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1 Hydrogen Concentrations and He Isotopes in Olivine From

2 Ultramafic Lamprophyres Provide New Constraints on a

3 Wet Tarim Plume and Earth's Deep Water Cycle

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- 20 Key Points:
- Olivines in the Wajilitag aillikites show high water contents (75-168 ppm H₂O).

• Hydrogen loss in olivines during decompression was limited to the outermost

- rims, and yields dehydration durations of 15-417 min.
- H_2O in these rocks derived from a plume source slightly contaminated by
- 25 recycled hydrous crustal material.

26 Abstract

27 Water enters Earth's mantle via subduction of hydrated oceanic slab and largely 28 returns to the exosphere through arc volcanism. However, the extent to which H_2O is 29 transferred into the deep mantle is poorly constrained. Here we address this question 30 by combining mineral chemistry and bulk-rock geochemistry data for aillikites related 31 to the deep mantle plume which generated the Permian Tarim large igneous province 32 (NW China). The water contents of olivine phenocrysts are 75-168 ppm H_2O and 33 positively correlated with Ti contents. These results, combined with infrared hydroxyl peaks at 3572 and 3525 cm⁻¹, suggest that H is mainly present in the form of Ti-34 35 clinohumite-like point defects. Hydrogen concentration profiles across olivine reveal 36 that H loss during decompression was limited to the outermost rims, and yield 37 dehydration durations of 15-417 min. Based on the water contents of the highest-Fo 38 olivine cores, the water contents of the primitive aillikite melts and their mantle source are estimated as 1.6-4.7 wt.% and 150-1200 ppm H₂O, respectively. ${}^{3}\text{He}/{}^{4}\text{He}$ 39 40 ratios (5.31-5.84 Ra) of olivine phenocrysts are slightly lower than MORBs and suggest involvement of recycled slab containing U (and hence radiogenic ⁴He) in the 41 42 plume source. This interpretation is consistent with Pb isotope compositions of the 43 aillikites which are intermediate between PREMA (Prevalent Mantle) and EM 44 (Enriched Mantle) compositions. These lines of evidence combined with the depleted 45 Sr-Nd isotopes and moderately radiogenic Os isotopes of the aillikites suggest that water in these rocks derived from a plume source marginally contaminated by deeply 46 47 subducted hydrated material.

48

49

50 Plain Language Summary

51 Water enters Earth's mantle via subduction of water-bearing oceanic crust and largely returns to Earth's surface through arc volcanism above subduction zones. 52 53 However, how much surface-derived water travels to the deep Earth locked in 54 subducted material is poorly constrained. Here we address this question by combining 55 mineral and bulk-rock geochemical data for carbonate- and water-rich volcanic rocks genetically linked to the Tarim mantle plume (convective upwelling of thermally 56 57 anomalous mantle). Olivines in these rocks contain high water contents that are 58 positively correlated with Ti due to a coupled mechanism of incorporation in the 59 olivine structure. Water content profiles across olivine reveal that water loss from 60 olivine during magma ascent was limited to the outermost rims. The calculated water 61 contents of the mantle source of these rocks based on water concentrations in the 62 olivine cores are higher than typical H₂O contents in the upper mantle. In combination 63 with bulk-rock Pb-Sr-Nd-Os isotope data of these volcanic rocks and new He isotope compositions for these olivines, these data suggest that the deep mantle source of 64 65 these rocks was polluted by subducted water-bearing oceanic crust. The elevated 66 water contents in mantle source help explain the occurrence of voluminous 67 magmatism of large igneous province.

68 **1. Introduction**

69 Water is the dominant volatile component in the Earth and plays a crucial role in 70 controlling surface climate, the emergence and evolution of life as well as rheology 71 and partial melting processes in the Earth's interior (e.g., Dixon et al., 2002; 72 Hirschmann, 2006, 2018; Kurokawa et al., 2018; Marty, 2012). It is well established 73 that water is continuously transferred into the mantle through subduction of hydrous 74 minerals, but largely returns to the ocean-atmosphere system through arc volcanism 75 (e.g., Bodnar et al., 2013; Cerpa et al., 2022). However, the proportion of water in the 76 slab that can ultimately be transferred to the deep mantle and perhaps contribute to 77 subsequent wet plume formation and flood basalt magmatism is poorly constrained 78 (e.g., Dixon et al., 2002; Hwang et al., 2017). One hypothesis entails that a large 79 fraction of subducted water can be carried into deep mantle (Hu et al., 2017; Hwang 80 et al., 2017), whereas other authors favor a scenario whereby water is efficiently 81 released to mantle wedge, crust and ocean-atmosphere system during subduction, 82 leading to low time-integrated H₂O contents in the deeply subducted slab (Cerpa et 83 al., 2022; Dixon et al., 2002). Combining He isotope with H_2O measurements has 84 been proven valuable for tracking the source of water in volcanic rocks including 85 inputs of crustal volatiles into the mantle (Marty, 2012). Helium isotopes are indeed 86 powerful tracers of primordial contributions to mantle-derived magmas, but can also 87 pinpoint the occurrence of (U-bearing) subducted material in the mantle (e.g., Hanyu 88 & Kaneoka, 1997; Jackson et al., 2020; Kurz et al., 1982) especially when combined 89 with other geochemical tracers such as Sr, Nd, Pb and Os isotopes.

90	Alkaline ultramafic magmas such as ultramafic lamprophyres (UMLs),
91	kimberlites and lamproites are volatile-rich (e.g., H ₂ O, CO ₂) and can provide valuable
92	information on the nature of fusible components in the mantle (>100 km) beneath
93	stable continents (Giuliani et al., 2017; Giuliani & Pearson, 2019; Rock et al., 1986;
94	Tappe et al., 2006). Olivine is the first phase to crystallize and, therefore, provides a
95	unique tool for deciphering the primitive composition of these magmas and the nature
96	of their mantle source (Giuliani et al., 2020; Nosova et al., 2018; Sokol et al., 2013;
97	Veter et al., 2017). Despite being a nominally anhydrous mineral (NAM), olivine can
98	incorporate trace amounts of hydrogen (H, bonded as hydroxyl) in crystal defects
99	(ionic vacancies and interstitials; e.g., Berry et al., 2005; Demouchy & Alard, 2021;
100	Mosenfelder et al., 2006; Newcombe et al., 2020; Tollan et al., 2018), and can act as a
101	potential important reservoir for H in the mantle (Peslier, 2010; Ohtani, 2021).
102	Alkaline ultramafic magmas generally ascend from the deep mantle very rapidly (10-
103	30 m/s) as indicated by the presence of diamonds (Kelley & Wartho, 2000; Sparks et
104	al., 2006) and thus can preserve the original water contents in the core regions of
105	olivine phenocrysts (Matveev & Stachel, 2007). For example, kimberlitic olivine
106	shows the highest water contents in natural olivine with up to 600 ppm $\mathrm{H_2O}$ (Bell et
107	al., 2004; Hilchie et al., 2014; Matsyuk & Langer, 2004; Matveev & Stachel, 2007).
108	The occurrence of fresh, olivine-rich aillikites (or carbonate-rich ultramafic
109	lamprophyres) in the northwest Tarim large igneous province (LIP) (Fig. 1a) provides
110	an ideal opportunity to examine Earth's deep water cycle because these magmas are
111	probably genetically linked to the Tarim plume (Wang et al., 2021a; Zhang et al.,

112 2013b). Based on detailed bulk-rock major- and trace-element and Sr-Nd-Os isotope, 113 olivine major- and trace-element and oxygen isotope, and bulk-carbonate C-O isotope 114 analyses, it was previously suggested that these rocks originated from a moderately 115 depleted sub-lithospheric (plume) source which was marginally contaminated by 116 deeply subducted oceanic crustal components based on radiogenic Os isotopes (Wang 117 et al., 2021a, 2022). In this work, we combine new H_2O content and He isotope data 118 for olivine with new and existing olivine major- and trace-element composition and 119 bulk-rock Sr-Nd-Os isotope results (Wang et al., 2021a, b, 2022), including newly 120 acquired bulk-rock Pb isotope data, to constrain the source of water in these rocks. 121 The water contents and distribution in the olivines were determined by secondary ion 122 mass spectroscopy (SIMS) and Fourier transform infrared spectrometry (FTIR). 123 Noble-gas analyses were undertaken by in-vacuo crushing of bulk olivine separates. 124 The goals of this study are (1) to quantify the H_2O contents of primitive aillikite 125 magmas and their mantle source; and (2) thereby to estimate the source and role of 126 water in the generation of the Tarim LIP.

