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## **NORTHWEST AFRICA (NWA) 12563 and ungrouped C2 chondrites: Alteration styles and relationships to asteroids**

R.H. Hewins, P.-M. Zanetta, B. Zanda, C. Le Guillou, J. Gattacceca, C. Sognzoni, S. Pont, L. Piani, T. Rigaudier, H. Leroux, et al.

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4 **NWA 12563 and ungrouped C2 chondrites: Alteration styles and relationships**  
5 **to asteroids**  
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33 **Abstract**—Many asteroids in the main belt have spectra like those of Mighei-type CM chondrites, but  
34 some Near Earth Objects (NEO) resemble less well known types of C2 chondrite. Northwest Africa  
35 (NWA) 12563, a new find with affinities to C2 chondrites, could help us understand the differences  
36 between observations of CM2 chondrites and bodies that are currently being studied by the Hayabusa 2  
37 and Osiris-Rex space missions. NWA 12563 contains 14% chondrules supported by 86% fine grained  
38 matrix consistent with CM2 chondrites, but differs in other respects. In both matrix and chondrules,  
39 olivine is unaltered and pyroxene shows incipient alteration. Metal in chondrules is pseudomorphed by  
40 serpentine, and mesostasis is replaced by serpentine- saponite and chlorite. Many Type I chondrules have  
41 gerrymandered shapes resulting from fracturing and selective metal replacement. Type II porphyritic  
42 chondrules are clusters of phenocrysts set in matrix-like material. Type II chondrules may be kinked and  
43 partially disbarred. The matrix of NWA 12563 is characterized by the absence of tochilinite-cronstedtite  
44 intergrowths. It contains hydrated and oxidized amorphous silicate ( $\text{Fe}^{3+}/\Sigma\text{Fe} \sim 75\%$ ) richer in magnesium  
45 than in other chondrites (with embedded sulfides). Serpentine-saponite is also present, as well as  
46 abundant framboidal magnetite.

47 NWA 12563 has similarities to a number of ungrouped magnetite-rich and  $^{18}\text{O}$ -rich chondrites (Bells,  
48 Essebi, Niger I, WIS 91600, Tagish Lake, Dho 1988 and MET 00432) that we call C2-ung1, as opposed to  
49  $^{18}\text{O}$ -poor C2-ung2 chondrites. The oxygen isotopic composition coupled with a magnetic susceptibility of  
50  $\log \chi = 4.67$  places NWA 12563 with these ungrouped chondrites in a cluster distinct from CM2 chondrites.  
51 NWA 12563 is closest to WIS 91600 among the C2-ung1 chondrites in alteration style and light element  
52 compositions. WIS 91600, however, has suffered light thermal metamorphism, suggesting that NWA 12563  
53 might represent its altered but unheated precursor material within the same parent body if it were zoned.  
54 The average Vis-NIR spectrum of NWA 12563 matches the asteroid taxonomic class K and resembles that  
55 of CO3 Frontier Mountain (FRO) 95002, but its spectra range from very “red” in dark matrix areas and very  
56 “blue” in magnetite-rich areas. The average MIR spectrum shows features indicating phyllosilicates,  
57 aliphatic CH compounds, hydrated silicates, and olivine. It is significantly different from those of other  
58 chondrites including FRO 95002, and closest to Bells (from which it differs in carbon isotopic composition)  
59 and WIS91600. The variety of mineralogical, chemical and isotopic properties among C2-ung1 chondrites  
60 requires several different parent bodies. However, the high abundance of magnetite common to this cluster  
61 of ungrouped chondrites, and to a lesser extent CI chondrites, indicates that they should be considered as  
62 possible material from Bennu, which has an 18  $\mu\text{m}$  magnetite signal in its spectrum not seen in the CM2  
63 chondrites (Hamilton et al., 2019).

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## 1. INTRODUCTION

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C-complex asteroids form a major fraction of the bodies in the main asteroid belt at ~3-4 AU (Burbine, 1998; Vernazza and Beck, 2017, Vernazza et al., 2017), and were probably formed beyond the snow line whether this was in the inner or outer solar system (Warren, 2011; Walsh et al., 2011). The largest category, the Ch- and Cgh-types, comprises ~70 asteroids in the main belt, as well as family members, and is well matched spectrally by CM carbonaceous chondrite meteorites (Vernazza et al., 2017). As the only well-established meteorite analogs of C-complex asteroids, the geologic history of the CMs is particularly important. They contain high temperature particles, including chondrules, and fine-grained hydrated matrix reflecting processes in the disk and in asteroidal bodies. The source of the ice accreted into asteroids and the timing and conditions of aqueous alteration are both key issues in early solar system history. From the CM group we have learned that water from isotopically different inner and outer solar system reservoirs was present (Piani et al., 2018).

Many other types of asteroids have not been definitively matched with chondrite groups as yet. D-type asteroids were proposed to be related to the Tagish Lake C2-ung chondrite (Hiroi et al., 2001) yet Vernazza and Beck (2017) report that they lack the hydrated minerals found in Tagish Lake. B-type asteroid spectra can be matched by CI chondrite provided that magnetite is added (Yang and Jewett, 2010). Of the two NEO asteroids recently visited by spacecraft, Ryugu is a Cb asteroid (Sugita et al., 2019) similar spectrally to some C2-ung and CY chondrites that experienced heating (Tonui et al., 2014; Kitazato, 2019; King et al., 2019). The other, Bennu, is classified as a B-type asteroid (Clark et al. 2011) and may be composed of C2 rather than CM2 material (de León et al., 2018; Tachibana et al., 2019; Kitazato et al., 2019; Lauretta et al., 2019; Hamilton et al., 2019). Deciphering the different properties and histories of rare ungrouped and anomalous C meteorites (C2-ung and CM-an) which have some similarities to CM2 and some to CI chondrites, and vary among themselves, could lead to a broader basis for interpreting C-complex and other asteroids.

Here we report on a new carbonaceous chondrite NWA 12563 which contains an unusual combination of mineral assemblages consistent with a different alteration history from CM2 meteorites. Its matrix contains abundant magnetite and serpentine-saponite rather than the cronstedtite and tochilinite characteristic of CM2 chondrites. Its similarities to several magnetite-rich C2-ung chondrites that have higher  $\delta^{18}\text{O}$  than CM2 chondrites (Greenwood et al., 2020) potentially illuminate the geological histories of the other asteroid groups. This work is timely, in view of current missions to explore NEO asteroids Ryugu and Bennu (Tachibana et al., 2019; Kitazato et al., 2019; Lauretta et al., 2019; Hamilton et al., 2019).

## 2. ANALYTICAL METHODS

### 2.1. Oxygen Isotope analysis

Three-oxygen-isotope analysis was carried out at the Stable Isotopes Laboratory of CEREGE. We analyzed one 1.5 mg aliquot of a powdered 20 mg bulk sample washed in HCl (1N) for 2 hours. Molecular oxygen was extracted using laser fluorination. The aliquot was heated with a 30 W CO<sub>2</sub> IR laser in the presence of 100 mbar of BrF<sub>5</sub>. The released gas was purified through two cryogenic nitrogen traps and one heated KBr trap. Molecular oxygen was trapped for 10 mn in a molecular sieve cooled at -196°C. The gas was then expanded at 100°C and passed through a molecular sieve cooled at -114°C slush for 5 mn to refreeze possible interfering gases. The gas was then trapped again for 5 mn in the molecular sieve cooled at -196°C, then expanded again at 100°C directly into the bellow of a dual-inlet Thermo-Finnigan Delta Plus mass spectrometer.

115 The isotopic compositions are expressed in standard  $\delta$ -notation, relative to Vienna standard mean  
116 ocean water (VSMOW):  $\delta^{18}\text{O} = (^{18}\text{O}/^{16}\text{O})_{\text{sample}} / (^{18}\text{O}/^{16}\text{O})_{\text{VSMOW}} - 1$  and  $\delta^{17}\text{O} = (^{17}\text{O}/^{16}\text{O})_{\text{sample}} /$   
117  $(^{17}\text{O}/^{16}\text{O})_{\text{VSMOW}} - 1$  (expressed in ‰),  $\Delta^{17}\text{O}$ , also expressed in ‰, is defined as  $\delta^{17}\text{O} - \lambda \times \delta^{18}\text{O}$ . For  
118 comparison with literature data, we use  $\lambda = 0.52$ . Measured  $\delta^{18}\text{O}$  and  $\delta^{17}\text{O}$  values of the samples were  
119 corrected daily using the laboratory quartz standard itself calibrated against the international standard  
120 NBS28 ( $\delta^{18}\text{O} = 9.60$  ‰,  $\delta^{17}\text{O} = 0.52 \times \delta^{18}\text{O} = 4.992$  ‰). Reproducibility (1 sigma) of the quartz laboratory  
121 standard are 0.119‰ for  $\delta^{18}\text{O}$ , 0.061‰ for  $\delta^{17}\text{O}$ , and 0.024‰ for  $\Delta^{17}\text{O}$  ( $n = 63$ ) with a  $\Delta^{17}\text{O}$  (defined as  $\delta^{17}\text{O}$   
122  $- \lambda \times \delta^{18}\text{O}$  with  $\lambda = 0.52$ ) of  $-0.015 \pm 0.024$ ‰.

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## 124 **2.2. Magnetic methods**

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126 Hysteresis measurements were performed on a 160.2 mg bulk sample at CEREGE with a Princeton  
127 Micromag Vibrating Sample Magnetometer with a maximum applied field of 1 T and a sensitivity of  $\sim 5 \times 10^9$   
128  $\text{Am}^2$ . The analysis of hysteresis loops provided the ratio of saturation remanent magnetization ( $M_{\text{RS}}$ ) to  
129 saturation magnetization ( $M_{\text{S}}$ ) and the coercive force (BC). High field susceptibility ( $\chi_{\text{HF}}$ ) was determined  
130 by a linear fit for applied fields  $> 0.9$  T of the hysteresis loops. Remanent coercive force ( $B_{\text{CR}}$ ) was  
131 determined by DC back field experiments performed with the VSM. Low-temperature remanence  
132 measurements were performed with an MPMS from Quantum Design®. This instrument has a moment  
133 sensitivity of  $10^{-11} \text{Am}^2$ . The low field specific susceptibility ( $\chi$  in  $\text{m}^3/\text{kg}$ ) and its evolution with temperature  
134 were measured using Agico MFK1 apparatus with sensitivity of  $5 \times 10^{-13} \text{m}^3$ , operating at 200 A/m and a  
135 frequency of 976 Hz, equipped with a CS3 furnace and a CSL cryostat. Isothermal remanent magnetizations  
136 (IRM) were imparted using a pulse magnetizer from Magnetic Measurements. Thermal demagnetization  
137 was performed using an MMTD furnace, under argon atmosphere above 250 °C. For most samples, we  
138 measured the S ratio that is the IRM obtained after applying a 3 T field and then a back field of 0.3 T  
139 normalized to the IRM acquired in 3 T. Remanence measurements were performed with a SQUID cryogenic  
140 magnetometer (2G Enterprises, model 755R, with noise level of  $10^{-11} \text{Am}^2$ ). All magnetic measurements  
141 were performed at CEREGE (Aix-en-Provence, France), with the exception of MPMS measurements (at  
142 IPGP, Paris, France). The magnetic susceptibility of NWA 12563 was measured on samples weighing 13.7  
143 g and 160.2 mg. The magnetic properties for the CY chondrite Dhofar 2066 were measured on 2.69 g for  
144 susceptibility and 250 mg for MS.

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## 146 **2.3. Scanning Electron Microscopy**

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148 A sample of NWA 12563 was cut at the Institut de Minéralogie, de Physique des Matériaux, et de  
149 Cosmochimie (IMPMC), Muséum National d'Histoire Naturelle (MNHN), to prepare a polished section for  
150 study using reflected light microscopy, scanning electron microscopy (SEM), and electron probe micro  
151 analysis (EPMA). A back-scattered electron (BSE) map and images of selected regions were made at  
152 MNHN using a Tescan VEGA II LSU SEM in conventional mode (mainly 15keV,  $< 20\text{nA}$  and 15.4 mm  
153 working distance). Minerals were characterized on the MNHN SEM with an SD<sup>3</sup> (Bruker) EDS detector.

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155 Images were acquired at the Ecole Normale Supérieure Paris using a ZEISS ZIGMA field  
156 emission gun (FEG) SEM equipped with an Oxford Instrument X-MAX EDS detector. The accelerating  
157 voltage was 15 keV with a working distance of 9 to 10 mm (in backscattered mode). The Inca Software  
158 (Oxford Instruments) was used for qualitative analysis and mapping by dispersive energy spectra (EDS).  
159 For high resolution images we used 5 keV and a working distance between 2 and 3mm.

160

161 Hyperspectral maps and high resolution images of chondrite matrix were acquired using a FEG-  
162 SEM JEOL JSM-7800F LV at the University of Lille equipped with an EDX/EBSD Aztec system from  
Oxford Instruments and a silicon drift detector (SDD XMaxN) of 80 mm<sup>2</sup>. The sample has been studied

163 using hyperspectral maps with 1024 by 768 pixels corresponding to a region of 250  $\mu\text{m}$  by 192  $\mu\text{m}$ . A  
164 representative matrix region of 125  $\mu\text{m}$  by 96  $\mu\text{m}$  using a resolution of 512\*384 pixels is discussed below.  
165 A working distance of 10 mm and an energy range of 10 keV for 2048 channels (5ev per channel) were  
166 used. During acquisition, a drift correction was applied, based on the corresponding BSE image. The phase  
167 map was based on a simplified version of the ACADEMY methodology (Analyzing the Composition, the  
168 modal Abundance and the Density using Electron Microscopy; Zanetta et al., 2019) using XMapTools  
169 software (Lanari et al., 2014). We used a BSE image mosaic and the Qgis® software to estimate the  
170 chondrule and matrix proportions.

## 171 **2.4. Transmission Electron Microscopy**

172 We prepared six electron transparent sections by the focused ion beam (FIB) technique using an  
173 FEI Strata dual beam 235 at IEMN, University of Lille. The sizes of the FIB sections were typically ~200  
174  $\mu\text{m}^2$  and 150 nm thick. The FIB sections were studied with an FEI Tecnai G2-20 TWIN (LaB6, 200 kV)  
175 equipped with an EDAX energy dispersive spectrometer (EDS) and a Titan Themis, at 300 kV using a four  
176 quadrant, windowless, super-X SDD system. K-factors for O, Fe, Mg, Al, S, Ca, and K were experimentally  
177 acquired on standards (fayalite, forsterite, basalt and rhyolite, serpentine, wollastonite and pyrite).  
178 Compositional data were deduced from elemental mapping after post-acquisition processing of data cubes  
179 using Hyperspy (De la Peña, et al., 2017; Zanetta et al., 2019).

## 181 **2.5. Electron Microprobe Analysis**

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183 All quantitative mineral analyses were made by wavelength-dispersive spectrometry on the  
184 Cameca SXFive electron microprobe at the Université Paris VI, mainly using 15keV and 10 nA, with a  
185 focused beam or small rasters. We used silicate, oxide and phosphate standards, and monitored conditions  
186 with Astimex chlorite, San Carlos olivine and Eagle Station olivine internal standards.  $K_{\alpha}$  lines were  
187 measured on standards using diffracting crystals as follows: Na, albite LTAP; Mg olivine LTAP; Si  
188 olivine TAP; Al orthoclase LPET; K orthoclase LTAP; Ca, diopside LPET; Ti, MnTiO<sub>3</sub>; Cr, Cr<sub>2</sub>O<sub>3</sub> PET;  
189 Mn, MnTiO<sub>3</sub>LLIF; Fe, FeS<sub>2</sub> LLIF; F, apatite LTAP; P, apatite LPET; Cl, scapolite PET; F (sic) CaF<sub>2</sub> TAP;  
190 Ni NiO LLIF. The counting time was 10 sec for peaks and backgrounds, increased to 20 or 60 sec for  
191 some minor elements.

## 193 **2.6. Scanning Transmission X-ray Microscopy (STXM)**

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195 X-ray Absorption Near Edge Structure (XANES) data were collected using a scanning transmission  
196 X-ray microscope on the HERMES STXM beamline (Belkhou *et al.*, 2015; Hitchcock, 2017) at the  
197 synchrotron SOLEIL. Optical elements were exposed to a flow of O<sub>2</sub> to remove carbon contamination from  
198 the beamline. Energy calibration was performed using the 3p Rydberg peak of gaseous CO<sub>2</sub> at 294.96 eV.  
199 X-rays are focused on a 30 nm probe using a Fresnel zone plate. Alignment of images of stacks and  
200 extraction of XANES spectra were done using the aXis2000 software. We collected images at energy  
201 increments of 0.1 eV over the carbon absorption range (282–295 eV) with a dwell time of ~ 1 ms per pixel  
202 to prevent irradiation damage, and at energy increments of 0.2 eV over the Fe-L edge absorption range (705-  
203 713 eV). The C-XANES spectra were normalized to the carbon quantity, deconvolved, and quantified  
204 following the method developed by Le Guillou et al. (2018). The Fe<sup>3+</sup>/ $\Sigma$ Fe ratio was obtained based on the  
205 calibration developed by Bourdelle et al. (2013) and Le Guillou et al. (2015).

