvenite bands within and at 282 the base of the Banded Unit are broadly parallel to banding in the peridotite, and to the contacts 283 between listvenite, peridotite, metamorphic sole, and the Hawasina nappe. These data are 284 consistent with our hypothesis that most of the listvenites formed by alteration of mantle 285 peridotite during the subduction of the underlying sediments via intra-oceanic thrusting and/or 286 later emplacement of the ophiolite onto the Arabian continental margin.

287 However, some of the listvenites formed or were modified during later events. The basal 288 thrust of the ophiolite was regionally reactivated – mainly with normal sense displacements – 289 during uplift and extension of the Jebel Akdar and Saih Hatat anticlinoria (Cawood et al., 1990; 290 Coffield, 1990; Hanna, 1990; Mattern & Scharf, 2018). Some or all of the cataclasites in BT1B 291 core and surrounding outcrops postdate listvenite formation and may record brittle deformation 292 associated with this reactivation (M.D. Menzel et al., 2020). In turn, the listvenite cataclasites are 293 fine-grained, indurated rocks – hard to drill or sample – that are cut by late, calcite and dolomite 294 rich veins. Thus, it is clear that local fluid-rock interaction of the listvenites continued after 295 brittle deformation.

On the basis of steep, fault-bounded contacts of listvenite with young, post-emplacement
conglomerates, several workers have inferred that the listvenites formed during later uplift and
extension (Nasir et al., 2007; Stanger, 1985; Wilde et al., 2002). Recently, Scharf et al. (2020)

reported 60 ± 16 Ma and 58 ± 6 Ma U/Pb ages for two carbonate veins that cut listvenite, and

300 structural observations indicating a top-down sense of shear along some faulted listvenite-

301 peridotite and listvenite-sole contacts. Following prior interpretations, they interpret these ages

302 as formation ages of the listvenites after ophiolite emplacement during uplift of the nearby Jebel

303 Akdar and Saih Hatat anticlinoria. We find that their interpretation is inconsistent with the field 304 observations reported above.

305 We have age-corrected our ⁸⁷Sr/⁸⁶Sr to the 96 Ma age reported by Falk and Kelemen (2015). This correction gives the lowest possible ⁸⁷Sr/⁸⁶Sr for all the samples based on reported 306 307 ages. For the corrections, we used Sr and Rb concentrations reported by Godard et al. 308 (submitted) for Hole BT1B and our analyses of Hawasina sediments. While Rb/Sr is low in most 309 BT1B samples and thus age corrections are small, this correction removes some of the apparent trends observed in measured ⁸⁷Sr/⁸⁶Sr versus depth. The age corrections particularly affect 310 311 listvenites with relatively abundant chromian mica (fuchsite-muscovite solid solutions, Falk & 312 Kelemen 2015, supplement) in the 115-163 m depth interval, as these micas host abundant Rb 313 (Godard et al. submitted). The age correction also affects the estimated ⁸⁷Sr/⁸⁶Sr values of the 314 metamorphic sole of BT1B and some of the Hawasina sediments as their Sr and Rb 315 concentrations are heterogeneous, ranging from 9 to 638 ppm for Sr and 0.4 to 97.7 ppm for Rb. Regardless, the age-corrected ⁸⁷Sr/⁸⁶Sr values of the listvenites are much higher than those of the 316 317 mantle and Cretaceous seawater. The only plausible source of this radiogenic Sr is from the

318 underlying Hawasina sediments.

319

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

320 ⁸⁷Sr/⁸⁶Sr and d¹³C data on MoD Mountain listvenites and Hole BT1B samples suggest 321 that replacement of peridotite by serpentinite and listvenite resulted from reaction with a single 322 fluid along a reaction path (Kelemen et al. 2021). The initial fluid was far from equilibrium with 323 peridotite, which converted olivine and serpentine in the protolith to carbonates + quartz and 324 approached equilibrium with serpentinite at higher extents of reaction progress and lower 325 integrated water/rock ratios (Beinlich et al 2020, Kelemen et al. 2021). Mg isotope data from a 326 set of samples studied by Falk and Kelemen (2015) show significant differences between dolomite and magnesite listvenites. Dolomite listvenites (average δ^{26} Mg ~ -1.33) are lighter in 327 Mg isotopes than magnesite listvenites (average δ^{26} Mg ~-0.33) (de Obeso et al., 2021), which 328 329 suggests magnesite dissolution and dolomite formation. This is consistent with the modelled 330 evolution of listvenites during fluid-rock reaction, with dolomite replacing magnesite at 331 increasing water/rock ratios (Kelemen et al. 2021).

As noted above, after the magnesite and dolomite listvenites formed they were cataclastically deformed, and then cut by late Ca-rich carbonate veins (Menzel et al. 2020). Thus, one might expect the veins to have formed from a later, geochemically distinctive fluid as suggested by variable clumped isotope (Δ 47) derived temperatures (Beinlich et al., 2020). This can be addressed in future studies via careful sampling of the post-cataclastic veins.