127 **2. Geological Background and Petrography**

The Tarim craton in northwestern China occupies an area of 600,000 km² (Fig. 1a) and is composed of Archean and Neoproterozoic crystalline basement overlain by Phanerozoic sedimentary strata (Tian et al., 2010; Zhang et al., 2013a). Permian magmatic activity identified in this region produced more than 260,000 km² continental flood basalts (Fig. 1a; Tian et al., 2010). In addition to the erupted flood basalts and rhyolites as well as minor nephelinites, the Tarim LIP also contains a diverse assemblage of mafic-ultramafic intrusive rocks including aillikites, nepheline
syenites, lamprophyres, carbonatites and mafic dyke swarms (Fig. 1b; e.g., Cheng et
al., 2018; Tian et al., 2010). The Permian magmatism can be divided into two major
pulses according to geochronological studies: (1) flood basalts and felsic volcanic
rocks at ~290 Ma (Jiang et al., 2004; Tian et al., 2010; Zhou et al., 2009); and (2)
mafic-ultramafic intrusions and dyke swarms at ~280-270 Ma (e.g., Zhang et al.,
2010; Zhang et al., 2013b).

141 Wajilitag is situated in the northwestern portion of the Tarim LIP (Fig. 1b). 142 Permian magmatic rocks in this region consist of diverse coeval intrusions and dyke 143 swarms of variable composition, i.e., aillikites, gabbros, pyroxenites, aegirine 144 nepheline syenites and alkaline lamprophyres-tephrites. Based on geological 145 observations and geochronological data, the formation sequence of these magmas is 146 inferred to be as follow: mafic-ultramafic intrusions (gabbros and pyroxenites; ~284-147 281 Ma) \rightarrow aillikites (284-268 Ma) \rightarrow carbonatites and nepheline symptomics (~273 Ma) 148 \rightarrow alkaline lamprophyres-tephrites and nephelinites (~268 Ma) (Wang et al., 2021a 149 and references therein). The Wajilitag aillikites are the first diamond-bearing rocks 150 identified in NW China (Fig. 1b; Jiang et al., 2004; Zhang et al., 2013b). The NWW-151 SEE trending dykes intruded upper Devonian strata, and were, in turn, crosscut by 152 tephrite and alkaline lamprophyre dykes. The aillikite pipes exhibit elliptical to 153 rounded shape with exposures at surface up to 180 m \times 100 m, but their extension at 154 depths is not known. The dimensions of the dykes range between 0.5-2 m in width 155 and 5-7 m in length, but their actual length should be longer due to ubiquitous desert 156 cover.

157 These aillikites display a distinctive inequigranular porphyritic texture with 158 abundant phenocrysts (30-40%) and nodules (polygranular aggregates, 5-10%) set in 159 a fine-grained matrix (Fig. 2a-c). The ferromagnesian silicate phenocrysts include 160 olivine, clinopyroxene and hydrous minerals (i.e., amphibole and phlogopite). Olivine 161 can be divided into two textural groups: fine-grained microcrysts (<5 mm) and 162 coarse-grained macrocrysts (>5-25 mm). The former are common in all aillikite pipes 163 and dykes (e.g., Dyke 1, 2 and 3), while the latter only occur in the most evolved 164 Dyke 3 aillikites (Fig. 2d-f vs Fig. 2f). As shown in Fig. 2i, primary melt inclusions 165 generally present in abundance in the olivine microcrysts. The matrix consists of 166 phlogopite, apatite, perovskite, titanomagnetite and calcite. Quantitative mineral mode 167 abundances for a representative aillikite sample are present in Fig. S1 in Supporting 168 Information S1. Herein, we mainly focus on olivines separated from three fresh 169 aillikite dykes previously named Dyke 1, 2 and 3 (Fig. 1b). Detailed petrographic 170 descriptions of these dykes can be found in Wang et al. (2021a).

171 **3. Analytical Methods**

172 **3.1. Major and Trace Element Analyses**

Petrographically fresh aillikite samples collected from three Wajilitag aillikite dykes (Dyke 1, 2 and 3; Fig. 1b) were initially crushed to millimeter sized fragments using a jaw crusher and then fragments of the mafic-ultramafic nodules and crustal xenoliths in the aillikites were carefully removed using a stereoscopic microscope. Then, the freshest olivine crystals were handpicked under a binocular microscope 178 either mounted in epoxy resin for H_2O , major-, trace-element determinations or 179 treated separately for bulk-mineral He isotope analyses.

The major and minor element abundances of olivine were analyzed using an
EMPA1600 electron probe micro analyzer at the Chinese University of Geosciences,
Beijing. Analytical conditions were 20 nA probe current, 15 kV accelerating voltage,
5 μm beam diameter, and peak counting times of 20 s. The results were calibrated by
employing synthetic oxides and natural minerals, and matrix corrections were
undertaken using a ZAF program. The analytical results are present in Table S1.

186 The minor and trace elements in olivine selected for major element and water content analyses were measured by LA-ICP-MS employing an Element XR HR-ICP-187 188 MS instrument (Thermo-scientific) coupled to a 193 nm ArF excimer laser-ablation 189 system at the National Research Centre for Geoanalysis, Beijing and the Institute of 190 Geology and Geophysics, Chinese Academy of Sciences. Ablation was undertaken in 191 a He atmosphere and Ar was used as the carrier gas (1.5 l/min). Every analysis was performed using a laser spot size of 50-100 μ m at 6 Hz with an energy of ~4 J/cm² for 192 40 s preceded by 20 s of gas blank measurement. ²⁹Si was used as the internal 193 standard. Three glass standards (GSE-1G, NIST SRM 610, NIST SRM 612; Jochum 194 195 et al., 2011) and one olivine standard (XEN; Batanova et al., 2015; Wu et al., 2020) 196 were analyzed as external references after every eight unknowns. Details of analytical 197 procedures can be found in Wu et al. (2018). The minor and trace element analytical 198 results are given in Table S2. The analytical precision and standard deviation are 199 listed in Table S3.