## 207 **2.7. Raman spectroscopy**

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209 Raman spectroscopy was used to document the degree of structural organization of the  
210 carbonaceous material. Raman data were obtained on a freshly fractured surface, at room temperature,

211 using a Renishaw INVIA spectrometer (IMPIC, Paris, France) in a confocal configuration equipped with  
212 a 514.5-nm wavelength 50-mW Modulaser Ar laser. The configuration used yields a horizontal resolution  
213 of  $\sim 1 \mu\text{m}$  for a laser power delivered at the sample surface below  $500 \mu\text{W}$ , thereby preventing irreversible  
214 thermal damage (Beysac et al., 2003; Bernard et al., 2008).

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## 216 **2.8 Hydrogen, nitrogen and carbon analysis**

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218 The measurements of the carbon and nitrogen concentrations and carbon isotopic composition  
219 (reported as  $\delta^{13}\text{C}$  calculated against the standard Pee Dee Belemnite with a  $^{13}\text{C}/^{12}\text{C}$  ratio of  
220 0.0112372) were performed on-line using the Thermo Scientific EA IsoLink - deltaV IRMS  
221 System at CRPG laboratory (Nancy, France) using a 2-mg aliquot of the meteorite powder. The  
222 precision of the measurement for nitrogen concentration, carbon concentration and isotopic  
223 composition are estimated to be 30 %, 2 %, and 2 ‰, respectively.

224 The measurements of the hydrogen concentration and isotopic composition were performed  
225 on-line using the Thermo Scientific EA IsoLink - deltaV IRMS System at CRPG laboratory  
226 (Nancy, France) according to the procedure detailed in Lupker et al. (2012). For hydrogen  
227 measurement, the meteorite was crushed into powder and two 2-mg-aliquots were loaded in tin  
228 capsules and degassed at  $120^\circ\text{C}$  under vacuum for 48h to minimize the contribution of adsorbed  
229 atmospheric water (Lupker et al., 2012; Vacher et al., 2016, 2020). The hydrogen isotopic  
230 composition is expressed as  $\delta\text{D}$  (‰) relative to Standard Mean Ocean Water (SMOW):  $\delta\text{D} =$   
231  $(\text{D}/\text{H}_{\text{sample}} / \text{D}/\text{H}_{\text{SMOW}} - 1) \times 1000$ , where SMOW represents the reference value of terrestrial oceans  
232 ( $\text{D}/\text{H}_{\text{SMOW}} = 155.76 \times 10^{-6}$ ). The reproducibility estimated on reference materials is better than 10%  
233 ( $2\sigma$ ) for the concentration of H and  $0.5 \times 10^{-6}$  for D/H (or 5‰ for  $\delta\text{D}$ ).

## 234 **2.9 X-ray computed tomography (X-CT)**

235 X-CT characterization was performed at the synchrotron X-ray beamline PSICHE at SOLEIL  
236 synchrotron (France). The measurement was performed with a parallel monochromatic beam of 25 keV  
237 and with an exposure time of 500 ms per each projection. A matrix sub volume with a volume of  $830 * 1500 * 700 \mu\text{m}$   
238 was studied in detail. We measured 5900 2D projections of the linear attenuation  
239 coefficient (LAC) to reconstruct the LAC variations in 3D, with a final voxel size of  $\sim 0.87 \mu\text{m}$ . Post-  
240 processing 3D reconstruction was achieved using the Python library Tomopy (Gursoy et al. 2014). The  
241 reconstruction was performed using PyHST2 (Mironne et al., 2014). Detection of pores and iron-rich  
242 phases was then performed thanks to a threshold segmentation (Dionnet et al. 2020), and the study of the  
243 structures' shape was conducted with Quant3D (Ketcham et al. 2005).

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## 245 **2.10 Visible-NIR micro-spectroscopy**

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247 Visible and near-IR diffuse reflectance micro-spectroscopy was performed in a clean room at  
248 IAS-Orsay (France), using a Vis-NIR (0.4-1.1  $\mu\text{m}$ ) grating spectrometer Maya2000 Pro (OceanOptics)  
249 with 4.5 nm spectral resolution coupled to a microscope (Leica Z16 APO). The sample is unilaterally  
250 illuminated at  $\sim 45^\circ$  incident angle by a 1000  $\mu\text{m}$  diameter fiber which is coupled to a halogen source. A  
251 collecting optical fiber is positioned orthogonally to the sample to collect the light diffused by the sample  
252 at  $0^\circ$  angle. By changing the magnification and/or the diameter of the collection fiber it is possible to  
253 adjust the collection spot down to about 7  $\mu\text{m}$ . We also measured spectra in a macroscopic configuration,  
254 using a collection spot of about 1 mm. In both cases, reference spectra were collected with respect to a  
255 99% Spectralon standard (from Labsphere). Spectra were measured at 20 different azimuth angles and  
256 averaged to take into account potential effects of the observation geometry. See Maupin et al. (2020) for  
257 more details.

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## 2.11 IR micro-spectroscopy

Mid-IR reflectance spectra and maps for polished sections of NWA 12563, Bells, and Essebi were collected at the SMIS beamline of the SOLEIL synchrotron (France) using an a Nicolet Continuum XL microscope (Thermo Fisher) equipped with a  $50 \times 50 \mu\text{m}^2$  liquid-nitrogen-cooled MCT/A detector (narrow-band,  $650 \text{ cm}^{-1}$  cut-off) and a  $32\times$  NA 0.65 Schwarzschild objective. The microscope was coupled to a Nicolet 5700 spectrometer (Thermo Fisher) equipped with a Michelson interferometer and a KBr beamsplitter. The IR source was the external synchrotron light source and the aperture was set to produce an analytical spot of 10-20  $\mu\text{m}$  on the focal plane. The spectral resolution was either  $4$  or  $8 \text{ cm}^{-1}$ , and at least 256 scans per spectrum were accumulated. Before each analysis, a background spectrum was acquired on a gold mirror standard. The average MIR spectrum of NWA 12563 was obtained by averaging more than 650 individual spectra obtained at different locations of the meteorite. All the spectra were taken from areas larger than  $500 \times 500 \mu\text{m}^2$ .

Additional MIR hyperspectral imaging data of NWA 12563, Bells, and Essebi were acquired with an Agilent Cary 670/620 micro-spectrometer equipped with a  $128 \times 128$  pixel Focal Plane Array detector ( $3900\text{-}800 \text{ cm}^{-1}$ ), installed at SMIS, using the internal globar source at  $8 \text{ cm}^{-1}$  spectral resolution. The IR maps were acquired in reflectance geometry, with respect to gold references, using a  $\times 15$  objective which produces a  $5.5 \mu\text{m}$  pixel size on the focal plane. More details about the setup are described by Brunetto et al. (2018) and Noun et al. (2019).

The far-IR (FIR,  $700\text{--}100 \text{ cm}^{-1}$  or  $14\text{--}100 \mu\text{m}$ ) spectra of NWA 12563, Bells, and Essebi were acquired at SMIS using a NicPlan microscope, coupled to an iS50 FTIR spectrometer (Thermo Fisher) operating in confocal reflection geometry, with respect to gold references, using a bolometer detector (boron doped silicon, 4.2 K cooled, Infrared Laboratories) and a solid-state Si beam splitter. The spectral resolution was  $4 \text{ cm}^{-1}$  and the spectral sampling  $0.5 \text{ cm}^{-1}$ . We mapped mm-sized areas using variable spot sizes of  $80\text{--}300 \mu\text{m}$  in diameter per spectrum. The various spectra were then averaged to finally produce average spectra representative of the meteorites. Thus, we averaged the small scale heterogeneity of the chosen chondrites. The bolometer also allowed us to acquire data in the  $750\text{--}1050 \text{ cm}^{-1}$  ( $13.3\text{--}9.52 \mu\text{m}$ ) range. Although noisy, this additional spectral range proved to be useful to properly combine spectral ranges obtained with different detectors. The FIR spectra were scaled to match the MIR spectra in the overlapping region, similarly to what was reported by Brunetto et al. (2020) for Tagish Lake, Murchison, Alais, Mighei, Paris, Allende, Lancé, and Frontier Mountain 95002 meteorites. In the case of NWA 12563, we also acquired additional MIR spectral maps using another Nicolet Continuum microscope at SMIS, equipped with a  $250 \mu\text{m}$  liquid-nitrogen-cooled MCT/B detector ( $450 \text{ cm}^{-1}$  cut-off) and a  $15\times$  Schwarzschild objective, to better combine the MIR and the FIR spectra, and to focus on the IR spectral signature of magnetite in one particular area of the meteorite.

The combined MIR-FIR spectra were finally baseline-corrected using a convex hull. The baseline correction was performed to focus on the position and shape of the IR spectral bands, and to allow comparison with emissivity IR remote sensing data of asteroids whose continuum is usually taken into account and removed in the thermal model of the spectral energy distribution (Emery et al., 2006).

## 3. RESULTS

### 3.1 Chondrite properties



307 *3.1.1 Petrography*

308 This chondrite with 14% chondrules supported by 86% fine grained matrix and accretionary  
309 chondrule rims is seen in Fig. 1a. The abundances were determined by using the image analysis program  
310 Qgis© and the term chondrule here includes complete chondrules, large chondrule fragments, and clusters  
311 or aggregates of phenocrysts (Fig. 1b). About 84% of the chondrules are Type I and about 16% Type II,  
312 while a large Al-rich chondrule is included with CAI and AOA in Fig. 1b as refractory material. Fig. 1b  
313 shows that only 46% of the chondrules have complete or partial fine-grained compact accretionary rims.  
314 The chondrule abundance is at the low end of the normal range for CM chondrites but the matrix includes  
315 isolated grains (tens of microns in size) likely derived from chondrules. Chondrule size ranges from 15  $\mu\text{m}$   
316 to 2 mm, with many smaller ones being fragments or disrupted loose crystals. Olivine (unaltered) is much  
317 more abundant than pyroxene, occurring both in Type I and II chondrules, and as crystal clasts in the matrix.  
318 Pyroxene (Mg-pigeonite and inverted protopyroxene) is found in some large chondrules. Amoeboid olivine  
319 aggregates and spinel-rich aggregates are rare.

320 The matrix is fine-grained (<100 nm to a few microns), with phyllosilicates, easily identifiable at  
321 the SEM scale by their fibrous nature, and amorphous silicate. It contains abundant magnetite (as framboids,  
322 spherules and plaquettes), plus pentlandite, troilite, sulfide-oxide aggregates, minor carbonate aggregates,  
323 but no scabby patches of tochilinite intergrown with cronstedtite. The matrix has a heterogeneous  
324 appearance (Fig. 1) with BSE intensity fluctuations due to the local variations in magnetite size distribution  
325 and abundance: bright areas have many small clusters of fine framboids while dark areas have scattered  
326 coarse framboid groupings. The matrix sample examined by X-CT with  $\sim 0.87 \mu\text{m}$  voxels contains a few  
327 crystal clasts up to 150  $\mu\text{m}$  in size and is very compact, with a very low porosity of  $0.7 \pm 0.5\%$  (Fig. S1a).  
328 The pores are anisotropic with average  $A = 5.6$ , the slight elongation meaning the meteorite has probably  
329 experienced weak shocks. The fractures are possibly related to shock or dehydration (Fig. S1b). At the scale  
330 of X-CT measurements, magnetite is fairly uniformly distributed in the three dimensions and its abundance  
331 in this chip is 5.8% (Fig. S1c).

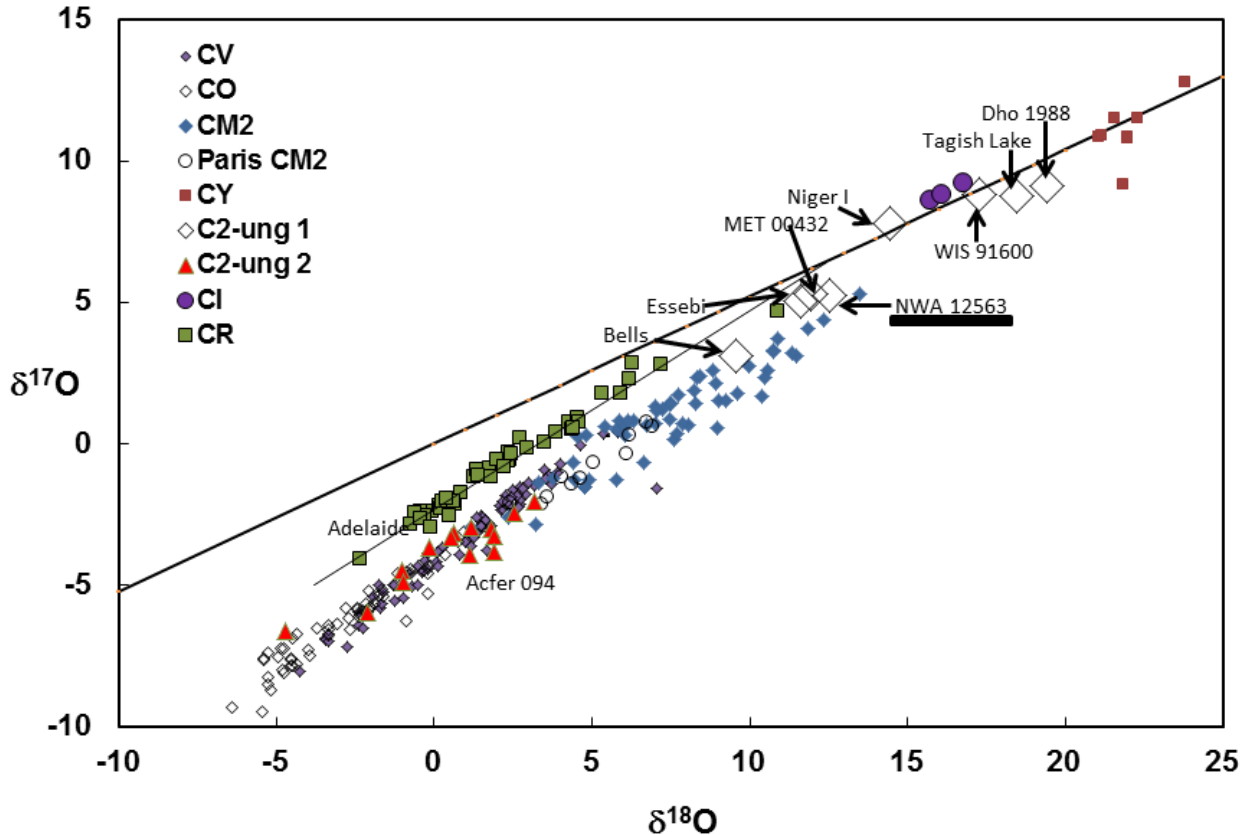
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333 Fig. 1. (a) BSE image of polished section of NWA 12563. Dark grey is magnesian olivine, medium grey is  
334 pyroxene and phyllosilicate, light grey is ferroan olivine, sulfide and magnetite, with a continuous grey  
335 serpentine-rich matrix. (b)The corresponding map is based on the BSE image.

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### 3.1.2 Oxygen Isotopes

The oxygen isotopic composition of NWA 12563 is  $\delta^{18}\text{O}=12.61\text{‰}$  and  $\delta^{17}\text{O}=5.26\text{‰}$ , with  $\Delta^{17}\text{O}$  of  $-1.33$  (linearized, slope 0.5247, analytical uncertainties 0.08‰, 0.12‰, 0.03‰ respectively). It plots at the upper end of the CO-CM trend line (Fig. 2), very close to the C2-ung Essebi (Rowe et al., 1994). We compare  $\delta^{18}\text{O}$  values, mineralogy and magnetic susceptibility with those of other chondrites of interest in section 7.2 and propose to call those with higher  $\delta^{18}\text{O}$  values C2-ung1 and those with lower  $\delta^{18}\text{O}$  values C2-ung2.



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Fig. 2. Oxygen isotopic composition of NWA 12563 compared to those of other carbonaceous chondrites (Greenwood et al, 2020). Ungrouped and anomalous C2 chondrites occur in two clusters separated by typical CM2 chondrites.