Returning attention to the source of the fluid that formed the bulk of the listvenites,
assuming that Sr and CO₂ were derived from the same fluid, Sr isotopes can be used to constrain
the source of the carbonating fluids that formed the serpentinites and listvenites. Falk and
Kelemen (2015) proposed three possible sources of fluids for carbonation: (1) compaction of
pore waters from underlying Hawasina sediments, (2) low temperature dehydration of opal and

342 clay minerals in calcite-bearing Hawasina sediments, and (3) higher-grade metamorphic

- devolatilization reactions involving subducted sediments similar to the Hawasina sediments
- 344 coupled with fluid that migrated up the subduction zone.

345 Unlike younger, mantle-peridotite-hosted carbonates, the listvenites do not contain a 346 significant fraction of seawater- or groundwater-derived Sr. The listvenites have ⁸⁷Sr/⁸⁶Sr ratios 347 that are distinct from recent, low-temperature carbonate veins and travertine in the mantle section 348 of the ophiolite (de Obeso & Kelemen, 2018; Peter B. Kelemen et al., 2011; Weyhenmeyer, 349 2000). Almost all of the listvenites and associated serpentinites from BT1B core and MoD Mountain outcrops have ⁸⁷Sr/⁸⁶Sr ratios at 96 Ma higher than Cretaceous seawater (Falk & 350 351 Kelemen 2015, and this paper). In contrast, young, peridotite-hosted carbonate veins and travertines in the Samail ophiolite consistently have ⁸⁷Sr/⁸⁶Sr lower than 0.709 and appear to 352 353 contain mixtures of Sr derived from seawater or groundwater and the mantle (de Obeso & 354 Kelemen, 2018; Gerbert-Gaillard, 2002; Peter B. Kelemen et al., 2011; Weyhenmeyer, 2000).

In addition, the listvenites do not contain significant Sr from the metamorphic sole sampled in Hole BT1B. Core samples of the sole have ⁸⁷Sr/⁸⁶Sr ratios that are similar to Indian Ocean MORB and near-ridge Pacific seamounts (Hofmann, 2013), which are systematically lower than the Sr isotope ratios of the listvenites. Perhaps the metabasalts are remnants of a subducted seamount, similar to accreted seamounts along the Cascadia margin of North America (e.g., Duncan, 1982), which derived from the enriched mantle source of some Indian Ocean MORB. Alternatively, the ⁸⁷Sr/⁸⁶Sr ratios may have increased during alteration.

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

369 Thermodynamic modeling of fluid-rock reactions (Kelemen et al. 2021) shows that the 370 characteristic listvenite mineral assemblages – magnesite + quartz – are attained from fluids with 371 $\sim 20,000$ ppm dissolved C for listvenite formation at 100-300 °C and 0.5 to 1 GPa, similar to the 372 assemblages modeled by Klein and Garrido (2011) at lower pressures. Such high dissolved 373 carbon contents are impossible to attain by congruent dissolution of pure calcite in aqueous 374 fluids at these P-T conditions (Peter B. Kelemen & Manning, 2015), which rules out silicate-375 poor limestones such as those from the continental margin as a carbon source. On the other hand, 376 metamorphic devolatilization of rocks composed of silicate-carbonate mixtures can produce C-377 rich fluids at temperatures above 400 °C and low to moderate pressures, depending on the rock 378 composition. Thus, we infer that the fluids that formed the listvenites derived from 379 devolatilization of subducting metasediments. During prograde subduction metamorphism, calc-380 silicate rocks, containing both clastic and carbonate components, undergo extensive 381 devolatilization at 2-3 GPa and 500 to 700°C (Gorman et al., 2006; Stewart & Ague, 2020) and

lose significant amounts of their CO_2 in this PT range due to reactions similar to the simplified reaction:

384 $CaMg(CO_3)_2$ (dolomite) + $CaMgSi_2O_6$ (diopside) + 2 $CaAl_2Si_2O_7$ (OH)₂·H₂O (lawsonite)

$$= 2 \operatorname{Ca_2MgAl_2Si_3O_{12}}(\text{garnet}) + 4 \operatorname{H_2O} + 2 \operatorname{CO_2}$$

In contrast, carbonate-rich compositions (limestone, dolomite, marble) are predicted to
 retain most of their CO₂ during subduction (e.g, (Kerrick & Connolly, 2001; Stewart & Ague,
 2020)).

At temperatures greater than ~ 300°C, dissolved CO₂ in aqueous fluids have δ^{13} C values 389 390 that are higher than those of co-existing calcite and dolomite (Chacko et al., 1991; Deines, 2004; 391 Horita, 2014). Thus, for example, fluid in equilibrium with calcite with δ^{13} C between -4‰ to -392 6‰ (as in Hawasina clastic sediments) would contain dissolved CO₂ with δ^{13} C of -2.3‰ to -393 0.3‰ at 500°C. At lower temperatures, like those estimated for listvenite formation, calcite and 394 dolomite have δ^{13} C higher than co-existing fluids. Thus, dolomite and presumably magnesite in 395 equilibrium with fluids with δ^{13} C of -2.3% to -0.3% would have δ^{13} C in the range of -0.9% to 1.1‰, similar to the δ^{13} C observed in the listvenites from Hole BT1B and the surrounding 396 397 outcrops.