200 **3.2.** Secondary Ion Mass Spectrometry (SIMS)

Olivine water contents were determined by a CAMECA IMS 1280-HR 201 202 multicollector ion microprobe at the Guangzhou Institute of Geochemistry, Chinese 203 Academy of Sciences, following the procedures described by Zhang et al. (2019). 204 Prior to the analysis, the machine (including the sample chamber without the sample 205 mounts) was baked for 24 hours. The vacuum in the sample chamber was enhanced 206 by keeping an alloy sample mount (52% Sn + 48% Bi) in the high-vacuum storage chamber overnight and then further vacuumed to 5 \times 10⁻⁹ mbar. Moreover, an 207 208 automatic liquid nitrogen cooling technique was applied to cool the sample chamber 209 in order to improve the vacuum performance. Analyses were conducted using a 210 primary Cs⁺ beam (4-5 nA) accelerated at 10 kV. Each spot was pre-sputtered for 200 211 seconds using a 30 \times 30 μ m raster beam and then the raster size was reduced to 20 \times 212 20 µm using a narrow field aperture to minimize the water background signals. A 213 normal incidence electron gun was applied to compensate for sample charging, and a 214 nuclear magnetic resonance (NMR) controller was employed to stabilize the magnetic 215 field during the measurement campaign. Other analytical conditions include a 400-216 contrast aperture, ~ 60 entrance slit, and 50 eV energy slit with a 5 eV energy window. 217 Although we focused on olivine H concentration quantification, the oxygen isotopic values were also analyzed simultaneously. The ¹⁶O⁻, ¹⁸O⁻, and ¹⁶O¹H⁻ were detected 218 219 by two Faraday cup detectors and an electron multiplier of monocollector, respectively. Collector slits of 500 mm were employed to collect ${}^{16}O^{-}$ and ${}^{18}O^{-}$, which 220 221 yielded a mass resolving power (MRP) of ~2500. A collector slit of ~173 µm was

applied for ${}^{16}O^{1}H^{-}$ to minimize the isobaric interference of ${}^{17}O^{-}$, which corresponds to 222 a ~7000 MRP. The olivine water contents were calculated from measured ${}^{16}O^{1}H^{-/16}O^{-}$ 223 ratios based on the calibration curve constructed by FTIR H₂O contents and SIMS 224 ¹⁶O¹H^{-/16}O⁻ ratios for four olivine reference materials (San Carlos, KLB-1, ICH-30, 225 Mongok; following Zhang et al., 2019). The calibration curve ($[H_2O] = 5.5 \times 10^6 \text{ x}$ -226 9.12: $R^2 = 0.99$. Fig. 3) was used here to invert the measured $x = {}^{16}O^1H^{-16}O^-$ ratios 227 into H₂O contents. The analytical accuracy and precision of water content 228 229 measurement were generally better than 10 % in this study.

230 **3.3.** Fourier-Transform Infrared Spectroscopy (FTIR)

231 The transmission FTIR spectroscopy analyses were carried out to determine the 232 speciation of water and structural position of hydroxyl groups in olivine at the 233 Institute of Geology and Geophysics, Chinese Academy of Sciences, Beijing. 234 Unpolarized and polarized infrared spectra were obtained using a Bruker Vertex 70V 235 coupled with a mercury cadmium telluride (MCT) detector cooled with liquid 236 nitrogen and a Bruker HYPERION 2000 IR microscope. Unpolarized mid-infrared radiation between 650-4000 cm⁻¹ was produced by a Ge-KBr beam splitter and a 237 Globar light source. The polarized IR radiation was generated using a KRS-5 238 239 polarizer (thallium bromideiodide) placed between the sample and the mid-IR source. 240 The stage was flushed with nitrogen gas during the analysis to limit impact from 241 ambient atmospheric moisture and a background determination was carried out prior 242 to each analysis. Optically clean areas of olivine in doubly polished section (100-150 243 μ m in thickness) were measured with a square aperture of sizes between 50 and 100

 μ m. For each measurement more than 128 scans were accumulated at 4 cm⁻¹ 244 245 resolution. Cross-sections with the di-electric vector (E) of the incident infrared light 246 parallel to the principal optical axes $(E//\alpha, E//\beta, \text{ or } E//\gamma)$ of olivine were acquired for 247 olivine grains in three rock samples. The crystallographic orientations of the olivine 248 were obtained from electron backscatter diffraction (EBSD, see Text 1 in Supporting 249 Information S1) analyses prior to FTIR measurements. The integrated absorbance (A) 250 for the olivine, corresponding to the integrated areas beneath the O-H bands, was 251 calculated using the Origin pro8 software. It should be noted that as water contents in 252 the Wajilitag olivine have been quantified using SIMS analysis, the unpolarized and 253 polarized FTIR spectroscopy mainly focused on examining hydroxyl speciation and 254 relative differences of hydrogen concentration (or integrated hydroxyl absorbance) in 255 olivine, respectively.

256

3.4. Helium Isotope Measurements

257 Helium isotope analyses were performed at the Institute of Mineral Resources, 258 Chinese Academy of Geological Sciences. Before analysis, mineral grains were 259 cleaned ultrasonically in deionized water and washed in acetone for 30 min, and 260 finally dried. Mineral separates (>500 mg each) were loaded into a crusher, baked 261 online at 150 °C for 48 h, and then pumped for 7-14 days until blanks were low and 262 stable. The gases released by crushing were purified using a titanium sponge pump at 263 780 °C and Zr-Al pump at 300 °C for 20 min to eliminate active gases including water 264 and CO_2 . The remaining gases were exposed to a second cold Zr-Al pump for 10 min. 265 Comparatively pure He was introduced into the analytical system and further purified

266 using a liquid nitrogen cooled titanium sublimation pump to remove other trace gases 267 such as H_2 . The noble gases isotope compositions were analyzed using a Helix SFT noble gas mass spectrometer. ⁴He was analyzed by a Faraday cup and ³He by an 268 269 electric multiplier. The mass resolution of >700 can separate ³He from the HD + H₃ 270 doublet peak. Air standard was analyzed during each analytical session. The hot blank levels of He at 1600 °C were ${}^{4}\text{He} = 2 \times 10^{-11} \text{cm}^{3} \text{ STP/g}$ (standard temperature and 271 272 pressure). The contribution of the blank to the analyzed samples was negligible. The 273 analytical results and standard deviation are listed in Table S4.

274 **3.5. Bulk-sample Pb Isotopes**

275 The Pb isotope compositions of nine representative aillikite samples were 276 measured by a GV Isoprobe-T thermal ionization mass spectrometer monitored by 277 standard NBS 981 at the Beijing Research Institute of Uranium Geology. About 100-278 200 mg of rock powder (<200 meshes) was completely dissolved in PFA beakers 279 using a mixture of $HF + HNO_3 + HClO_4$ for 24 h. Pb was separated and purified using 280 anion exchange resin AG1-X8 with diluted HBr as an eluant. The purified Pb was 281 loaded onto rhenium filaments using a phosphoric acid-silica gel emitter. Total 282 procedure blanks were <100 pg for Pb. During the measurement campaign, repeated analyses of the NBS 981 standard yielded values of $^{206}Pb/^{204}Pb = 16.937 \pm 0.002$, 283 207 Pb/ 204 Pb = 15.457 ± 0.002, and 208 Pb/ 204 Pb = 36.611 ± 0.004, in agreement with the 284 285 reference values (e.g., Belshaw et al., 1998). The Pb isotope analytic errors were 286 reported as $\pm 2\sigma$.

287 **4. Results**

288 4.1. Olivine Major and Trace Element Concentrations

289	The major and minor element compositions of olivine grains which have been
290	selected for water content analyses are present in Figs. 4-6 and reported in Table S1.
291	The forsterite contents (Fo = $100 \times Mg/(Mg + Fe)$ a.p.f.u.) of the olivine in the
292	aillikites range from 78 to 88. Similar to previous studies (Wang et al., 2021a, b), the
293	olivine can be distinguished between high-Ni (>0.10 wt.% NiO) and low-Ni (<0.10
294	wt.% NiO) groups (Fig. 4) based on major and minor element compositions. The
295	high-Ni olivine group corresponds to fine-grained microcrysts (<5 mm) identified
296	petrographically in Dyke 1, 2 and 3 (Fig. 2d-f), while the low-Ni olivine group
297	includes large macrocrysts (5-25 mm; Fig. 2f) that only occur in Dyke 3. The high-Ni
298	olivines have higher Fo78-88 and NiO (0.10-0.47 wt.%) contents but lower MnO (0.10-
299	0.47 wt.%) contents than the low-Ni ones (Fo = 78-81; NiO <0.10 wt.%; MnO = 0.40-
300	0.74 wt.%). Their CaO concentrations overlap (0.06-0.18 wt.% and 0.09-0.14 wt.%,
301	respectively). These compositions are entirely consistent with those reported by Wang
302	et al. (2021a, b; Fig. 4). X-ray element maps show that the SiO ₂ , FeO and MgO
303	contents of the olivine microcrysts are almost homogenous with only slight variation
304	along the edges (Fig. S2 in Supporting Information S1).
305	The trace element compositions of olivine grains are quantified by LA-ICP-MS
306	(Table S2). Concentrations of Ni, Cr, Ti and Al are higher in the high-Ni olivine group
307	compared to the low-Ni one, i.e., Ni = 1149-4164 vs. 23.4-259 ppm, Cr = 16.3-106 vs.