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### 3.1.3 Magnetic Properties

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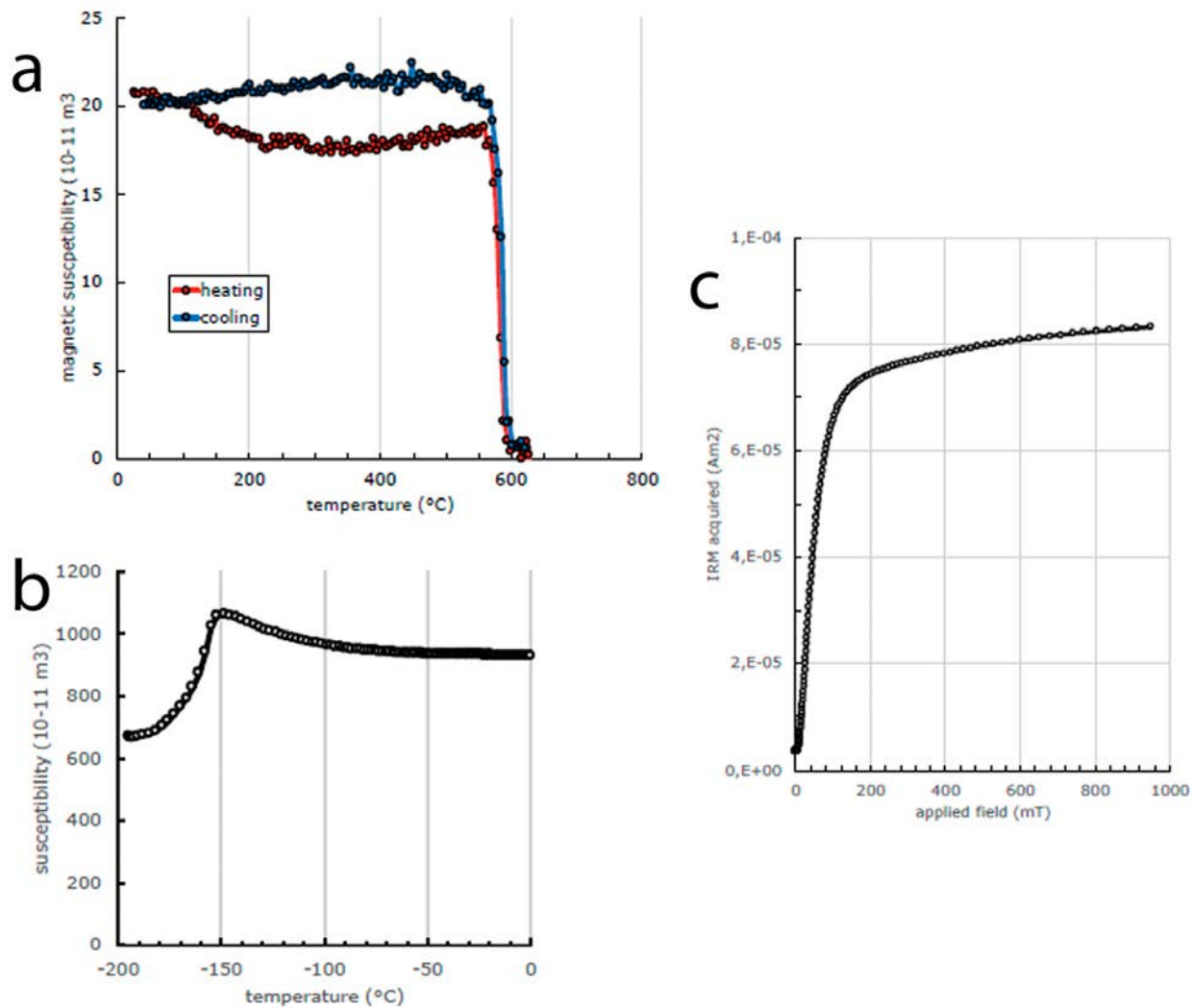
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Thermomagnetic measurements reveal a sharp and reversible magnetic transition at  $585^{\circ}\text{C}$  (Fig. 3a), which is the Curie temperature of magnetite. Magnetite is thus the ultra-dominant ferromagnetic mineral in this meteorite. The presence of magnetite is confirmed by the magnetic Verwey transition observed at  $-153^{\circ}\text{C}$  in the low temperature magnetic measurements (Fig. 3b). The magnetic susceptibility values of  $\log \chi = 4.67$  (where  $\chi$  is in  $10^{-9} \text{ m}^3 \cdot \text{kg}^{-1}$ ) indicate small scale homogeneous distribution of magnetite. This value is close to the values for Bells, Essebi, and Niger I C2-ung at 4.82, 4.85, and 4.89 respectively, and higher than all except 4 of 52 CM chondrites that give an average  $\log \chi = 3.90 \pm 0.43$  (Rochette et al., 2008). Saturation magnetization  $M_s$  is  $6.26 \text{ Am}^2/\text{kg}$ , corresponding to 6.80 wt% of magnetite. This is similar to the value for Niger I,  $M_s=9.85 \text{ Am}^2/\text{kg}$  that we measured on four samples totaling 2.55 g. For comparison, the

363 average  $M_S$  for CM chondrite falls is  $1.01 \pm 0.55 \text{ Am}^2/\text{kg}$  ( $N=9$ ), data from Cournède et al. (2015) and new  
 364 data from this study for Boriskino, Murchison, Pollen, Santa Cruz ( $M_S=1.76, 0.671, 0.234, 1.45 \text{ Am}^2/\text{kg}$ ,  
 365 measured on total masses of 31, 944, 20, 221 mg, respectively. The magnetite content of NWA 12563 is  
 366 thus more than five times higher than in the average CM chondrite.

367  
 368 In more detail, IRM acquisition is not completed around 300 mT (Fig. 3c), as should be the case if  
 369 magnetite was the only ferromagnetic mineral. IRM is still acquired up to 1 T (the maximum field used in  
 370 our experiments), which implies that a high coercivity ferromagnetic mineral is present in addition to  
 371 magnetite. This is confirmed by the  $S_{-300}$  ratio (ratio of SIRM superimposed with a back-field isothermal  
 372 remanent magnetization at 300 mT over SIRM) of -0.86, also indicative of the presence of a high-coercivity  
 373 mineral in addition to magnetite. Fe,Ni metal is unlikely because it usually has a low coercivity except in  
 374 the form of tetrataenite in the cloudy zone structure (Gattacceca et al., 2014), or as kamacite grains in dusty  
 375 olivines (Lappe et al. 2011)), none of which are observed in NWA 12563, and also because the magnetic  
 376 susceptibility is negligible above  $585^\circ\text{C}$  (Fig. 3a). Pyrrhotite is a candidate because it has much lower  
 377 susceptibility than magnetite so a minor pyrrhotite abundance compared to magnetite would go totally  
 378 unnoticed in the thermomagnetic experiments.



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382 Fig. 3. (a) Thermomagnetic measurements show the presence of magnetite by the magnetic transition at  
 383 585°C, its Curie temperature. (b) The magnetic Verwey transition observed at -153°C confirms the presence  
 384 of magnetite. (c) IRM acquisition is incomplete at 300 mT (Fig. 3c), probably due to pyrrhotite.

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### 386 3.1.3 Hydrogen, nitrogen, and carbon

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388 The carbon concentration of NWA 12563 is 2.13 wt.% C, with a  $\delta^{13}\text{C}$  of -12.2 ‰, and the nitrogen  
 389 content is 0.095 wt.% N. The hydrogen concentrations measured for the two aliquots after degassing at  
 390 120°C correspond to 0.73 and 0.72 wt.%, equivalent to 6.6 and 6.5 wt.% H<sub>2</sub>O, with an isotopic composition  
 391  $\delta\text{D}$  of -57.7 and -57.6 ‰, respectively. These values are typical of normal CM chondrites (Alexander et al.,  
 392 2012; Vacher et al., 2016; Vacher et al., 2020), although the hydrogen concentrations are in the lower end  
 393 of the concentration range measured for CM chondrites (from ~5 to 13 wt.% H<sub>2</sub>O). The hydrogen  
 394 concentrations and C/H ratios (C/H at.% = 0.24) measured for NWA 12563 are close to the one measured  
 395 for the ungrouped chondrites Tagish lake, Bells and Essebi (5.0 to 8.6 wt.% H<sub>2</sub>O and C/H at.% from 0.15  
 396 to 0.45; Alexander et al., 2012). These values might also resemble to the one of heated CM-chondrites  
 397 (Alexander et al., 2012), though our studies of matrix (below) show no evidence of dehydration.

## 398 3.2 High temperature materials

### 399 3.2.1 Refractory Inclusions

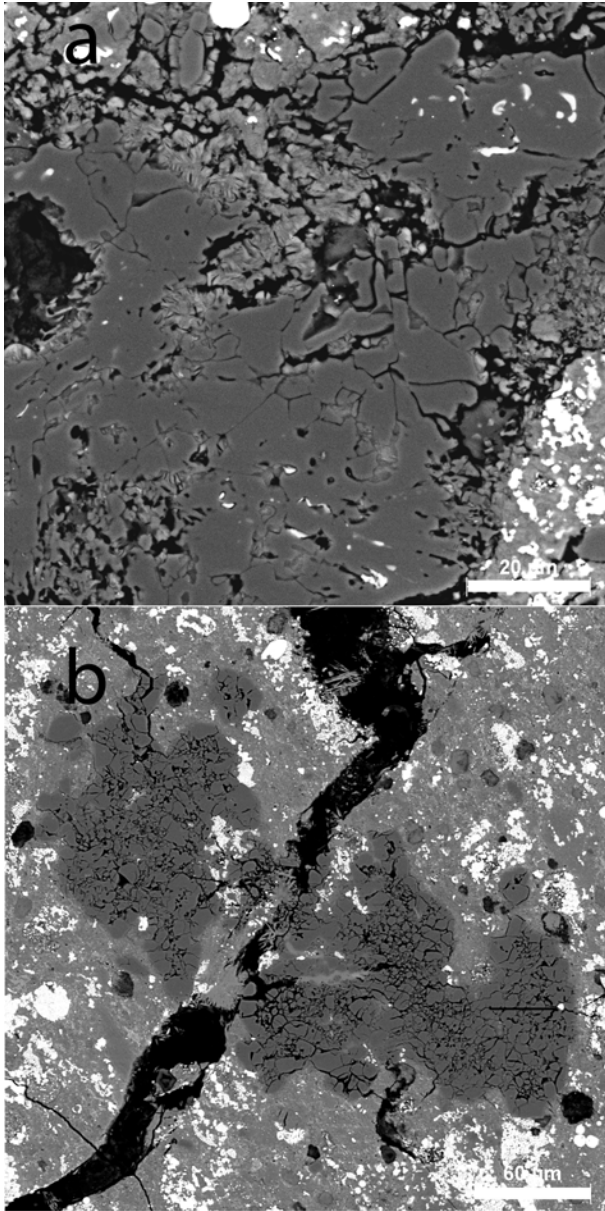
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401 Refractory inclusions are rare (~0.2%). They include spinel-rich inclusions in which other primary  
 402 phases have been destroyed, except for accessory perovskite (Fig. 4a), and amoeboid olivine aggregates  
 403 (AOA) with accessory Ni-rich schreibersite (Fig. 4b). The olivine in AOA overlaps in composition with  
 404 olivine in chondrules and isolated grains, but Fig. 5 shows that it is closer on average to pure forsterite in  
 405 minor elements as well as Fe and Mg (Fo<sub>99.3±0.7</sub> vs Fo<sub>98.9±0.7</sub>). Its concentrations of Fe and minor elements  
 406 resemble those of AOA olivine in primitive chondrites like Acfer 094 (Krot et al., 2004). The  
 407 compositions of anhydrous silicates are given in Tables 1 and S1.

Table 1. Analyses of olivine and pyroxene from AOA, chondrules, and isolated in matrix.

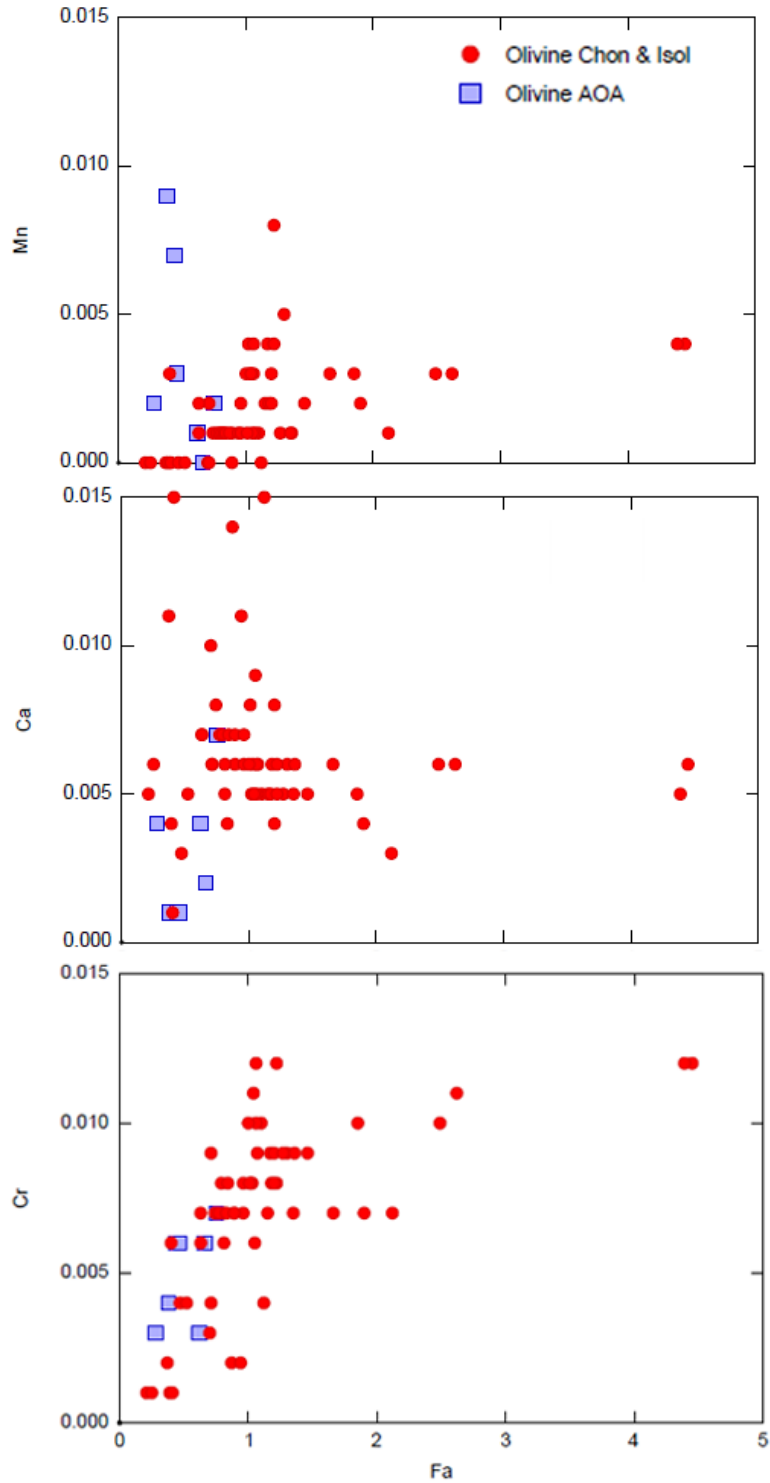
Point	Object	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	TiO <sub>2</sub>	Cr <sub>2</sub> O <sub>3</sub>	FeO	MnO	MgO	CaO	Na <sub>2</sub> O	Total	Fo/En	Fa/Fs	Wo
55	AOA	42.36	0.05	0.03	0.14	0.29	0.08	57.45	0.17	0.03	100.59	99.72	0.28	
32	AOA	43.67	0.04	bd	0.34	0.47	0.14	57.24	0.05	bd	101.96	99.54	0.46	
33	Al-rich	43.66	0.05	0.04	0.38	0.81	0.05	55.88	0.20	bd	100.85	99.19	0.81	
60	IA BO	43.18	0.12	0.08	0.10	0.37	bd	57.15	0.43	bd	101.68	99.63	0.37	
58	isolated	42.54	0.06	0.04	0.18	0.71	bd	56.52	0.42	0.02	100.47	99.30	0.70	
68	IA ch 63	42.90	0.05	0.05	0.51	1.32	0.26	56.01	0.23	bd	101.33	98.70	1.30	
51	IIA PO 51	40.57	0.05	0.03	0.40	13.16	0.26	45.78	0.13	bd	100.50	86.12	13.89	
58	IIA BO	38.84	0.06	0.01	0.42	20.71	0.27	39.78	0.19	0.01	100.49	77.40	22.60	
59	isolated	37.31	0.01	bd	0.43	28.90	0.38	33.58	0.16	bd	100.80	67.44	32.56	
52	IIA PO 51	35.49	bd	0.02	0.34	36.47	0.29	27.28	0.20	0.02	100.13	57.15	42.86	
64	isolated	34.86	0.04	bd	0.43	42.24	0.41	21.83	0.25	bd	100.49	47.95	52.05	
31	Al-rich	59.10	2.05	0.44	0.73	0.48	0.11	35.26	3.55	bd	101.84	92.60	0.71	6.70
109	IB	59.90	0.55	0.14	0.76	1.01	0.10	38.84	0.51	bd	101.85	97.66	1.42	0.92
162	IAB 158	58.23	1.03	0.15	0.59	1.61	0.03	37.94	0.47	0.07	100.14	96.83	2.31	0.86

408



409

410 Fig. 4. (a) Spinel-rich refractory inclusion with abundant pores containing phyllosilicate, and accessory  
411 perovskite (white). (b) Amoeboid olivine aggregate with accessory Ni-rich schreibersite (arrow). Both  
412 objects are embedded in magnetite-rich matrix.