398 Figure 4a illustrates that the listvenites and serpentinites from Hole BT1B lie along the 399 reaction path proposed by Kelemen et al. (2021) at 200°C and 0.5GPa that forms listvenites with 400 W/R~100. In our calculations, we assume that reacting fluids enriched in Sr (250 ppm) with 401 87 Sr/ 86 Sr values @96Ma that are similar to those of clastic Hawasina metasediments (0.7110 to 0.7135) reacted with the peridotite with mantle-like ⁸⁷Sr/⁸⁶Sr (0.7027±0.0011). The listvenites 402 403 and serpentinites follow a mixing trend (Figure 4b) between mantle peridotite like compositions 404 $(Sr=1.5 \text{ ppm}, {}^{87}Sr/{}^{86}Sr=0.7027\pm0.0011, C=680\text{ ppm} \text{ and } \delta^{13}C=6.0\pm2.0\%)$ and low temperature 405 carbonates that crystallized from a fluid produced by high-pressure, high-temperature 406 devolatilization with Sr isotope ratios in the range of the Hawasina clastic sediments (Sr=250ppm, ⁸⁷Sr/⁸⁶Sr~0.7100 to 0.7135) and fractionated carbon isotopes as described above. 407 408 These trends together suggest that the CO₂-bearing aqueous fluids that formed the listvenites 409 from Hole BT1B and surrounding outcrops were derived by devolatilization of calc-silicate metasediments, with 87 Sr/ 86 Sr and δ^{13} C similar to clastic sediments in the Hawasina Formation 410

411 along a subduction zone geotherm. These fluids migrated up dip to lower pressures and

412 temperatures to form the listvenites.

413



Figure 4. (a. left) Total carbon (wt%) vs ⁸⁷Sr/⁸⁶Sr @96Ma. Black lines are reaction paths of 415 carbon rich fluid reacting with peridotite at 200°C and 0.5GPa at variable water/rock (tie lines) 416 from Kelemen et al. (submitted). The reacting fluid is assumed to have ⁸⁷Sr/⁸⁶Sr values of 0.7100 417 to 0.7135 like those of clastic Hawasina metasediments. (b. right) Inorganic carbon δ^{13} C vs 418 419 ⁸⁷Sr/⁸⁶Sr @96Ma. Black lines are mixing lines between mantle and carbonate minerals 420 precipitated from a metamorphic fluid. The fluid has the same carbon concentration as the reaction path on the figure 4a and the δ^{13} C fractionation between the carbonate and the fluid are 421 described in the body of the text with values for high and low PT shown top. We assume that the 422 423 fluid has the Sr isotope ratios of Hawasina clastic sediments. ⁸⁷Sr/⁸⁶Sr values for the depleted mantle are for mid-ocean-ridge-basalt (MORB) from (Hofmann, 2013). Mantle δ^{13} C is from 424 425 (Deines, 2002).

426 **5 Conclusions**

427 Listvenites and spatially associated serpentinites from Hole BT1B and surrounding outcrops that 428 replace residual mantle peridotites from the base of the Samail ophiolite have Sr isotope ratios 429 that are more radiogenic than their peridotite protoliths, Cretaceous seawater, modern seawater, 430 groundwater in the ophiolite, and the underlying metamorphic sole. We suggest that the 431 radiogenic Sr isotope component was transported via carbon-rich aqueous fluid that reacted with the peridotite to form the listvenites and serpentinites. The ⁸⁷Sr/⁸⁶Sr values of this component 432 433 resemble those of calcite-bearing clastic sediments in the Hawasina Formation underlying the 434 ophiolite and the metamorphic sole. However, the fluid must have contained higher dissolved 435 carbon contents than feasible for congruent dissolution of pure calcite at < 2 GPa and/or < 436 550°C. Thus, we hypothesize that this fluid was derived by devolatilization of carbonate- and 437 silicate bearing meta-sediments akin to the Hawasina clastic sediments at 0.5 to 2.3 GPa and 400 438 to 700°C in the subduction zone (M. P. Searle et al., 1994). This fluid then migrated up dip to

- 439 react with hanging wall peridotite at <1 GPa and <250°C, forming the listvenites and
- 440 serpentinites. Carbon isotope fractionation during high temperature devolatilization followed by
- 441 low temperature carbonate precipitation during the reaction with peridotite likely controlled the
- 442 isotopic characteristics of the listvenites and the serpentinites with δ^{13} C from -10.6‰ to 1.92‰ 443 and 87 Sr/ 86 Sr from 0.7090 to 0.7145.

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- 462 All data will be uploaded to EarthChem or a similar repository prior to final publication.
- 463
- 464 **7 References**
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