308 3.6-11.9 ppm, Ti = 26.7-117 vs. 44.7-82.9 ppm and Al = <22.9 vs. <2 ppm. In

309 contrast, Mn, Zn and Li contents are enriched in the latter: Mn = 1319-3133 ppm vs.

310 3835-5588 ppm; Zn = 109-231 vs. 277-349 ppm; Li = 1.8-4.8 vs. 3.6-7.3 ppm. Other
311 elements show little difference between the two olivine groups, including Ca (579312 1101 vs. 559-871 ppm), P (88.9-178 vs. 91.4-218 ppm), and Sc (2.4-4.6 vs. 1.4-9.1
313 ppm). These trace element compositions are consistent with those reported by Wang
314 et al. (2021a, b; Fig. S3 in Supporting Information S1). A LA-ICP-MS transect across
315 a representative high-Ni olivine microcryst shows that Ni contents decrease and Mn
316 and Ca contents increase from core to rim (Fig. 6).

317 4.2. Content and Distribution of Hydrogen in Olivine

318 Water contents in the Wajilitag olivine grains measured by SIMS vary from 75 to 319 168 ppm H₂O (Table S1; Fig. 5). To avoid potential inclusions of hydrous phases such 320 as serpentine or clinohumite, we used the small beam ($\leq 30 \mu m$) and shallow depth of 321 penetration (<2 µm) of SIMS analysis, as well as monitored the H/Si and O/Si count 322 to notice potential contamination by silicate rich (and hydrous) components. For the 323 high-Ni olivine microcrysts, the H₂O contents increase gradually with decreasing Fo 324 values, and increasing Mn, Ca and Ti contents (Fig. 5). Based on their water contents, 325 the microcrysts can be further divided into high-H₂O (144-168 ppm H₂O) and low-326 H_2O (75-146 ppm H_2O) subgroups, with the latter showing higher Fo, Ni, Cr and 327 lower Ca and Ti concentrations, i.e., more primitive composition. The lower-H₂O, 328 higher-Fo microcrysts predominantly occur in the less evolved Dyke 1 and 2 329 aillikites, while the higher-H₂O, lower-Fo microcrysts are mainly present in evolved 330 Dyke 3 (Table S1). The H₂O contents of the low-Ni macrocrysts are 103-161 ppm 331 H₂O, and do not show any correlation with Fo or minor elements, except for a 332 positive correlation with Ti concentrations (Fig. 5).

To assess hydroxyl speciation in complement to SIMS measurements, 333 334 unpolarized FTIR spectra were obtained from optically clear areas of crystallographically unoriented olivine crystals in thick sections from the aillikite pipe 335 336 and dykes (Fig. 7 and Table S5). Absorption bands due to O-H vibrations in the olivine consist of prominent overlapping peaks at 3572 and 3563 cm⁻¹, a peak at 3525 337 cm⁻¹ with a broad asymmetric shoulder at 3542 cm⁻¹, and a peak at 3484 cm⁻¹. 338 Additional four low intensity peaks are observed at 3637, 3620, 3612, 3597 cm⁻¹. 339 340 These spectra resemble hydroxyl spectra of kimberlitic olivines from the Siberian 341 (Matsyuk & Langer, 2004) and Kaapvaal (Peslier et al., 2008) cratons. Although the 342 beam was focused to $<100 \mu m$, the FTIR collects data through the entire sample 343 thickness; hence, it was unavoidable to measure minor inclusions. The peak at 3693 cm⁻¹ is related to serpentine inclusions (e.g., Matsyuk & Langer, 2004). The infrared 344 345 absorption spectra of low-Ni olivines are similar to those of high-Ni ones (Fig. 7). No 346 hydrous fluid inclusions related peaks are present in the spectra as no characteristic broad absorption bands centered around 3400 cm⁻¹ were observed (Fig. 7). 347

Hydrogen distribution within individual crystals was also determined for three high-Ni and one low-Ni olivine grains using polarized FTIR analyses on crystallographically oriented olivine grains (Figs. 6a and 8). Cross sections of these olivines reveal bell-shaped profiles with higher and homogeneous absorbance at the center (long flat plateau) compared to the grain edges (Fig. 8).

353 4.3. Helium Isotopes in Olivine

354 Helium contents and isotopic compositions in the mineral separates from the 355 Wajilitag aillikites are summarized in Table S4. Helium isotopic ratios are reported as R/Ra ratios, i.e., ³He/⁴He ratio of measured sample is normalized to the atmospheric 356 ratio (Ra =1.39 \times 10⁻⁶). As the olivine macrocrysts and microcrysts in Dyke 3 357 358 aillikites could not be confidently separated using a binocular microscope, only 359 olivine separates from Dyke 1 and 2 were analyzed because they only contain high-Ni microcrysts. The measured ⁴He abundances of the high-Ni olivine separates are 6.27-360 38.8×10^{-8} cm³ STP/g and the ³He/⁴He ratios are 5.31-5.84 Ra. 361

362 4.4. Bulk-Rock Pb Isotopic Compositions

The Pb isotopic compositions of the Wajilitag aillikite pipe and dykes are present 363 364 in Fig. 9 and Table S6. The Pb isotopic compositions of the examined aillikite samples yield present-day values for ${}^{206}Pb/{}^{204}Pb = 19.21-20.52$, ${}^{207}Pb/{}^{204}Pb = 15.65-$ 365 15.74 and ${}^{208}\text{Pb}/{}^{204}\text{Pb} = 39.31-41.00$. The measured U/Pb and Th/Pb ratios range 366 between 0.24-0.71 and 0.87-2.51, respectively. The age-corrected (270 Ma) initial Pb 367 isotope ratios define a relatively small range, including ${}^{206}Pb/{}^{204}Pb_{(i)} = 18.30-18.90$, 368 ${}^{207}Pb/{}^{204}Pb_{(i)} = 15.60-15.66$ and ${}^{208}Pb/{}^{204}Pb_{(i)} = 38.54-39.50$. These compositions 369 feature relatively high in ²⁰⁷Pb/²⁰⁶Pb and ²⁰⁸Pb/²⁰⁶Pb and are broadly intermediate 370 371 between typical Enriched Mantle (EM) and Prevalent Mantle (PREMA) compositions 372 (Fig. 9).

373 5. Discussion

In the following sections, we first use major- and trace-element compositions of olivine combined with FTIR analyses to investigate the mechanism that may control the incorporation of water into the Wajilitag olivines. We then discuss the factors that contribute to the high and variable water contents in these olivines, and assess H_2O contents of primitive aillikite magmas and their mantle source based on H concentrations in high-Fo olivines. Finally, by combining new olivine water contents and bulk-mineral He isotope analyses with new and existing Pb-Sr-Nd-Os isotope constraints, we attempt to examine the origin and role of water in the generation of the Tarim LIP.