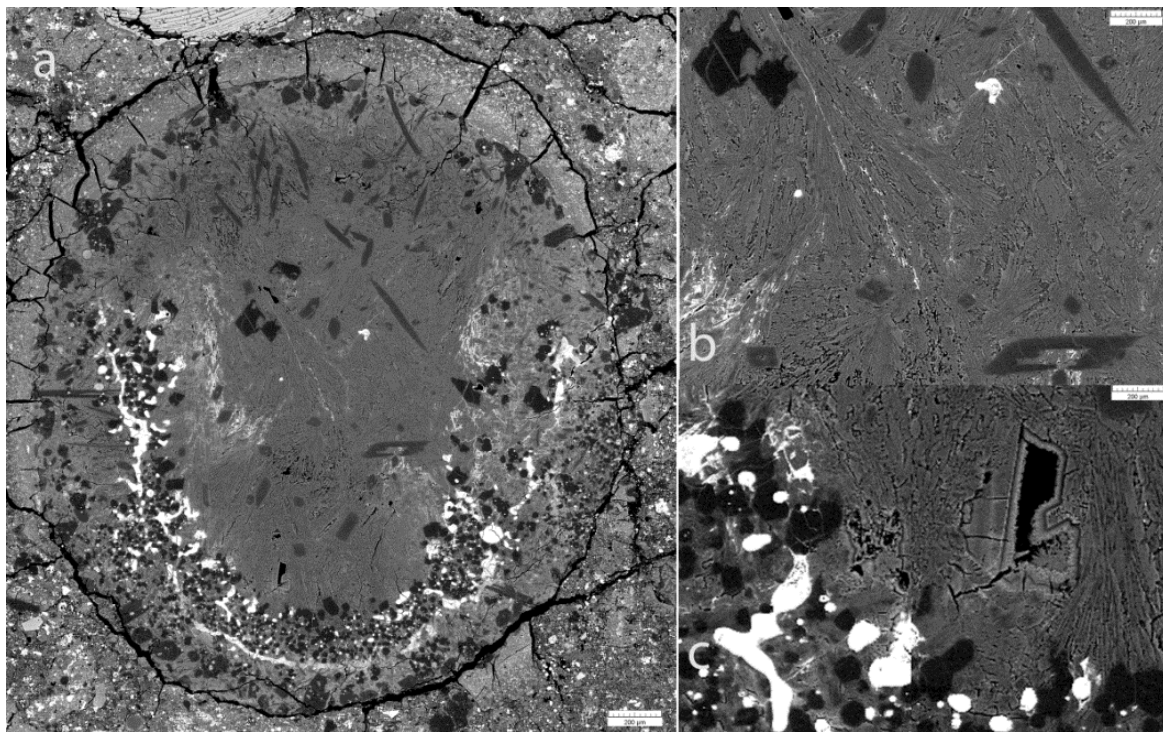


413  
 414 Fig. 5. Concentrations of Mn, Ca, and Cr in olivine in a.f.u. Olivine in AOA is closer to pure forsterite on  
 415 average than that in chondrules (chon) and isolated (isol) in matrix.  
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418 3.2.2 Composite Al-rich chondrule

419 NWA 12563 contains one chondrule-like object (Fig. 6a) zoned with a core, a mantle with olivine  
420 and metal, and an igneous rim (in addition to the matrix-like fine-grained accretionary rim). We describe  
421 this chondrule as composite and Al-rich to call attention to the exceptional core that consists of a Ca- and  
422 Al-rich groundmass of plumose and acicular anorthite and diopside crystals (Fig. 6b). There are bladed  
423 and elongate hopper microphenocrysts of Ca-poor pyroxene, a magnesian pigeonite ( $\text{En}_{90-94}\text{Fs}_{1-4}\text{Wo}_{4-7}$ ),  
424 occurring mainly in this core and the igneous rim. The mantle is discontinuous and resembles metal-rich  
425 microporphyritic Type IAM chondrule material with forsterite ( $\text{Fo}_{99.1 \pm 0.2}$ ). Where the mantle is absent  
426 there are a few forsterite hopper crystals, as well as pigeonite set in the groundmass of the adjacent core.  
427 The igneous rim resembles Type IAB chondrules. Alteration is confined mainly to the rim and metal in  
428 the mantle, particularly in the continuous ring segment. A vug in the core contains amphibole growing  
429 inward, as well as some calcite whiskers (Fig. 6c). The Al-Ca-rich plumose core groundmass is similar to  
430 crystalline mesostasis in a Type I chondrule in CR3 chondrite QUE 99177 (Abreu and Brearley, 2010). Its  
431 texture is similar to those of rapidly cooled synthetic Al-rich chondrules (Tronche et al., 2007).



432  
433 Fig. 6. BSE images of refractory chondrule. (a) Whole chondrule with core, mantle, igneous rim, and fine-  
434 grained rim. (b) Core of plumose anorthite and diopside crystals with elongate and hopper pigeonite  
435 microphenocrysts. (c) Amphibole-lined vug in core (center-right) and Type IAM chondrule mantle with  
436 unaltered metal (bottom left).

437 3.2.3 Chondrules

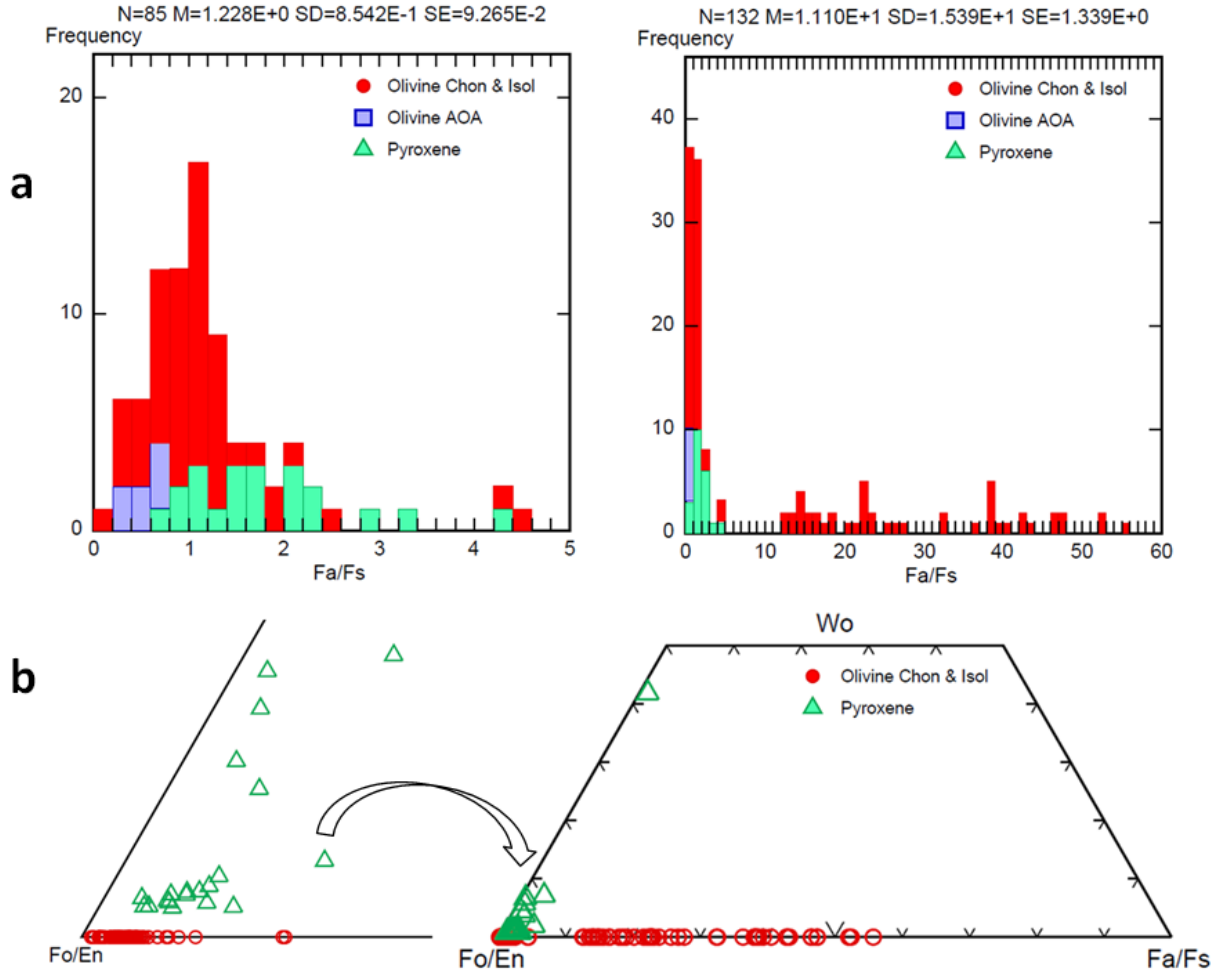
438  
439 The chondrules are mainly Type IA (PO and BO) and IAB, including originally metal-rich  
440 chondrules, and Type IIA (PO and BO, with relict grains and minor chromite). They are illustrated in Fig.  
441 7. Pyroxene-rich chondrule fragments are rare, and olivine is much more abundant than pyroxene in the



442 meteorite, as indicated by the histograms (Fig. 8a). Similarly, Type I chondrules and chondrule debris are  
443 more abundant (84%) than Type II material (16%).

444

445 Fig. 7. BSE images of chondrules. (a) Type IA porphyritic olivine chondrule with rare metal inside  
446 olivine. (b) Poikilitic Type IAB chondrule: ol olivine, op orthopyroxene, Di diopside, ps pseudomorph  
447 after metal. (c) Type IIA porphyritic olivine. (d) Type IIA barred olivine.



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 449  
 450 Fig. 8. (a) Histograms of olivine (in AOA and Types I and II chondrules) and pyroxene compositions. (b)  
 451 Olivine and pyroxene compositions. Detail of Type I olivine and pyroxene at left in both (a) and (b).

452 The forsteritic olivine of Type I chondrules has an average composition of  $Fo_{98.9 \pm 0.7}$  with Fe correlating  
 453 with Cr and Mn (Fig. 5). Type IAB chondrule and isolated pyroxene in NWA 12563 are generally  $En_{98-}$   
 454  $_{94}Fs_{1-4}Wo_{1-2}$  though there is minor interstitial diopside ( $En_{57}Fs_1Wo_{42}$ ) in some chondrules (Fig. 7b). Type  
 455 Type II olivine has a composition range of  $Fo_{84-44}$  and an average FeO/MnO of 90, typical of carbonaceous  
 456 chondrites (Berlin et al., 2011; Hewins et al., 2014). The standard deviation of wt. %  $Cr_2O_3$  in olivine is  
 457 lower than that in the CO 3.0 ALH 77307 (Fig. S2), placing it at 3.0 on the scale of Grossman and  
 458 Brearley (2005). Selected mineral compositions are given in Table 1.

459

### 460 3.3 Chondrule deformation and alteration

461 A few chondrules survive as complete objects with rims, but the majority are fragmented and/or  
462 deformed with many unusual shapes. In addition to hydrous replacement of many phases, especially glass  
463 (Fig.9a), many Type II barred olivine chondrules display fractures and kinking (Fig. S.3), and there is a  
464 kind of disbarring where olivine bar segments are scattered into adjacent matrix (Fig. 9b). Some Type I  
465 chondrules have highly irregular, gerrymandered shapes (Fig. 9c) in part caused by selective alteration of  
466 specific phases, probably metal, eating away at their borders. Similar embayments are observed in Tagish  
467 Lake (Takayama et al., 2012). Clusters of ferroan olivine crystals are interpreted as defunct Type II  
468 porphyritic chondrules that suffered replacement of mesostasis by a matrix-like assemblage with  
469 concentrations of phosphate (Fig. 9d).

470

471 Fig. 9. BSE images of chondrules. (a) Type IIA BO, with interstitial glass replaced by phyllosilicates. (b)  
472 Disbarred Type II BO, fragmented, kinked with bar fragments dispersed in matrix (center bottom). (c)  
473 Gerrymandered Type IA chondrule; embayments (e.g. dashed line) caused by replacement of least stable  
474 material. (d) A defunct Type IIA chondrule, with a cluster of phenocrysts and microphenocrysts in matrix-  
475 like altered mesostasis.

476 Though olivine in chondrules and matrix is essentially unaltered, pyroxene in chondrules shows  
477 incipient alteration. Kamacite and Ca-Al-rich glass survive only as rare inclusions in the forsterite of Type

478 I chondrules. The dominant chondrule alteration phases are fibrous to lathy Mg-Si-rich and Fe-Al-rich  
479 phyllosilicates visible in BSE as mixtures or intergrowths interstitial to microphenocrysts, i.e. replacing  
480 mesostasis (Fig. 10a,b). Pyroxene alteration in chondrules occurs along etched cleavage planes marked  
481 with voids and some phyllosilicate (Fig. 10c). Ovoid inclusions in olivine and particularly pyroxene on the  
482 margins of Type I chondrules (as shown in Fig. 10b,c,d,e). They have a form like that of large metal  
483 grains and we interpret them as pseudomorphs after kamacite, as in Wisconsin 9600 and Tagish Lake  
484 (Brearley, 2004; Takayama et al., 2012). There is a continuum (of smooth clear glassy-looking serpentine  
485 ovoids Fig. 10, particularly d), Si-poor ovoids speckled with high-Z material including sulfides, and  
486 manifestly altered, sometimes two-phase, Fe-rich nodules (Fig. 10f).

487  
488 Fig. 10. BSE images of alteration in chondrules. (a) Type IA rich in altered metal and with  
489 phyllosilicates replacing mesostasis. (b) Metal-poor Type I chondrule with two-phase phyllosilicate (dark  
490 grey serpentine-saponite and light grey more ferroan chlorite). (c) Type IAB with fresh metal only in  
491 olivine at top left; phyllosilicates replace mesostasis and pseudomorph metal. (d) Metal pseudomorphs  
492 within pyroxene in a Type IAB chondrule composed of clear serpentine or serpentine speckled with  
493 FeNiCrPS phases. (e) Type IAB with phyllosilicate in etched pyroxene. (f) Type IA chondrule with

494 silicate contrast set to black; metal pseudomorphs are two-phase, Si-rich and S-rich respectively. Ol:  
 495 olivine, PX: pyroxene, PH: phyllosilicate, AK: altered kamacite, and psd: pseudomorph.

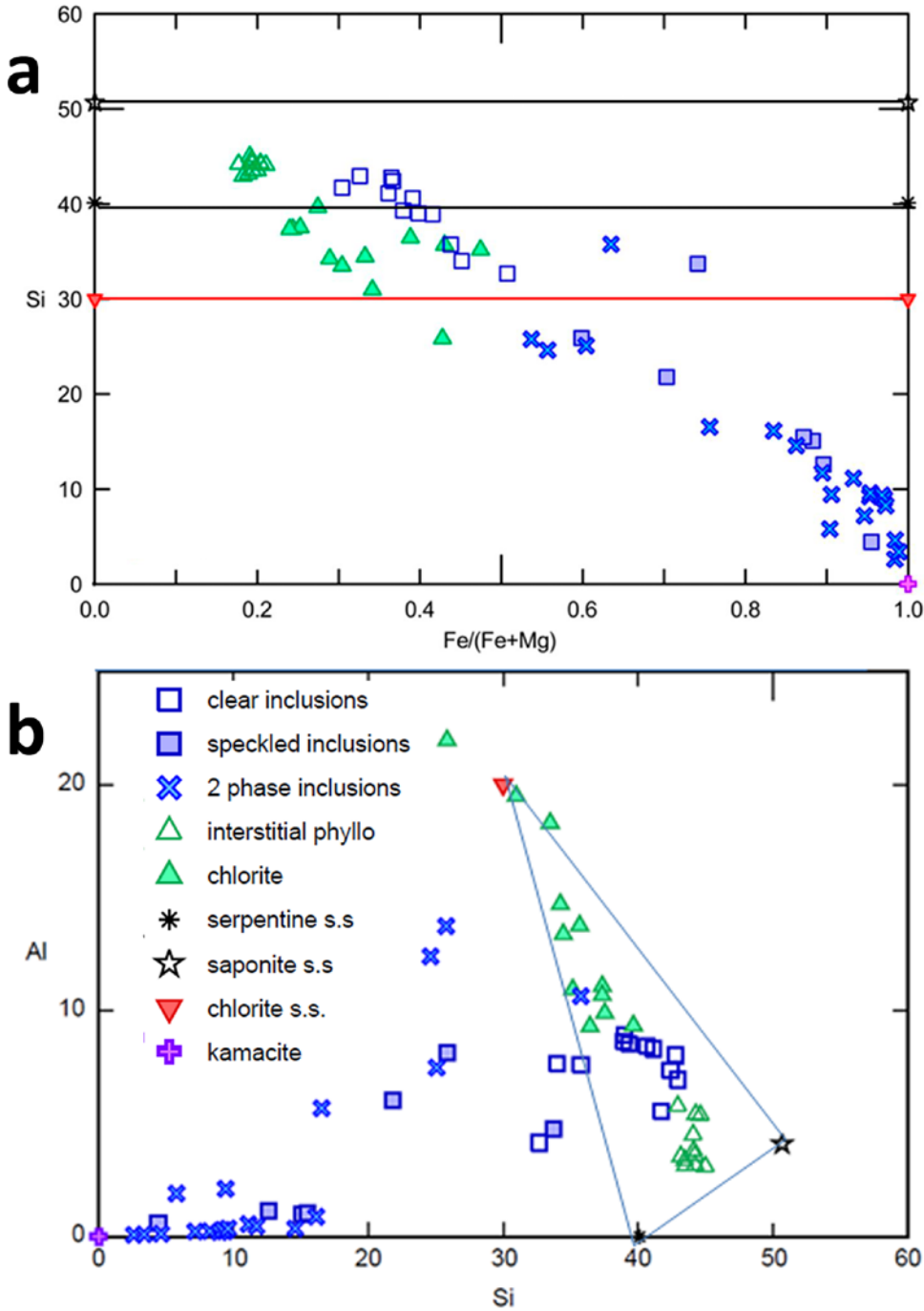
496 Analyses of alteration phases found in chondrules are given in Table 2. The two phases seen in  
 497 BSE images of mesostasis are called serpentine and chlorite here; altered metal ovoids consist of  
 498 assemblages of phyllosilicates and Fe-rich phases distinguished in Table 2 by the texture type. Some  
 499 analyses do not represent pure phases, as there may be inclusions of sulfides and other minerals, and  
 500 mixtures of phyllosilicates at the one micron scale. A secondary amorphous silicate phase does not seem  
 501 probable for the fibrous and lathy material in the chondrule mesostasis, but it cannot be ruled out for the  
 502 smooth ovoids.