383 5.1. Hydrogen Incorporation in Olivine

384 As mentioned above, olivines in the Wajilitag aillikites can be divided into high-385 Ni microcrysts and low-Ni macrocrysts. The well-defined linear correlations between 386 Fo, minor and trace elements (e.g., increasing Mn with decreasing Fo and Ni) (Fig. 4) 387 in the olivine microcrysts suggest that they are phenocrysts crystallized during 388 magmatic differentiation of the host aillikite magmas (Wang et al., 2021a, b). In 389 comparison, the origin of the macrocrysts, whose compositions are displaced from the 390 evolution trend of the microcrysts (Fig. 4), is more complex. These grains probably 391 represent antecrysts derived from precursor highly evolved aillikite melts based on 392 their large size, homogeneous composition, very low Fo78-81, Ni and Cr and high Mn 393 and Zn contents (Wang et al., 2021a).

Water contents in the analyzed high-Ni olivine microcrysts exhibit a relatively wide variation of 75-168 ppm H₂O (Table S1; Fig. 5). For these olivines, water contents are positively correlated with Ti concentrations, but increase gradually with decreasing forsterite contents from Fo_{78} to Fo_{88} (Fig. 5a). The positive correlation (R²

398	= 0.71 (square of the correlation coefficient); Fig. 5b) between H_2O and Ti contents
399	likely suggests a common incorporation mechanism in these olivines. As shown in
400	Fig. 7, the high-Ni olivine grains show prominent peaks at 3572 and 3525 cm^{-1} in the
401	FTIR spectra, reflecting the significance of Ti-related point defects in the olivine
402	structure (e.g., Berry et al., 2005; Kovacs et al., 2010). Alternatively, these peaks
403	could also relate to the presence of Ti-clinohumite exsolutions lamellae (e.g., Matsyuk
404	& Langer, 2004). However, the homogeneous BSE response of these olivine grains
405	(Figs. 2g-h) demonstrates that the exsolved phases, if present, must be of nanometer
406	size. It is notable that these olivines have considerably higher water contents relative
407	to hypothetical pure Ti-clinohumite-like point defects or lamellae (De Hoog et al.,
408	2014), thus the composition of the point defects or exsolution phases must deviate
409	from the ideal stoichiometry MgTi[]O ₂ (OH) ₂ . Additional peaks at 3637, 3620, 3612,
410	3597, 3563, 3542 and 3484 cm^{-1} (Fig. 7) are linked to Si-related substitution
411	mechanisms, i.e., OH is incorporated as $Mg_2[V_{Si}]OH_4$, where $[V_{Si}]OH_4$ represents a
412	Si vacancy (e.g., Berry et al., 2005; Kovacs et al., 2010). The lack of correlation ($R^2 =$
413	0.13) between H_2O and SiO_2 seems to be in contradiction with the FTIR observations.
414	However, since Si is a major element in olivine, trace quantities of H substitutions in
415	the tetrahedral site are unlikely to be detected in SiO_2 variations. As shown in Fig. 7d,
416	the two spectra from the low-Ni olivine macrocrysts are similar to the high-Ni
417	microcrysts, suggesting that H-related substitutions associated with Ti-related point
418	defects and Si vacancies may also account for the OH incorporation in the low-Ni
419	olivines. We thus propose that hydrogen in the Wajilitag olivines is related to coupled

substitution of H with various vacancies in olivine, not only Ti or not solely Si
vacancies, even if dominant. Consequently, the main H diffusion mechanism will be
rate-controlled by vacancy diffusivity and not by the faster proton-polaron exchange
or interstitials jumps (see Thoraval and Demouchy, 2014).

424 5.2. Origin of H₂O Variations in Olivine

425 Water contents in olivine from the Wajilitag aillikites are higher (75 to 168 ppm 426 H₂O; Fig. 5) than those from alkali basalts (4-70 ppm H₂O, Demouchy & Bolfan-427 Casanova, 2016) and South Africa cratonic peridotites (up to 120 ppm H_2O , see 428 Peslier, 2010), while the highest water contents are as high as those reported from the Siberian cratonic peridotites (200 ppm H₂O, Doucet et al., 2014, within 20 % error 429 430 bar). Five possible mechanisms may account for these observations, including (1) 431 post-emplacement hydrothermal processes (e.g., serpentinization; Stripp et al., 2006); 432 (2) assimilation of hydrous crustal materials; (3) addition of H by ionic diffusion from 433 the host volatile-rich aillikite melts after olivine crystallization (Demouchy & 434 Mackwell, 2006); (4) incomplete H loss from olivine upon magma ascent (Demouchy 435 et al., 2006; Ferriss et al., 2018); and (5) H acquired by magmatic olivine during 436 crystallization (Sokol et al., 2013).

Aillikites may contain abundant serpentine precipitated from post-emplacement
hydrothermal fluids (Tappe et al., 2006) and the Wajilitag aillikites are no exception.
Thus, H could be added into olivine during interaction with post-emplacement
hydrothermal fluids. However, the extremely low diffusivity of H in olivine at typical
serpentinization temperatures for these or similar rocks (commonly ≤400 °C; Stripp et

442 al., 2006) and at the shallow pressures (<3 km depth) of magma intrusion implies that 443 more than 10,000 years would be required for a millimeter sized olivine crystal to 444 attain high water contents (e.g., >30 ppm H₂O; Demouchy & Mackwell, 2006). 445 Considering that the timescale for alkaline ultramafic rocks cooling and hydrothermal 446 activity is centuries to a few millennia for large pipes (Sparks et al., 2006), 447 hydrothermal alteration is thus an unlikely reason for the high H concentrations in the 448 Wajilitag olivines. In addition, at these low-temperature conditions, interaction of 449 olivine with hydrous fluids would be more likely to serpentinize olivine rather than 450 increasing water contents by diffusion.

451 Assimilation of hydrous crustal materials can lead to high water contents in 452 magma and olivine crystallized from the system. However, the presence of diamonds 453 in these aillikites (Zhang et al., 2013b) indicates that these magmas ascended rapidly 454 toward the Earth's surface and did not stagnate in crustal magma chambers where 455 assimilation would have occurred. Moreover, the lack of correlations between bulkrock SiO₂ contents, trace element ratios (Nb/U and Ce/Pb), ⁸⁷Sr/⁸⁶Sr_(i) and 456 ¹⁸⁷Os/¹⁸⁸Os_(i) (Fig. S4) also rule out a prominent role for crustal assimilation (see 457 458 Wang et al., 2021b, 2022). Thus, it is less likely that the magma became more hydrous 459 through crustal contamination.

Alkaline ultramafic rocks such as aillikites may contain several weight percent
H₂O retained in various hydrous magmatic phases such as amphibole and phlogopite
(Figs. 2c and S1 in Supporting Information S1), suggesting that their parental magmas
are enriched in water (e.g., Rock et al., 1986; Tappe et al., 2006). Moreover, as H is an

464 incompatible element during igneous differentiation (Demouchy & Alard, 2021), the 465 water content in evolving mafic-ultramafic magmas will continuously increase 466 following fractional crystallization of early-stage nominally anhydrous mineral 467 (NAM) such as olivine and pyroxene. Coupled with the high diffusivity of H in 468 olivine at magmatic temperatures, addition of H into olivine via ionic diffusion from 469 the evolved H_2O -rich aillikite melts is thus possible (Demouchy & Mackwell, 2006; 470 Peslier et al., 2008). However, this process would generate reversely zoned diffusion 471 profiles with lower H in the cores than at the edges. In contrast, as shown in Figs. 6a 472 and 8, OH absorbance profiles across the olivine grains display distinctive core-to-rim 473 depletion of H rather than enrichment. These results rather suggest that the Wajilitag 474 olivines may have been affected by H loss during syn-eruptive degassing.