503 Table 2. Analyses of alteration phases in mesostasis and ovoid inclusions replacing metal in chondrules.

Type	Sample	SiO2	Al2O3	TiO2	Cr2O3	FeO	MnO	MgO	CaO	Na2O	K2O	P2O5	S	Ni	Total
serpentine	134	45.02	2.74	0.09	0.43	11.86	0.17	27.73	0.04	0.46	0.06	0.02	0.31	0.06	88.99
	135	44.31	3.23	0.15	0.37	12.20	0.09	26.61	0.14	0.49	0.04	0.01	0.39	0.05	88.08
	42	44.27	3.85	0.07	1.43	12.03	0.04	25.27	0.33	0.79	0.25	0.03	0.16	0.11	88.64
	41	45.10	4.60	0.09	1.23	10.64	0.02	24.98	0.51	1.06	0.29	0.10	0.10	0.03	88.75
chlorite	17	40.63	8.12	0.21	1.01	15.91	0.13	23.59	0.30	0.84	0.16	0.07	0.25	bd	91.22
	26	37.90	9.54	0.32	0.90	14.28	0.08	24.83	0.12	0.60	0.14	0.23	0.24	0.03	89.21
	24	36.27	11.89	0.22	0.49	23.83	0.13	17.70	0.21	1.68	0.18	0.14	0.25	0.02	93.00
	20	31.96	17.08	0.27	0.40	18.58	0.27	20.13	1.63	0.69	0.09	0.51	0.33	0.02	91.96
ovoid (clear)	15	39.29	6.25	0.17	1.72	18.40	0.10	17.99	0.12	0.37	0.14	0.05	0.14	0.04	84.78
	14	36.84	6.50	0.42	3.14	19.54	0.20	17.07	0.16	0.19	0.11	0.11	0.29	0.04	84.60
	12	34.79	6.40	0.51	2.52	18.73	0.17	17.17	0.55	0.26	0.10	0.12	0.39	0.19	81.90
	33	29.46	5.30	0.10	4.30	20.46	0.08	14.73	1.20	0.08	0.07	0.28	0.42	1.25	77.73
ovoid (speckled)	32	29.37	3.54	0.29	5.86	34.58	0.08	6.76	2.20	0.22	0.13	0.49	1.54	2.88	87.94
	37	21.81	5.82	0.08	6.19	30.19	bd	11.35	1.31	0.26	0.13	0.44	1.80	2.18	81.56
	22	13.09	0.75	1.46	4.54	60.07	0.48	4.47	0.79	0.19	0.02	0.45	3.68	1.94	91.92
	36	10.10	0.78	1.49	3.68	51.70	0.38	3.37	1.47	0.17	0.07	0.83	5.06	3.37	82.47
ovoid (2-phase)	45/1	29.50	7.45	0.18	1.13	27.96	0.40	9.02	0.61	0.53	0.29	0.33	1.43	0.67	79.50
	36/1	2.81	0.08	0.03	1.67	81.37	0.29	0.52	0.22	0.11	bd	0.53	2.64	3.36	93.61
	47/1	5.71	1.60	0.05	0.42	53.68	0.03	3.20	0.23	0.11	0.03	0.25	15.88	10.31	91.50

504

505 Understanding of chondrite phyllosilicate compositions involves the roles of variations in  
 506 tetrahedral-octahedral layer combinations (serpentine vs. saponite, and interlayer cations), ionic  
 507 substitutions (especially Al), and the presence of inclusions (especially of sulfide) or mixed layer  
 508 structures. Charge balancing of Al in phyllosilicate is complex (Wiewora and Weiss, 1983) and here we  
 509 use the  $M_6Si_4$  serpentine endmembers chrysotile-greenalite, where  $M = Mg, Fe^{2+}$ , and the  $M_5AlSi_3Al$   
 510 chlorite midmembers clinocllore-chamosite to evaluate the identity of the phases. Analyses are plotted  
 511 along with serpentine, chlorite and saponite endmembers in Fig. 11. All the phyllosilicate analyses free of  
 512 obvious high-Z material plot between serpentine and saponite tie lines, with some analyses near the  
 513 chlorite tie line (Fig. 11a). A large range of Al contents is seen in Fig. 11b: most of the phyllosilicate  
 514 compositions (except ovoid pseudomorphs with high-Z inclusions) fall in a triangle bounded by  
 515 endmember serpentine, and mid-member chlorite and saponite compositions. Those with moderate Al



516

517 Fig. 11. Compositions of chondrule mesostasis (green) and metal alteration phases (blue) in atomic  
 518 percent (excluding O) compared to reference s.s. (solid solution) serpentine, saponite, chlorite  
 519 (clinocllore) and kamacite compositions. (a) Ovoid inclusion material ranges from compositions near  
 520 kamacite to near chlorite and serpentine. (b) mesostasis phyllosilicates are magnesian serpentine-saponite  
 521 mixtures or intergrowths and a ferroan chlorite. The distribution within the three-phase triangle suggests  
 522 that chlorite is a mixture.

523 contents, with a maximum near clinochlore and a minimum near penninite (not shown), and with roughly  
524 equal quantities of <sup>IV</sup>Al and <sup>VI</sup>Al, are designated as chlorite. We cannot show that those with a  
525 composition near penninite are not mixtures of clinochlore-like chlorite and serpentine, or a mixed-layer  
526 phase, but compositions near clinochlore exist. The magnesian serpentine-like phyllosilicate is quite  
527 tightly clustered in composition (Fig. 11a,b open triangles) with some coupled Al substitution (Fig 11c) as  
528 in chlorite. However, chlorite is more ferroan than the serpentine-saponite and has a wide range of Fe/Mg  
529 ratios (Fig. 11a). The correlation of Fe/Mg is positive with Al and negative with Si, for both interstitial  
530 chlorite and ovoid inclusions. The phyllosilicate with lower Al contents falls close to a tie line between  
531 Mg-rich serpentine and saponite (Fig. 11b). In general, it has an octahedral to tetrahedral cation ratio  
532 between 3:2 and 3:4, corresponding on average to serpentine with about 45% saponite.

533           There is a continuum of compositions for two-phase, speckled, and clear ovoid inclusions (Fig. 10  
534 f, d, c, respectively) between metal and phyllosilicate replacing mesostasis, particularly chlorite (Fig. 11,  
535 12). Fe and Ni concentrations are naturally lower in the silicate inclusions than in reference kamacite, but  
536 P, S and Cr are higher, especially in the ovoids speckled with inclusions. Clear ovoids are close in  
537 composition to chlorite, except for higher Cr, lower Al (Fig. 11, 12) and lower P. The least altered ovoids  
538 appear in BSE to be two-phase mixtures (Fig. 10f). These ‘phases’ are dominantly Si-rich (phyllosilicate)  
539 and dominantly Fe- or S-rich, i.e. in reality three-component mixes. The Fe-rich phase could be residual  
540 kamacite, magnetite or a hydroxide; the S-rich component is one or more sulfides, as tochilinite (Tomeoka  
541 and Buseck, 1985) is eliminated by the low Mg content of the replacing high-Z phases (Fig. 11a). The  
542 “two-phase” pseudomorphs have Ni, Cr and P contents higher than those of the speckled inclusions.

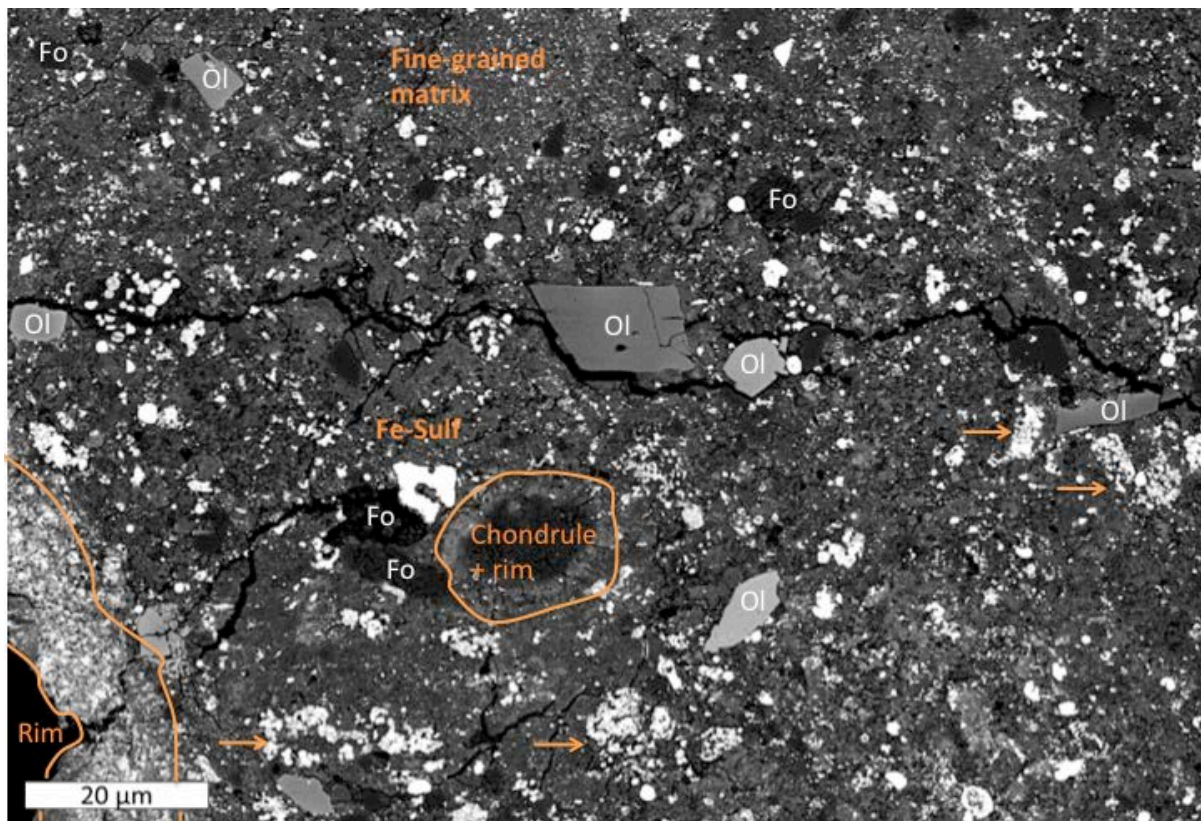
544 Fig. 12. Minor element concentrations of ovoid inclusions pseudomorphing metal in atomic percent  
545 (excluding O) compared to mesostasis phyllosilicates and a reference kamacite composition.  
546 Concentrations of S, Cr, and P are higher than for kamacite, even for clear ovoids whose composition is  
547 close to chlorite for major elements.

548

### 549 3.4 Matrix and rims, mineralogy and alteration

#### 550 3.4.1 Overview of matrix

551



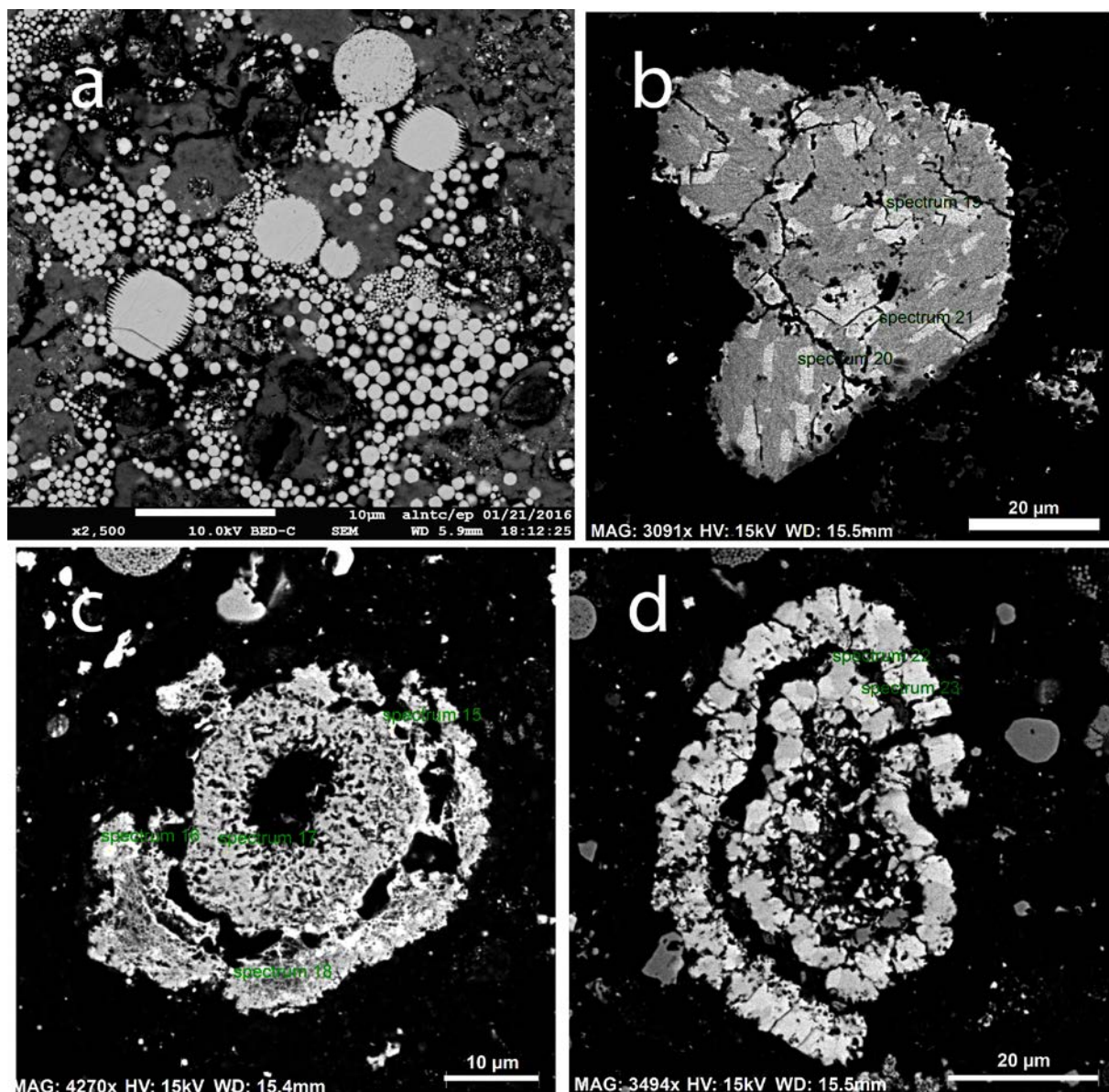
552

553 Fig. 13. BSE image of a representative area of the matrix of NWA 12563. The different minerals are Ol  
554 (olivine with various Fe contents), Fo (forsterite), and Fe-sulf (Fe-rich sulfide). The orange arrows indicate  
555 the presence of framboidal magnetite aggregates.

556 A general view of the matrix of NWA 12563 is shown in Fig. 13. The grain size ranges from <100  
557 nm to 20 μm, but some grains are larger, such as patches of carbonate, and type I and type II chondrule  
558 fragments. The type I chondrule fragments are mainly pure forsterite which sometimes contain preserved  
559 metal grains as inclusions. The type II fragments have sharp outlines and generally show Mg/Fe zonation  
560 in olivine with a more Mg-rich core. The pyroxene grains are anhedral, as in the chondrules, and show  
561 chemical zonation, from a stoichiometric pyroxene in the center of the grain to more oxygen-rich towards  
562 the edges. EPMA data do not reveal the presence of the tochilinite-cronstedtite intergrowths usually  
563 observed in CMs and other altered carbonaceous chondrites.