475 Alkaline ultramafic magmas typically ascend very rapidly through the 476 lithosphere (Sparks et al., 2006), and hence olivine phenocrysts and megacrysts in 477 these rocks can preserve pristine water contents (i.e., minimally affected by diffusion) 478 at least in the cores (Bell et al., 2004; Matveev & Stachel, 2007; Peslier et al., 2008). 479 As shown in Figs. 6a and 8, the Wajilitag olivine crystals have homogenous H 480 distribution in the core regions forming flat plateaus (see also Thoraval & Demouchy, 481 2014). In addition, the water contents of high-Ni olivine microcrysts increase 482 gradually with decreasing Fo values (Fig. 5a) and all the examined olivine grains have 483 almost homogeneous Fo values within individual grains (e.g., Figs. 6b and S2) but 484 exhibit wide variability in the grain population (Figs. 4-5). Moreover, the lower- H_2O , 485 higher-Fo₈₁₋₈₈ olivine microcrysts predominantly occur in the less evolved Dyke 1 and 2 aillikites, while the higher-H₂O, lower-Fo₇₈₋₈₁ microcrysts are mainly present in evolved Dyke 3 (Table S1). These lines of evidence suggest that (1) the variation of water contents in the Wajilitag olivines is likely mainly controlled by fractional crystallization (recorded by Fo decrease, Fig. 5a); and (2) the H values observed in the cores of most grains represent pristine water contents gained by olivine during crystallization from differentiating and gradually water-richer aillikite magmas.

492 **5.3. Estimation of Dehydration Duration**

493 As illustrated in Figs. 6a and 8, the H values at the edges of the Wajilitag olivines 494 are generally lower than those in the cores, possibly indicating partial loss of H during 495 transport of the magmatic olivine to the surface. We modelled the H profiles of these 496 olivine crystals using the diffusion equations of Demouchy & Mackwell (2006) to 497 estimate the duration of H loss. Hydrogen diffusion is governed by the following 498 parameters: (1) temperature and thus H diffusion coefficients (D) in olivine at the 499 given temperature; (2) the diffusion mechanism, either the proton-polaron exchange 500 or proton-vacancy diffusion; and (3) the crystallographic direction of H diffusion in 501 olivine crystal (Demouchy & Mackwell, 2006). The equilibrium temperatures of the 502 Wajilitag olivines were computed from olivine-spinel thermometers and yielded 950-503 1150 °C (average 1040 °C; Wang et al., 2021b). The crystallographic orientations for 504 olivine profiles were established through EBSD analysis. The hydrogen diffusion 505 coefficient in the olivine can be calculated using the following equations established 506 for olivine of Fo₉₀ composition (Demouchy & Mackwell, 2006):

507
$$D_{\alpha \text{ and } \gamma} = 10^{-4.5} \exp(-204,000/\text{RT})$$

508
$$D_{\beta} = 10^{-1.4} \exp(-258,000/RT)$$

where $D_{\alpha, \beta, \gamma}$ are the H diffusion coefficients in olivine along the α , β and γ optical axes ($\gamma = [100]$, $\alpha = [010]$ and $\beta = [001]$), R is the gas constant (8.31447 JK⁻¹ mol⁻¹) and T is the temperature (K).

512 The 1-dimensional diffusion equation of Demouchy & Mackwell (2006) was 513 used to model the entire H diffusion profiles (e.g., from edge to edge) across the 514 analyzed olivine grains (Fig. 8):

$$C_{OH}(x) = C_0(\operatorname{erfc}(\frac{x}{2\sqrt{Dt}}) + \operatorname{erfc}(\frac{(X-x)}{2\sqrt{Dt}}))$$

515 where $C_{OH}(x)$ represents the hydrogen concentrations (or as in this study, the 516 integrated hydroxyl absorbance) at a distance x (m) from the edge, C_0 represents the 517 initial integrated hydroxyl absorbance (e.g., the core absorbance), D is the diffusion coefficient (m^2s^{-1}) , X is the width of the diffusion profile from edge to edge, t is the 518 519 diffusion time (s). Using the H diffusion coefficients (e.g., $D_{\alpha, \beta \text{ or } \gamma}$) as calculated 520 above and the olivine equilibration temperature of 1040 °C, resulting diffusion 521 profiles produced by the fitting curves yield a dehydration duration of between 15 and 522 417 min (Fig. 8). As aillikite melts do not stagnate in crustal magmatic chambers and 523 ascend rapidly to the surface (e.g., Sparks et al., 2006), thus the temperature 524 difference between olivine crystallization and H diffusion during magma degassing is 525 expected to be small (e.g., <100 °C). Using experientially determined diffusion 526 coefficients of H (e.g., D α , β or γ) for a shallow magma temperature of 940 °C, the 527 calculated diffusion profiles yield durations of between 40-660 min for the four 528 olivine grains in Fig. 8. These dehydration durations significantly overlap with the

range (15-417 min; Fig. 8) calculated from a higher temperature of 1040 °C,
indicating a limited effect of a slightly lower temperature. This is in agreement with
previous study by Thoraval & Demouchy (2014).

532 We also modelled diffusion in 3-dimensions following the method of Thoraval & 533 Demouchy (2014) to calculate the duration of dehydration for the observed 534 concentration profiles across the four olivine grains (Ol-1, Ol-2, Ol-3, and Ol-4; Fig. 535 S5 in Supporting Information S1). The fastest direction of diffusion for H is [001] and 536 dehydration is predominantly driven by this crystallographic direction. As profiles 537 were acquired on thin section, there is uncertainty on the initial geometry of each 538 olivine grain (only 2 directions known and not 3). Nevertheless, we decided to 539 approximate the grain shape with a rectangle shape with [001] twice the length of 540 [100] and [010] for Ol-1, Ol-2 and Ol-4; and [010] twice the length of [001] and [100] 541 for Ol-3, with the initial water concentration set as the concentration in the central 542 plateau. Modeled diffusion profiles are calculated using the H diffusion coefficient (e.g., D_{[100],[010] or [001]}) calculated above for a temperature of 1040 °C. Figure S5 543 544 displays the results of the modelling for each crystallographic axis and for each 545 olivine. Similar to 1D diffusion models, the 3D model yields very short durations (16 546 min) of dehydration for Ol-1 and Ol-2 and longer durations (135-592 min) for Ol-3 547 and Ol-4 (see Fig. S5 in Supporting Information S1).

548 5.4. Water Content in the Parental Aillikitic Magmas and their Mantle Source

549 As the water contents of the Wajilitag olivines are affected by magma 550 differentiation and, to a very localized extent, volatile degassing, only the cores of

551	olivine crystallized at the earliest stage can be used to infer the H ₂ O contents of the
552	primitive aillikite melt (Peslier et al., 2008). The highest forsterite values (Fo _{87.7}) of
553	the analyzed olivines (Table S1) are similar to those of the most forsteritic olivine
554	phenocrysts crystallized from primitive mantle-derived melts (Herzberg et al., 2011).
555	Furthermore, the Mg# value (74.1) of the equilibrium melts for these high-Mg olivine
556	grains calculated using the Fe-Mg partition coefficient ($Kd_{Fe-Mg}^{olivine-melt}$) of 0.40 for
557	silicate-carbonate melts (Giuliani, 2018 and references therein) is marginally higher
558	than the aillikite sample with highest Mg# (73.1, Table S7; Wang et al., 2021a).
559	Therefore, it is suggested that these high-Fo olivines crystallized from primitive
560	aillikite melts and can be used to infer the initial melt H_2O contents. The
561	experimentally determined partition coefficient for H between olivine and carbonated
562	silicate melt $(D_{H2O}^{Ol-melt})$ is the key parameter for this calculation and ranges from
563	0.006-0.009 to 0.002-0.004 with molar CO ₂ /(CO ₂ +SiO ₂) in the melt varying from 0 to
564	0.4 (Aubaud et al., 2004; Bell et al., 2004; Sokol, et al., 2013). Using a H_2O content of
565	93 ppm H_2O for olivine (e.g., average value for the four highest-Fo olivine
566	compositions, Fig. 5a), and partition coefficients between olivine and primitive
567	aillikite melt $(CO_2/(CO_2+SiO_2) = 0.3-0.4$; Tappe et al., 2006; Wang et al., 2021a) of
568	0.002-0.006, the water contents of the melt are estimated as 1.6 to 4.7 wt.%. The
569	water content of the primitive aillikite melt is therefore higher than the range shown
570	by ocean island basalts (OIB, 0.3-1.0 wt.%) and mid-ocean ridge basalts (MORB, 0.1-
571	0.3 wt.%), but overlaps those of arc magmas (2.0-8.0 wt.%) (Xia et al., 2016 and
572	references therein). The water content resembles primary H ₂ O content values

573 proposed for natural kimberlite melts (2-9 wt.%) (Moussallam et al., 2016).

574 Next, we estimate the H₂O content of the mantle source to infer if aillikites 575 derive from mantle domains enriched in H₂O. The garnet-bearing peridotite versus carbonated silicate melt partition coefficient $(D_{H20}^{peridotite-melt})$ for H₂O depends on 576 577 mineral abundances in the peridotite source and their respective mineral D values 578 (Bell et al., 2004; Sokol et al., 2013). This bulk D_{H20} for peridotite-melt is calculated 579 by combining individual mineral D values with their modal proportions in typical 580 garnet-bearing peridotite source (mineral modes taken from Sokol et al. (2013): 72% olivine, 21% orthopyroxene, 6% garnet, 3% clinopyroxene, and 1% spinel). 581 Assuming that $D_{H2O}^{Ol-melt}$ values = 0.002-0.006, $D_{H2O}^{Px-Ol} = 2$ (Ardia et al., 2012; Aubaud 582 et al., 2004; Hirschmann et al., 2009) and $D_{H20}^{Grt-Ol} = 5$ (Mookherjee et al., 2010), 583 $D_{\rm H2O}^{\rm peridotite-melt}$ between garnet-bearing peridotite source and carbonate-silicate melt 584 585 $(CO_2/(CO_2+SiO_2) = 0.3-0.4)$ ranges from 0.005 to 0.01. The water content of the 586 peridotite source for the Wajilitag aillikites is estimated using models for both batch 587 and fractional melting, and assuming a partial melting degree of 0.7-1.5% (Tappe et 588 al., 2006; detailed calculations are present in Text 2 in Supporting Information S1). 589 The results show that the calculated water contents in the mantle source of these 590 aillikites are 150-1200 ppm H_2O (average 520 ppm H_2O), which is significantly 591 higher than those of the depleted upper mantle (MORB source) (e.g., <200 ppm H₂O; 592 Hirschmann, 2018; Shimizu et al, 2019), but falls in the range of source water 593 contents for OIBs (300-1000 ppm H₂O; Bizimis & Peslier, 2015). A previous study 594 (Xia et al., 2016) based on the analyses of clinopyroxene phenocrysts in flood basalts also proposed that the Tarim mantle plume was enriched in water (up to 3000-5000 ppm H_2O). If these elevated water contents are typical of the Tarim plume, they help explain the occurrence of voluminous magmatism in the Western Tarim LIP due to the dumping effect of water on the peridotite solidus (e.g., Kawamoto & Holloway, 1997; Liu et al., 2022).

600 5.5. Implications for Earth's Deep Water Cycle

601 It has been previously shown that the majority of igneous units in the Tarim LIP 602 including basalts, aillikites, alkaline lamprophyres, nephelinites and mafic-ultramafic 603 intrusions are characterized by Fe- and Ti-rich signatures and enriched (OIB-like) 604 primitive mantle nominalized trace element patterns (Cheng et al., 2018; Zhou et al., 605 2009). Combination of these geochemical features with constraints from Sr-Nd-Os-C-606 O-Mg isotopes has been used to invoke a mantle plume source polluted by 607 subduction-related components for these magmas and related carbonatites (e.g., 608 Cheng et al., 2018; Wang et al., 2022). This interpretation is further reinforced by 609 previous regional geological studies, which suggest that the Southern Tianshan 610 oceanic plate was subducted beneath the Tarim craton in Paleozoic period (e.g., ca. 611 200 Ma before the formation of the Tarim LIP; Charvet et al., 2011; Ge et al., 2012; 612 Wang et al., 2011). Our new results for the Wajilitag aillikites that were emplaced 613 shortly after the eruption of the Tarim flood basalts provide further insights into the 614 nature of the Permian Tarim plume.

615 The high-Ni olivine microcrysts in Dyke 1 and 2 originated from primitive 616 aillikite melts (Wang et al., 2021a). Therefore, the noble gases of these olivines can be

617	used to constrain the mantle source compositions of the aillikites. The ${}^{3}\text{He}/{}^{4}\text{He}$
618	isotope ratios (5.31-5.84 Ra; Table S4) of these olivine grains are much higher than
619	those of continental crust and atmosphere (Allegre et al., 1987; Ballentine & Burnard,
620	2002), but slightly lower than those of MORBs (8 \pm 1 Ra; Graham, 2002) and
621	significantly lower than those of hot and buoyant plumes sourced from the lowermost
622	mantle (~8-50 Ra; Jackson et al., 2017). These He isotope compositions overlap the
623	range of xenoliths from the sub-continental lithospheric mantle (SCLM, 5-6 Ra) (Day
624	et al., 2005, 2015; Gautheron et al., 2005), which may indicate that these aillikites
625	derived from a SCLM source. However, the depleted Sr-Nd isotopes (87 Sr/ 86 Sr _(i) =
626	0.70340-0.70390; $\epsilon Nd_{(i)} = +1.3$ to +4.9; Wang et al., 2022) of primary magmatic
627	apatite and perovskite in the aillikites are more consistent with an asthenospheric
628	(plume) mantle source rather than metasomatised domains in the lithospheric mantle,
629	which would rather feature geochemically enriched Sr-Nd isotope compositions.
630	Therefore, the lower-than-MORB He isotopes are better attributed to the influence of
631	deeply recycled hydrous crustal material in the source, which contained some U and
632	therefore in-grew ⁴ He during mantle residence (e.g., Hanyu & Kaneoka, 1997). This
633	interpretation is consistent with moderately radiogenic Os isotope compositions of
634	these rocks (Table S7; Wang et al., 2022) and new bulk-sample Pb isotope data, which
635	are intermediate between PREMA (i.e., the possible deep-mantle component common
636	to aillikites and kimberlites; Giuliani et al., 2021) and EM (i.e., the recycled hydrous
637	crustal component) (Fig. 9). This multi-isotopic approach highlights the complex
638	make-up of the Tarim plume and the presence of deeply subducted (or delaminated)

639 hydrous crustal components, which contributed to the formation of voluminous640 volcanic outpours.

641 The subducted (or delaminated) hydrous crustal components in the Tarim plume 642 are likely derived from the deeply subducted Southern Tianshan oceanic slab (Cheng 643 et al., 2018). To constrain how much water was retained in the subducted crustal 644 component, we here employ the approximate H₂O contents (i.e., 520 ppm) of the 645 mantle plume source of the Wajilitag aillikites. Based on Os isotope mass balance 646 calculations Wang et al. (2022) suggested that less than one third of recycled oceanic 647 crustal material can occur in the Tarim plume. If we assume 30% of water in the 648 mantle plume source was originated from deeply subducted oceanic material and the 649 remaining 70% from a depleted mantle source containing 200 ppm H_2O , the recycled 650 oceanic crust should contain ca. 1300 ppm H_2O . It is suggested that globally oceanic 651 crust contains 2-3 wt.% H₂O (Dixon et al., 2002). If so, the deeply subducted oceanic 652 slab might have lost more than 93% of its original water content since subduction. 653 These dehydration estimates are consistent with other approaches (i.e., based on H 654 isotope and H₂O/Ce variation of oceanic crust before and after subduction; Dixon et 655 al., 2002; Shaw et al., 2008).