564 A striking feature of the matrix is the occurrence of sulfides and magnetite: both occur as small  
 565 grains, embedded within the amorphous silicates/phylosilicate, and also as scattered rosette-like aggregates.  
 566 The particle size of the matrix-embedded magnetite is very well constrained since it varies between 100 nm  
 567 and a few  $\mu\text{m}$ , with rare grains 10-20  $\mu\text{m}$  in size. Magnetite mainly occurs as framboidal and spheroidal  
 568 aggregates, but also as plaquettes (Fig. 14a). Its composition is  $\text{Fe}_3\text{O}_4$  and no other elements are present  
 569 above the detection limits. There is also magnetite occurring as a fine meshwork within the sulfide rosettes  
 570 (Fig. 14c,d). A bulk magnetite content of 6.80 wt. % was estimated based on the magnetic properties. This  
 571 translates into 2.94 vol% assuming a CM2 bulk density of 2.25 for NWA 12653 (Consolmagno et al., 2008).

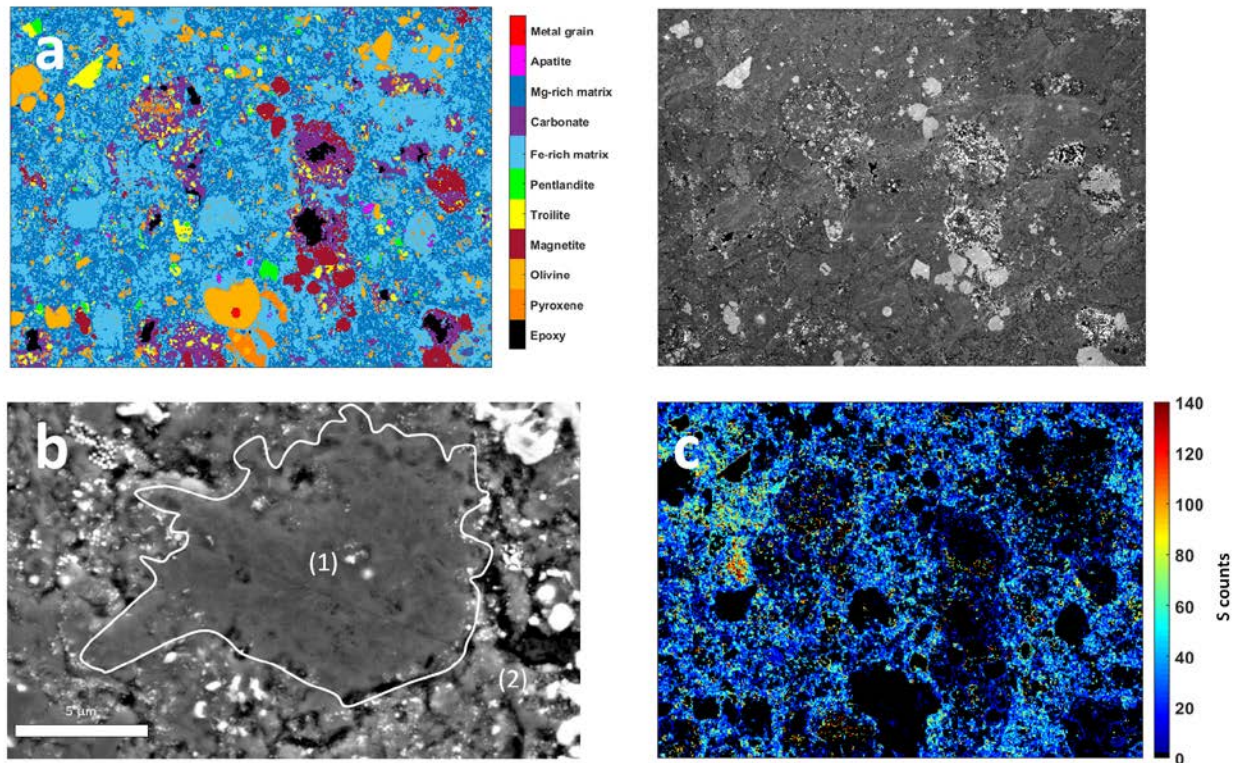


572  
 573 Fig. 14. BSE images of fine-grained magnetite and sulfide in matrix. (a) Framboidal and plaquette  
 574 magnetite. (b) Troilite with exsolved pentlandite. (c,d) Rosette aggregates of pentlandite, troilite and  
 575 magnetite.

576 Pentlandite is the main sulfide mineral in the matrix, occurring as isolated or clustered tiny laths, as  
 577 rosettes, and rarely as exsolution lamellae within troilite (Fig. 14b). The rosette or flower structures are  
 578 mainly double rings. They contain pentlandite, troilite and magnetite (Fig. 15c,d). Pentlandite  
 579 compositions plot on the linear field for pentlandite in the 200°C section of the Fe-Ni- S system,  
 580 reconstructed using reference compositions from Craig (1973) and Harries and Langenhorst (2013). Their  
 581 most Fe-rich compositions are in equilibrium with the pyrrhotite near stoichiometric troilite (Fig. S4).  
 582 Magnetite occurs in a fine meshwork with sulfide (Fig. 14c), the lacey texture suggesting replacement of  
 583 either phase by the other.

### 584 3.4.2 Matrix mineralogy using ACADEMY

585 We used the ACADEMY method (Zanetta et al., 2019) to characterize matrix. This method allows  
 586 us to establish phase maps as well as modal abundances using low voltage SEM and then to quantify the  
 587 mapped area using EPMA.  
 588



589 Fig. 15. (a) Phase map (125 μm\*96 μm, pixel size ~ 250 nm) obtained using the ACADEMY  
 590 methodology (Zanetta et al., 2019) and corresponding secondary electron image. It reveals in particular  
 591 two types of fine-grained matrix material (different blues) with different texture and composition. (b)  
 592 High resolution BSE image of these two types of fine-grained material. (1) is a homogeneous and fibrous  
 593 phyllosilicate and contains few inclusions. (2) contains amorphous silicate richer in nanosulfides and is  
 594 more porous. (c) X-ray map showing the localization of S-rich matrix near some sulfide grains seen in (a).  
 595  
 596

597 The phase map (Fig. 15a) reveals that the matrix of NWA 12563 is dominated by a groundmass of  
 598 amorphous silicate and phyllosilicate. Sulfides, oxides, anhydrous silicates and carbonates are embedded in  
 599 this groundmass. The bulk composition of this area is near-chondritic (Table 3), with the exception of  
 600 calcium which is enriched due to the presence of large dolomite grains. The abundance of magnetite is

601 around 4 % in this area (Table 3) and reaches up to 10 % in other areas of the meteorite. Framboidal  
602 magnetite is associated with Mg-rich carbonate. The amorphous silicate/phyllsilicate is seen on the BSE  
603 map to be divided into two sub-types (Fig. 15b). The first one (1) displays a fibrous texture and is richer in  
604 Mg (i.e. Mg-rich matrix). The second one (2) contains a higher content of nanosulfides which are below the  
605 mapping resolution and thus it appears richer in Fe (Fe-rich matrix). It is also more porous. In a few locations  
606 enrichment in S was visible in the amorphous/phyllsilicate material around the nanosulfides (Fig 15c). We  
607 classified these areas containing fine troilite and pentlandite as S-rich matrix.

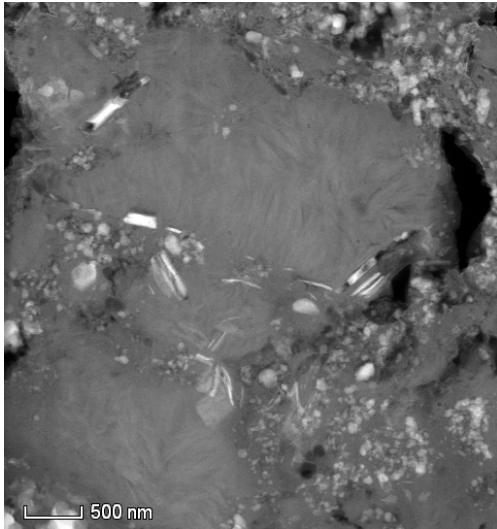
Table 3. Area abundances of matrix  
phases from ~1 mm<sup>2</sup> phase map.

Mg-rich matrix	33.94%
Fe-rich matrix	30.18%
S-rich matrix	10.66%
Si-rich matrix	1.55%
Olivine	4.09%
Metal grain	0.04%
Pyroxene	4.18%
Magnetite	4.06%
Carbonate	8.02%
Troilite	2.29%
Pentlandite	0.79%
Apatite	0.19%

608

#### 609 3.4.3 TEM analysis of matrix and chondrule rims

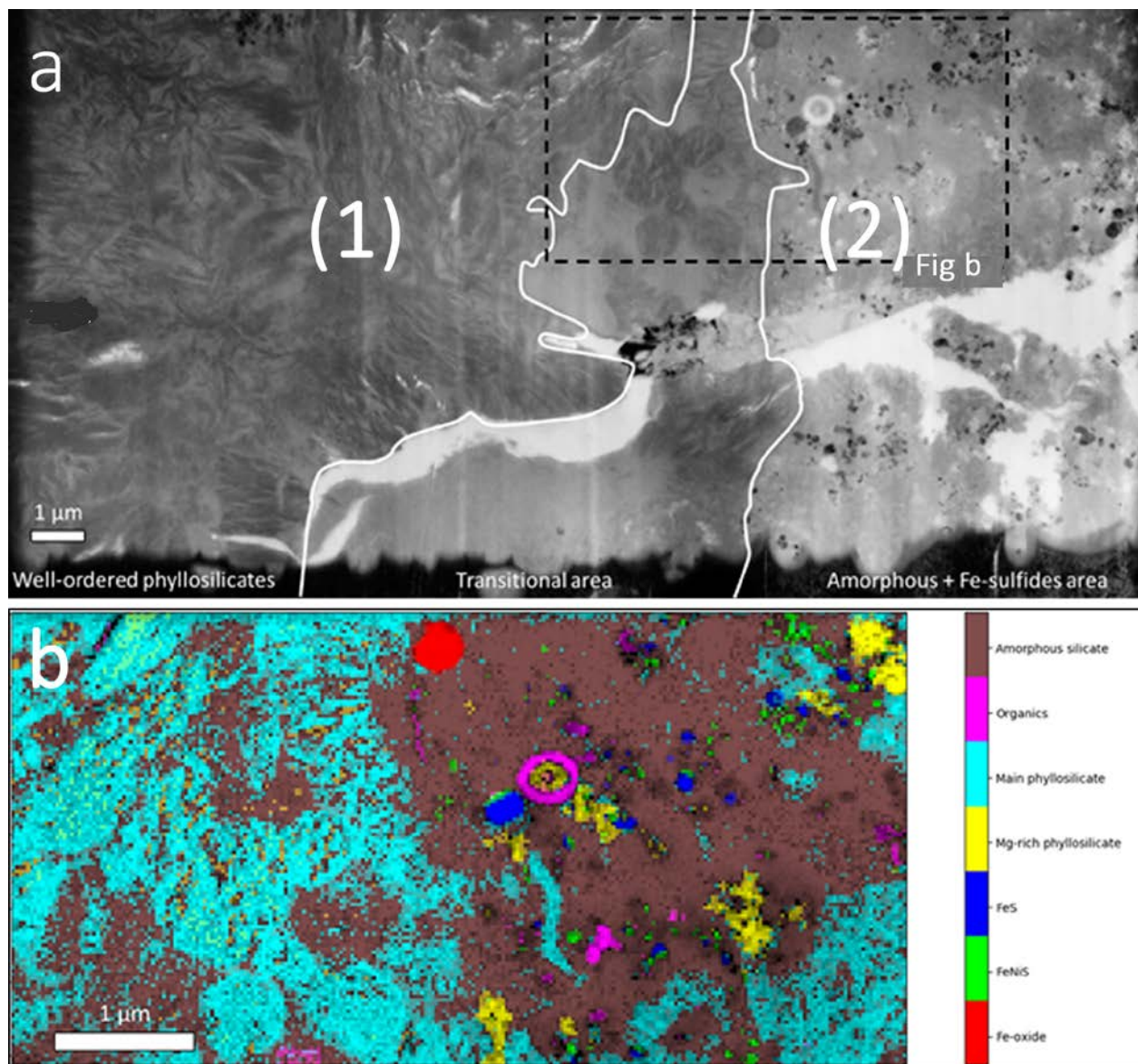
610 We studied several FIB sections which all present relatively homogeneous crystalline fibrous  
611 phyllosilicates next to more heterogeneous areas containing amorphous silicate and nanosulfides, oxides  
612 and organics. The finer scale TEM map showed very similar matrix details to those made by SEM (Fig.  
613 15b). We observed wide fields of relatively homogeneous fibrous phyllosilicates, juxtaposed with more  
614 heterogeneous areas rich in amorphous silicate and nanosulfides (Fig. 16). Sulfides occur principally as  
615 nano-crystalline aggregates and scattered needles. An Mg-Fe-Ni-Ca-S x-ray map of the same area is shown  
616 in Fig. S5.



617

618 Fig. 16. STEM Bright-field (BF) image of a typical area of the matrix of NWA 12563, showing  
619 phyllosilicate-rich patches and multi-phase regions with amorphous silicate and sulfide inclusions.

620



621  
 622 Fig 17. (a) STEM Bright-field (BF) image of a typical area of the matrix of the NWA 12563 chondrite,  
 623 showing (1) phyllosilicate-rich patches and (2) heterogeneous regions dominated by amorphous silicates.  
 624 The box outlined is shown in (b) a phase map of sulfide-poor fibrous phyllosilicate regions (1) and their  
 625 transition to sulfide-rich amorphous regions.

626  
 627 Phyllosilicate fibers are ~500 nm long in Fig. 15b and 16, but well ordered fibers ~1 μm long are  
 628 seen in Fig. 17a, where there are transitional regions between phyllosilicate patches and amorphous silicate  
 629 patches. We quantified the compositions of phyllosilicate and amorphous silicate at each pixel across the  
 630 three regions seen in Fig. 17 (Table 4 and S3). The aluminum content of matrix phyllosilicate, unlike that  
 631 for chondrules, is too low to be compatible with chlorite. The phyllosilicate has the same composition  
 632 throughout, with an (Mg+Fe)/Si ratio of 1.16, intermediate between the ratios of saponite and serpentine, as  
 633 in chondrule mesostasis but more ferroan (Fig. 18). Amorphous silicate displays an (Mg+Fe)/Si ratio of  
 634 0.97 in region (2) and 1.05 in the transitional region, with much less Fe than in CR and CM chondrites  
 (Chizmadia and Brearley, 2008; Abreu and Brearley, 2010; Le Guillou et al., 2015; Vinogradoff et al.,

635 2018). The Fe/Si ratio is fairly constant (~20.30) through the three regions, including intermediate areas,  
 636 but the concentration of S and Ni differs. Iron-rich analyses in Fig. 18b are also Ni- and S-rich, reflecting  
 637 numerous nanosulfide grains in the amorphous silicate. The heterogeneous distribution of sulfide grains in  
 638 amorphous silicate is seen In Table 4, where the standard deviations for S and Ni are much higher than the  
 639 mean values. The Fe-rich sulfide is pyrrhotite based on the (Fe+Ni)/S ratio of the most S-rich analyses, and  
 640 consists of 100-200 nm grains. It is surrounded by smaller grains of the less abundant pentlandite. Organic  
 641 particles, clusters of anhydrous silicates, and one magnetite grain (~ 400 nm) are also observed in Fig. 17b  
 642 but in low abundances. However, C and N are present in all analyses of amorphous silicate and phyllosilicate,  
 643 with similar abundances and C/N ratios (Table 4). We corrected the compositions for the presence of  
 644 organics and sulfide, and calculated a structural formula for the phyllosilicate, assuming 3 octahedral cations  
 645 per formula unit. The average analysis of the amorphous silicate is presented in the same format for  
 646 comparison purposes.

Table 4. Average compositions (ATEM) of matrix amorphous silicate (ams) and phyllosilicate (phyllo) in atomic % and atoms per formula unit.

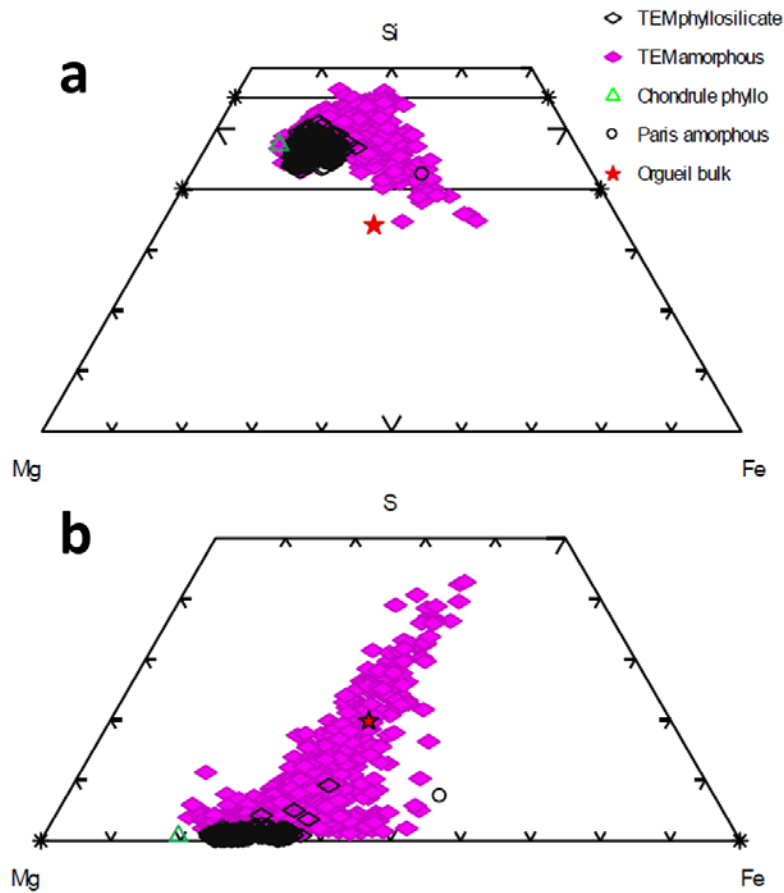
Atom %	ave ams	s.d.	ave phyllo	s.d.	ams SIL	phyllo SIL	ams afu	phyllo afu
Si	13.21	0.87	14.63	0.69	15.47	16.42	2.97	2.58
Al	1.02	0.16	1.35	0.24	1.19	1.52	0.23	0.24
Ti	0.017	0.007	0.006	0.004	0.020	0.010	0.000	0.000
Cr	0.114	0.018	0.129	0.022	0.130	0.140	0.030	0.020
Fe	4.56	1.02	4.72	0.50	5.34	5.30	1.03	0.83
Mn	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Mg	8.78	1.26	12.26	1.06	10.28	13.76	1.97	2.17
Ca	0.33	0.12	0.09	0.05	0.39	0.10	0.07	0.02
Na	0.28	0.06	0.53	0.07	0.33	0.59	0.06	0.09
K	0.029	0.007	0.013	0.006	0.030	0.010	0.010	0.000
P	0.001	0.002	0.000	0.001	0.000	0.000	0.000	0.000
S	0.97	1.43	0.14	0.11				
Ni	0.111	0.271	0.007	0.009				
Cl	0.063	0.059	0.050	0.023	0.070	0.060	0.010	0.010
C	12.42	2.39	9.90	1.95				
N	1.12	0.30	0.82	0.24				
O	56.97	2.32	55.34	1.34	66.74	62.09	12.81	9.77
Total	100.00		100.00		100.00	100.00	100.00	100.00

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648

649

650

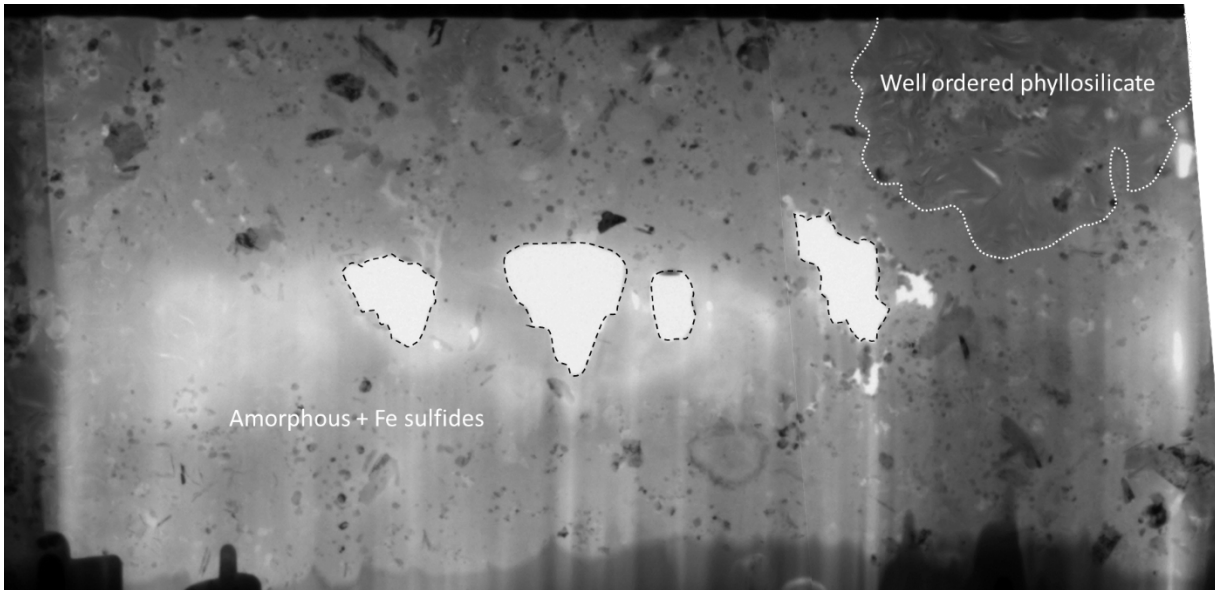


651  
 652 Fig.18. Compositions in atom % determined by ATEM (a) Mg-SiAl-Fe diagram showing that the  
 653 phyllosilicate and amorphous silicate compositions are intermediate between saponite and serpentine  
 654 stoichiometry (tie lines for reference). The phyllosilicate has a narrower range of composition and generally  
 655 plots on the Mg-rich end of the compositional range. (b) Mg-S-Fe diagram showing that the more Fe-rich  
 656 points of the amorphous silicates are due to mixture with Fe-Ni nanosulfides. Orgueil composition from  
 657 Jarosewich (1990); Paris amorphous silicate from Hewins et al. (2014).

658

### 659 3.4.4 Fine-grained rims

660 Many chondrules in NWA 12563 are surrounded by accretionary rims that are bright in BSE  
 661 because of very fine Fe-rich grains, and discrete clusters of fine magnetite spherules (Fig. 1, 6, 10, 14). The  
 662 large composite Al-rich and relatively metal-poor chondrule of Fig. 6 has a rim with little obvious magnetite.  
 663 Similarly, AOA and Type II chondrules do not have bright rims. Some, especially Type IAB chondrules  
 664 have a halo of altered chondrule material with pseudomorphs after metal beneath the fine-grained rim. The  
 665 sporadic nature of rim occurrence suggests a regolith history but a detailed study of fine-grained rims  
 666 suggests that chondrules in Semarkona suggests rimming and fracturing of chondrules in the disk before  
 667 accretion together with matrix dust (Zanetta et al., 2021).



668  
 669 Fig. 19. STEM BF image of the FIB section ~20  $\mu\text{m}$  long sampled in a fine-grained rim with amorphous  
 670 silicate (light grey) locally with fibrous phyllosilicate crystals, organic material and anhydrous silicate  
 671 (medium grey), pores (black) and holes (white).

672 An FIB section has been sampled in one of those fine-grained rims (Fig. 19). This rim consists  
 673 mostly of amorphous silicates with embedded nanosulfides, small anhydrous silicate grains ( $<1\mu\text{m}$ ) and  
 674 organic matter. Rare patches of ordered phyllosilicates are visible. The rim differs from the amorphous  
 675 patches in matrix in that the porosity is much lower. The large holes in the middle of the section are due to  
 676 FIB damage during the final thinning operation. The compositions of the amorphous silicates and  
 677 phyllosilicate patches are comparable to those of the matrix.

678 *3.4.5 Iron valency in matrix material*

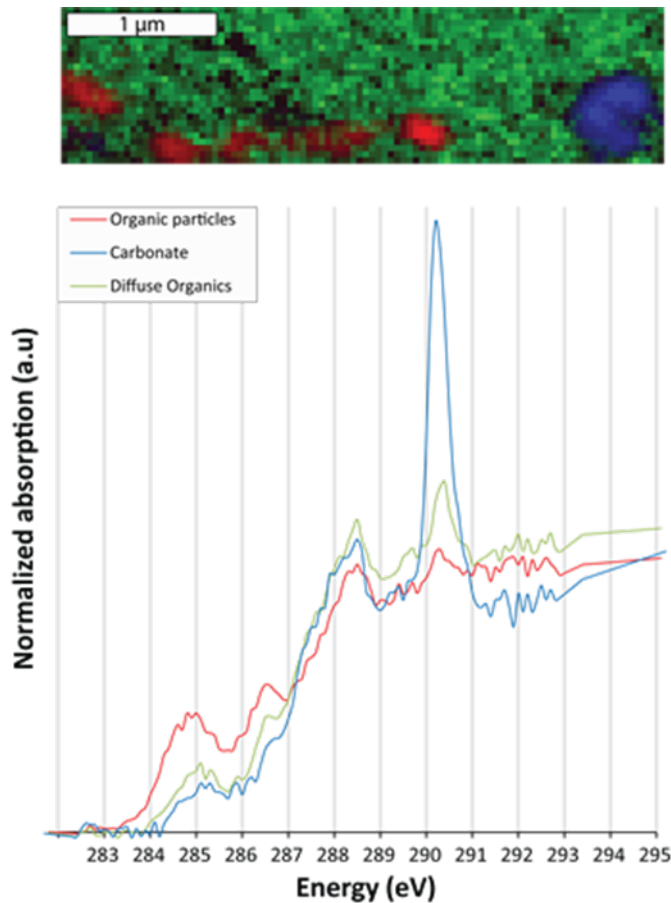
679 The iron valency of amorphous silicate and phyllosilicate was measured by STXM on an FIB  
 680 section that had not been previously observed by TEM. The spatial resolution is not sufficient to allow us  
 681 to always distinguish between amorphous silicate and phyllosilicate, and some pixels might contain a  
 682 mixture of both. Using the calibration proposed by Le Guillou et al. (2015), we quantified the  $\text{Fe}^{3+}/\Sigma\text{Fe}$  ratio  
 683 and obtained a value of  $\sim 75\%$  ( $\pm 5$ ), a value similar to that found in most CR chondrites (Le Guillou et al.,  
 684 2015), in Acfer 094 (Hopp and Volmer, 2018) and in Semarkona (Dobrica et al., 2019).

685 *3.4.6 The nature of organic matter*

686 We studied individual organic particles, diffuse organics trapped within amorphous/phyllosilicates  
 687 and carbonate grains by STXM on a FIB section. XANES spectra of the organic matter at the carbon K-  
 688 edge were obtained. The main functional groups observed are: aromatic-olefinic at 285 eV, ketone-phenol  
 689 at 286.5 eV, aliphatic at 287.5 eV, carboxylic acid at 288.6 eV and carbonate at 291.3 eV. The color map  
 690 (Fig. 20) represents the three different components that we identified: individual organic particles (red),  
 691 diffuse organics trapped within amorphous/phyllosilicates (green) and ( $\sim 500\text{ nm}$ ) carbonate grains. We  
 692 found individual organic particles slightly more aromatic-olefinic rich than the surrounding diffuse organic



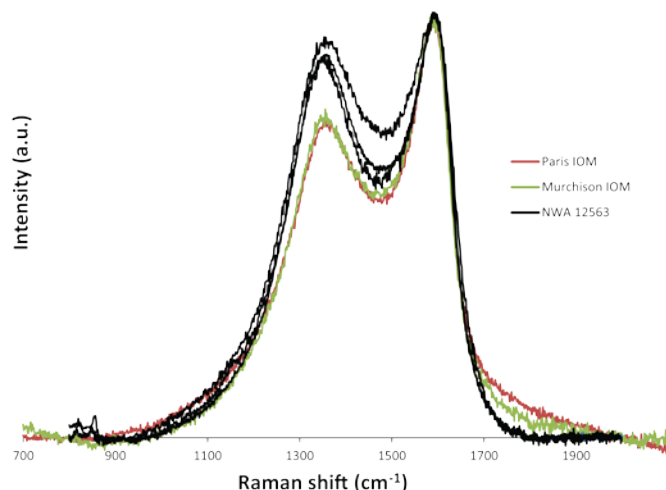
693 matter (dark red), associated with amorphous silicate and/or phyllosilicates, that is richer in aliphatics and  
694 carboxylic functional groups.



695  
696 Fig. 20. XANES spectra at the carbon K edge obtained on a matrix FIB section. The color map shows  
697 individual organic particles, diffuse organics trapped within amorphous/phyllosilicates and carbonate  
698 grains.

699 We acquired Raman spectra on an epoxy-free section and compared them in Fig. 21 to the spectra  
700 of the insoluble organic matter of Murchison and Paris CM chondrites (Vinogradoff et al., 2018). The D  
701 band of NWA 12563 spectra is slightly more intense. However, NWA 12536 spectra were measured on  
702 meteorite chips and therefore include the contribution of the soluble organic compounds, whereas this is not  
703 the case for the Murchison and Paris spectra, and could explain the difference. They are also similar to  
704 published data for CM2 chondrites (Bonal et al., 2006, 2007; Busemann et al., 2007; Quirico et al., 2018).  
705 Therefore, the subtle differences between them cannot be clearly interpreted. The main result is that no  
706 significant difference is observed, and that, within a few tens of degrees, the peak temperature encountered  
707 by organics was similar to that of CM2 chondrites, i.e. below 250°C (Busemann et al., 2007).

708



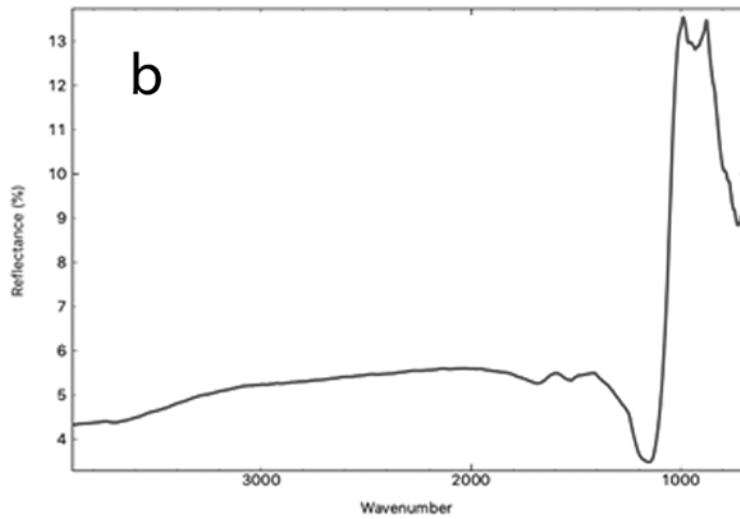
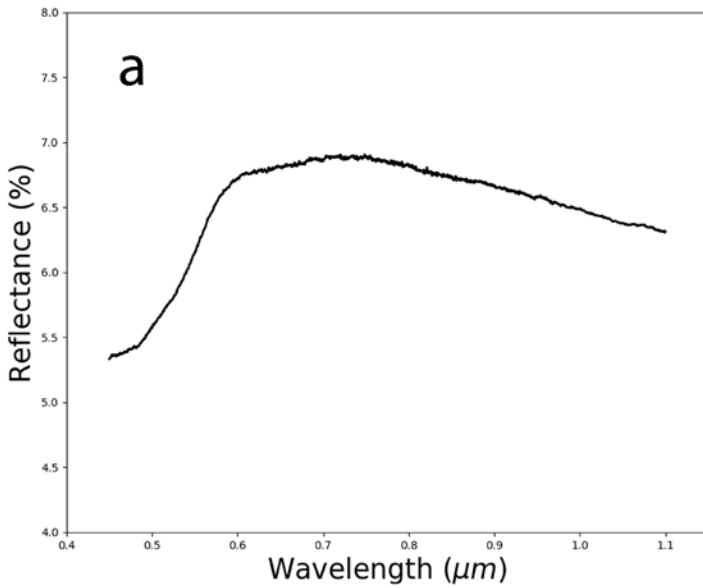
709