656 **6.** Conclusions

This study presents H_2O , major-, trace-element and bulk-mineral He isotope compositions of olivine from the Wajilitag aillikites in the Tarim large igneous province, NW China. Olivines in the aillikites are divided into two groups: (i) high-Ni microcrysts (<5 mm) and (ii) low-Ni macrocrysts (5-25 mm). The former are

661	phenocrysts crystallized directly from the aillikite melts, while the latter probably
662	represent antecrysts produced from evolved aillikite melts during early pulses of
663	magmatic activity. The water contents are high for both the microcrysts and
664	macrocrysts and range between 75-168 ppm $\rm H_2O$ and 103-161 ppm $\rm H_2O,$ respectively.
665	The positive correlation between H_2O and Ti in these olivines, combined with infrared
666	hydroxyl bands at 3572 and 3525 cm ⁻¹ , suggests that H is mainly incorporated as Ti-
667	clinohumite-like point defects. FTIR profile analyses using polarized infrared
668	radiation reveal water loss during decompression, but limited to the rims of olivines.
669	Conversely, long flat plateaus with homogeneous water concentration in the cores
670	provide pristine magmatic water contents of liquidus olivine. Using experimental
671	diffusion data for H, the duration of H loss is estimate as 15-417 min for the olivines
672	with lower water contents in the rim zones. Based on the water contents of the cores
673	of primitive high-Fo olivines, the minimum water contents of the primitive aillikite
674	melts and their mantle source are estimated as 1.6-4.7 wt.% and 150-1200 ppm $\mathrm{H_2O},$
675	respectively. These water contents in the mantle source are higher than those typical
676	of the depleted upper mantle (i.e., MORB source) hence requiring addition of a water-
677	bearing component. Additionally, the ${}^{3}\text{He}/{}^{4}\text{He}$ ratios (5.31-5.84 Ra) of primitive high-
678	Ni olivines are slightly lower than the range of MORBs (i.e., depleted upper mantle).
679	In combination with depleted Sr-Nd isotopes of primary apatite and perovskite, and
680	moderately radiogenic Pb and Os isotopes of bulk samples (Wang et al., 2022), these
681	new results suggest that water in the aillikites was mainly sourced from a plume
682	contaminated by deeply recycled (U-bearing) hydrated crustal material. This study

reinforces the view that the Tarim plume was wet (Xia et al., 2016), but clarifies that much of this water was stored in deeply subducted material that polluted the plume source in the lowermost mantle.

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Data Availability Statement

The authors comply with the AGU's data policy, and the data sets of this paper are available at https://doi.org/10.6084/m9.figshare.21067804.v5.

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994 Figure captions

Figure 1. (a) Location of the Tarim large igneous province in NW China, which
shows the position of Permian Tarim flood basalts, rhyolites and mafic-ultramafic
intrusions (after Zhou et al., 2009). (b) Detailed geological map of the Wajilitag area
showing the distribution of aillikites (after Cheng et al. 2018).

999 Figure 2. (a) Representative field photograph of the Wajilitag aillikites. (b) 1000 Photograph of a representative aillikite hand specimen. (c) Optical microphotograph 1001 showing the presence of abundant phenocrysts of olivine, clinopyroxene and hydrous 1002 silicate minerals (amphibole and phlogopite) distributed in the aillikite matrix, plane-1003 polarized light. (d -f) Representative photomicrographs of olivine microcrysts in Dyke 1004 1 (d), 2 (e) and 3 (f) and macrocryst in Dyke 3 (f), plane-polarized light. (g-h) Back-1005 scattered electron (BSE) images of representative olivine microcrysts. (i) Primary 1006 melt inclusions with vermicular shapes in the interiors of the olivine microcrysts. Ol, 1007 olivine; Cpx, clinopyroxene; Phl, phlogopite; Amp, amphibole; Ap, apatite; Cal, 1008 calcite.

Figure 3. Water content calibration curve for olivine analyses by SIMS. Independent analyses of the water content of the olivine references were acquired using FTIR analyses. Note that the error bars of KLB-1 and San Carlos are smaller than the symbols. The solid line represents linear regressions to the data, and R^2 is the square of the correlation coefficient.

1014 Figure 4. Fo versus NiO (a), CaO (b), MnO (c) and Ti concentrations for olivine
1015 microcrysts (orange) and macrocrysts (blue) from the Wajilitag aillikite dykes. Note

that FeO, MgO, NiO, CaO and MnO contents of the olivines were measured by
EMPA and Ti concentrations by LA-ICP-MS. Previous data for the Wajilitag olivine
microcrysts (grey cycle) and macrocrysts (white diamond) are from Wang et al.
(2021a, b).

Figure 5. Water contents determined by SIMS as a function of selected major and trace element concentrations for olivine microcrysts (orange) and macrocrysts (blue) in the Wajilitag aillikite dykes. FeO and MgO contents for the olivines were measured by EMPA and Ti, Ca and Cr concentrations by LA-ICP-MS. The dotted line in Fig. 5a shows the assumed (without calculation) trend of Fo and H₂O concentration during fractional crystallization. The dashed lines in Fig. 5b represent linear regressions to the data, and R^2 is the square of the correlation coefficient.

Figure 6. Compositional traverse for a representative olivine microcryst shown inFig. 2h (red line). FeO and MgO contents for the olivines were measured by EMPA.

1029 Water contents of the olivine are from FTIR measurements and Ni, Mn, Ca, and Ti

1030 concentrations are from LA-ICP-MS measurements.

1031 Figure 7. Unpolarized Fourier transform infrared absorption spectra of randomly

1032 oriented olivines in aillikite pipe (a) and Dyke 1 (b), 2 (c) and 3 (d). Olivines from

- 1033 different aillikite pipe and dykes display similar FTIR spectra. Spectra are normalized
- 1034 to thickness of 1 cm and each spectrum represents an individual measurement.
- 1035 Figure 8. Profiles of H absorbance values (normalized to thickness of 1 cm) and
- 1036 diffusion modeling for (a-c) three olivine microcrysts (Ol-1, Ol-2 and Ol-3) and (d)
- 1037 one macrocryst (Ol-4) from the Wajilitag aillikites. O-H integrated absorbance are

1038	measured from polarized FTIR spectra in the 3700-3000 cm ⁻¹ region due to O-H
1039	vibrations. Modeled diffusion profiles are calculated following methods described in
1040	the text and using the H diffusion coefficient (D) from Demouchy and Mackwell
1041	(2006) for a temperature of 1040 °C. The input parameters into modelling are $A_0 =$
1042	initial absorbance in the core, $t = diffusion$ time and $X = the$ width of the diffusion
1043	profile from edge to edge. Error bars on the y-axis (water content) represent 10 %
1044	uncertainty, and those on the x-axis represent the aperture size during FTIR analyses.
1045	All integrated absorbances are calculated for a sample thickness of 1 cm.
1046	Figure 9. Age-corrected (270 Ma) initial ${}^{206}Pb/{}^{204}Pb_{(i)}$ vs. ${}^{87}Sr/{}^{86}Sr_{(i)}$ (a), $\epsilon Nd_{(i)}$ (b),
1047	207 Pb/ 204 Pb _(i) (c) and 208 Pb/ 204 Pb _(i) (d) diagram for the Wajilitag aillikites. The Pb, Sr
1048	and Nd isotope compositions of MORB, HIMU (high ²³⁸ U/ ²⁰⁴ Pb ratio), enriched and
1049	PREMA mantle are also plotted for comparison (Stracke, 2012 and references
1050	therein). NHRL: Northern Hemisphere Reference Line (Hart, 1988).

Figure 1.



E78°57'15.6"



Figure 2.



Figure 3.



Figure 4.



Figure 5.



Figure 6.



Figure 7.



Figure 8.

Figure 9.