710 Fig. 21. Raman spectra of the NWA 12563 chondrite compared to spectra from CM2 chondrites.

### 711 3.4.7 Spectral properties

712 The large scale (mm-sized spot) diffuse Vis-NIR and confocal MIR reflectance spectra of NWA 12563  
 713 are reported in Fig. 22. The reflectance in the visible at 0.55  $\mu\text{m}$  is found to be  $R=6.2\pm 0.5\%$ , while the  
 714 spectral slope in the 0.45-1.0  $\mu\text{m}$  range is  $\text{slope} = 0.27\pm 0.07 \mu\text{m}^{-1}$ . In both cases, uncertainties represent a 1  
 715 sigma error bar. The left shoulder of a 1- $\mu\text{m}$  band typical of olivine can be seen in the spectrum. Other  
 716 features are an inflection between 0.6 and 0.7  $\mu\text{m}$ , and a dramatic drop in reflectance for wavelengths below  
 717 0.59  $\mu\text{m}$ . At much smaller spatial scale (less than 50  $\mu\text{m}$ ), the meteorite show very heterogeneous values of  
 718 both albedo and spectral slope. The reflectance at 0.55  $\mu\text{m}$  can be as low as  $R=3.9\pm 0.7\%$  in dark matrix  
 719 areas, whereas it rises up to about  $R=24.6\pm 0.7\%$  in bright magnetite-rich areas. The corresponding spectral  
 720 slopes vary from very “red” in dark matrix areas ( $0.39\pm 0.1 \mu\text{m}^{-1}$ ) to very “blue” in magnetite-rich areas ( $-$   
 721  $0.54\pm 0.02 \mu\text{m}^{-1}$ ). A potential weak hydration feature around 0.64  $\mu\text{m}$  is detected in some matrix locations.  
 722

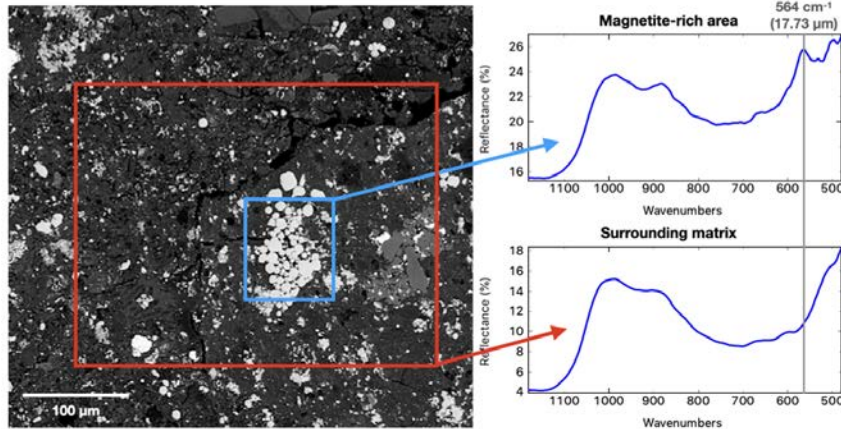
723 The average MIR spectrum of NWA 12563 (Fig. 22b) shows a clear transition between the volume  
 724 scattering regime and the surface scattering regime around 1150  $\text{cm}^{-1}$  ( $\sim 8.7 \mu\text{m}$ , main Christiansen feature).  
 725 Bands at higher wavenumbers (lower wavelengths) are detected as absorption features towards the bottom.  
 726 These include a weak hydration feature at 3700  $\text{cm}^{-1}$  (2.7  $\mu\text{m}$ ), typical of phyllosilicates, and an extremely  
 727 weak aliphatic CH feature around 2900-3000  $\text{cm}^{-1}$  (3.4  $\mu\text{m}$ ). Overall, the OH and CH features in the volume  
 728 scattering spectral range appear relatively weak, especially with respect to features at longer wavelengths,  
 729 and this is probably due to the geometry of our measurements (backscattering) which tends to favor the  
 730 surface scattering rather than the volume scattering regime. However, this observation is consistent with the  
 731 H content observed, which is low compared to average CM2 chondrites. For wavenumbers lower  
 732 (wavelengths higher) than the position of the main Christiansen feature, strong reststrahlen bands are  
 733 observed as peaks towards the top, such as the peaks at 985  $\text{cm}^{-1}$  (10.15  $\mu\text{m}$ ) and at 874  $\text{cm}^{-1}$  (11.44  $\mu\text{m}$ ),  
 734 that are generally attributed to hydrated silicates and olivine respectively. Smaller features between 945-  
 735 960  $\text{cm}^{-1}$  (10.4-10.6  $\mu\text{m}$ ) may be attributed to pyroxenes and other anhydrous phases. A small spectral  
 736 contribution from carbonates is probably observed between 1410-1470  $\text{cm}^{-1}$  (6.8-7.1  $\mu\text{m}$ ). Minor bands are  
 737 present around 765-785  $\text{cm}^{-1}$  (12.7-13.1  $\mu\text{m}$ ), which might be related to different types of oxides.



738  
 739 Fig. 22. Diffuse Vis-NIR (a) and confocal MIR reflectance spectra (b) of a chip of NWA 12563. (a) shows  
 740 the left shoulder of a 1- $\mu\text{m}$  band typical of olivines. (b) shows a weak hydration feature at  $3700\text{ cm}^{-1}$  (2.7  
 741  $\mu\text{m}$ ), typical of phyllosilicates, and an extremely weak aliphatic CH feature around  $2900\text{--}3000\text{ cm}^{-1}$  (3.4  
 742  $\mu\text{m}$ ), and peaks at  $985\text{ cm}^{-1}$  (10.15  $\mu\text{m}$ ) and  $874\text{ cm}^{-1}$  (11.44  $\mu\text{m}$ ), due to hydrated silicates and olivine  
 743 respectively.

744 The presence of magnetite is not obvious in spectra measured on large surface areas of NWA  
 745 12563, e.g. a 400 mm matrix region in Fig. 23. However, the spectrum for the center of this area  
 746 containing a clump of framboidal magnetite shows the  $564\text{ cm}^{-1}$  (17.7  $\mu\text{m}$ ) band for magnetite seen at  $555$   
 747  $\text{cm}^{-1}$  in Bennu (Hamilton et al., 2019).

748



749  
 750 Fig. 23. The MIR spectral signature of magnetite at  $564\text{ cm}^{-1}$  ( $17.73\text{ }\mu\text{m}$ ) is observed from a small body of  
 751 framboids but is obscured when a small region of matrix is analyzed.

## 752 4. DISCUSSION

### 753 4.1 NWA 12563 is unlike CM2 chondrites

754 Though the abundances of anhydrous silicates, matrix and phyllosilicates suggest that NWA  
 755 12563 is a C2 chondrite, considering secondary minerals, it is not immediately clear what class it  
 756 represents. Although the matrix is more abundant than in most CM2 chondrites, relatively little of its  
 757 amorphous silicate is converted to phyllosilicate. We examine several key properties and alteration  
 758 mineral assemblages in NWA 12563 in Table 5. Its O isotope composition is at the  $\delta^{18}\text{O}$ -rich end of the  
 759 CM range and close to Bells and Essebi (Fig. 2). The C and N abundances, and H isotopic composition for  
 760 NWA 12563 are typical of CM chondrites (Pearson et al., 2006; Alexander et al., 2010, 2013. Vacher et  
 761 al., 2020) but not that far from those of CY (heated) chondrites (Table 5). The  $\delta^{13}\text{C}$  is considerably lower  
 762 than in CM chondrites and similar to that in CI chondrites.

763  
 764 From Acfer 094 (Bischoff and Geiger, 1994; Newton et al., 1995; Greshake, 1997), Asuka 12169  
 765 (M. Kimura et al., 2019; Noguchi et al., 2020), DOM 08006 (Alexander et al., 2018; Nittler et al., 2018;  
 766 Davidson et al., 2019) and MET 00426 and QUE 99177 (Abreu and Brearley, 2010; Le Guillou and  
 767 Brearley, 2014; Harju et al., 2014; McAdam et al., 2018), among the most pristine carbonaceous  
 768 chondrites, we can infer that the main primary phases in chondrules and matrix in 3.0 carbonaceous  
 769 chondrites are olivine, pyroxene, glass, kamacite, sulfide (including nanosulfide  $<100\text{ nm}$ ) and amorphous  
 770 silicate. The transformation of these phases, along with ice and organic material, has led to different suites  
 771 of secondary phases in C2 chondrites.

772  
 773 Aqueous alteration of olivine mainly produces serpentine, and carbonaceous chondrite matrices  
 774 contain serpentine minerals, yet olivine phenocrysts in chondrules do not disappear until the CM2.0/2.1  
 775 alteration stage e.g. LAP 02277 (Hanowski and Brearley, 2001; Rubin et al., 2007). Though alteration of  
 776 olivine can be used to designate subgroups (Hanowski and Brearley, 2001; Velbel et al., 2015), many other  
 777 primary phases are more useful in subdividing CM2 chondrites: kamacite, matrix amorphous silicate, and  
 778 chondrule glass are replaced before olivine is much affected. Approaches tracking single facets of alteration,  
 779 e.g. replacement of kamacite (M. Kimura et al., 2011; Palmer and Lauretta, 2011), hydrogen abundance  
 780 (Alexander, 2013), or total phyllosilicate abundance (Howard et al., 2015), are useful individually but are  
 781 difficult to combine in a consistent manner. Holistic approaches (e.g. Rubin et al., 2007; Velbel et al., 2015)  
 782 have been used successfully with moderately to totally altered CM chondrites, where key steps are the  
 783 development of cronstedtite and tochilinite and their eventual disappearance.

784 The chondrule kamacite in NWA 12563 is pseudomorphed by serpentine with no tochilinite,  
785 whereas in CM2 chondrites the metal is replaced by tochilinite and cronstedtite (Tomeoka and Buseck,  
786 1985; McSween, 1987; Rubin et al., 2007; Maeda et al., 2009; Palmer and Lauretta, 2011; Vacher et al.,  
787 2019). The occurrence of tochilinite is almost confined to CM2 chondrites (Zolensky et al., 1993; Rubin et  
788 al., 2007; Pignatelli et al., 2017) and its abundance is much lower in CM1. CM1 chondrites differ from  
789 NWA 12563, in having less olivine, less magnetite, less amorphous silicate, and more phyllosilicate (King  
790 et al., 2017). For other chondrite groups, minor amounts have been observed only in CR chondrites (Le  
791 Guillou and Brearley, 2014; Changela et al., 2018) though other aspects of their alteration are similar to  
792 those of CM chondrites (Harju et al., 2014). We have not observed tochilinite in NWA 12563. Its  
793 chondrule mesostasis contains amorphous silicate, serpentine-saponite, and chlorite rather than serpentine  
794 and cronstedtite, as in CM2 chondrites. Its matrix contains abundant amorphous silicate, serpentine-  
795 saponite, and magnetite, whereas CM2 matrix contains amorphous silicate in the least altered samples  
796 (Hewins et al., 2014), serpentine, cronstedtite, and tochilinite (Ikeda, 1983; Zolensky et al., 1993; Rubin et  
797 al., 2007; Velbel et al., 2015; Pignatelli et al., 2017). NWA 12563 alteration is difficult to categorize on  
798 the CM2 scale of Rubin et al. (2007) because of the lack of tochilinite and cronstedtite. Using the scale of  
799 Velbel et al. (2015) we find that NWA 12563 would be stage 1 based on olivine and metal alteration level.  
800 The most striking feature of NWA 12563 is the abundance of magnetite, a phase associated with heavy  
801 alteration, along with the surviving primary phases olivine and porous amorphous silicate (associated with  
802 small (<100nm) nanosulfides). Magnetite is anomalously abundant from the perspective of CM2  
803 chondrites and indicates significantly different alteration conditions (temperature, fO<sub>2</sub>, and/or fluid  
804 compositions, etc...) for NWA 12563.

#### 805 **4.2 A cluster of ungrouped C2 chondrites**

806 In Table 5 we note similarities to type 2 ungrouped chondrites (as defined in Fig. 2) as well as  
807 significant differences from CM2 chondrites. Oxygen isotopic compositions are indicators of both  
808 provenance and processing, and similar alteration processes in chondrites may lead to overlap of  
809 taxonomic classes. In this case mineral abundances and in particular the dominant Fe-rich mineral,  
810 kamacite, tochilinite or magnetite, is also a useful criterion. Ungrouped chondrites like Bells and Tagish  
811 Lake all contain ~10% magnetite (Table 5), whereas Watson et al. (1975) noted that nine tochilinite-rich  
812 CM chondrites contain <1 wt.% magnetite, and Type 1 CMs have ~3% (King et al., 2017); more pristine  
813 ungrouped chondrites like Acfer 094 are kamacite-rich. Using the oxygen isotope data base and chondrite  
814 nomenclature of Greenwood et al. (2020), we plotted the ungrouped and major group chondrites. The  
815 well-defined distribution for CM chondrites overlaps slightly with those of CO and CV chondrites and is  
816 distinct from the other coherent groups in Fig. 2. The type 2 ungrouped chondrites divide into two  
817 clusters, C2-ung1 and C2-ung2, at either end of the CM O isotope distribution. Clayton and Mayeda  
818 (1999b) had already noted that three of the C2-ung1 cluster (Bells, Essebi, and Niger I) were distinct,  
819 calling them “CI2?” chondrites. Rochette et al. (2008) proposed a new association of ungrouped  
820 chondrites with high magnetic susceptibility (i.e., high kamacite or magnetite content) including Bells,  
821 Essebi, and Acfer 094. We plot magnetic susceptibility log  $\chi$  against  $\delta^{18}\text{O}$  (Fig. 22) and show that the  
822 magnetite- and  $^{18}\text{O}$ -rich cluster C2-ung 1 (Table 5), is separated from the field of CM chondrites. The  
823 kamacite-rich chondrites like Acfer 094 plot in a cluster (C2-ung 2) poor in  $^{18}\text{O}$  overlapping the CM and  
824 CO chondrite fields.  
825

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Table 5. Alteration minerals and properties of seven cluster C2-ung1 and other chondrites.

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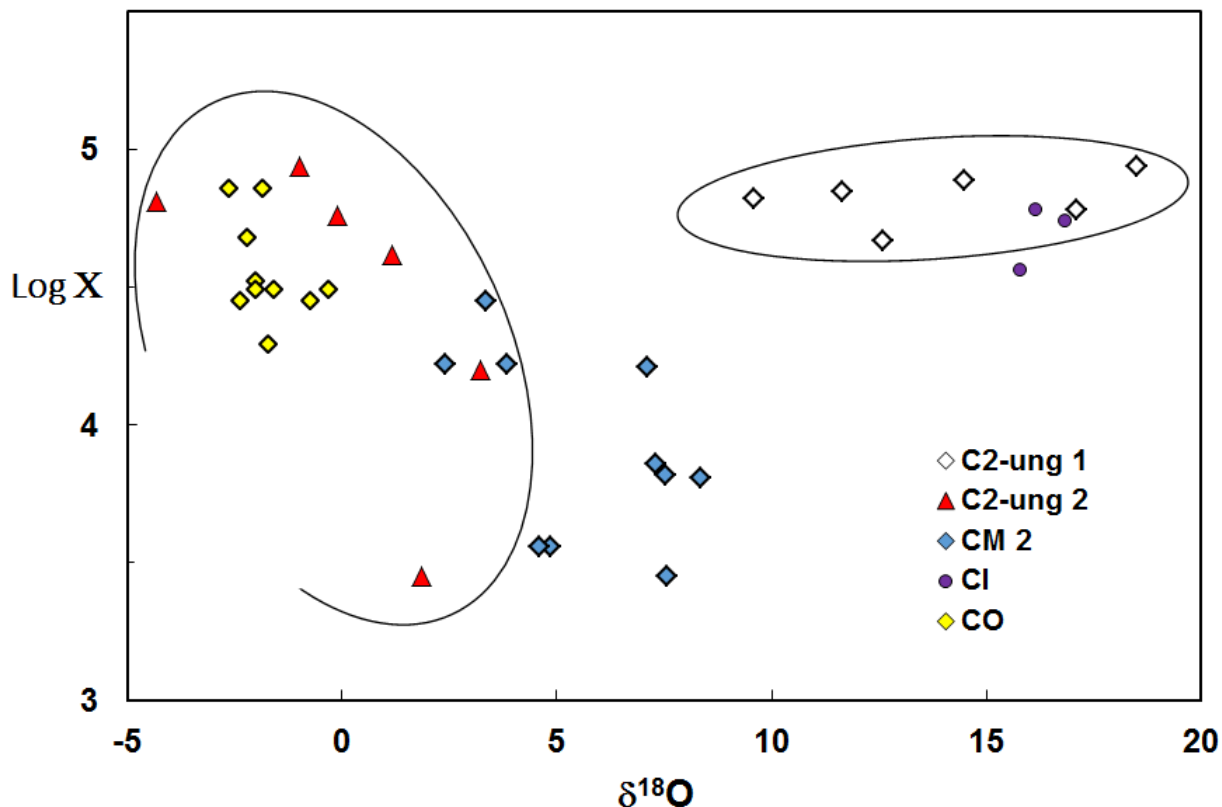
	Bells	Essebi	NWA 1256 3	Niger I	WIS 91600	Tagish Lake	MET 00432	CI Orgueil	CM2 Murchison	CY (heated)
Metal	~alt	~alt	pse	pse	pse	Pse			0.16	K+T
Phyllo	s+s	s+s	s+s		dehyd	s+s	sap	s+s	s+cr	dehyd
TCl	none	none	none	none	none	None	none	none	15-40%	none
Magt	4-11	5-11	6.8	fr	6-8	Fr	fr	11	1.1	fr
Sulfide	fg	fg	ros		ros	Ros				
$\delta^{18}\text{O}$	9.59	11.65	12.60	14.5	17.1	18.5	14-18	16.16	7.3	21.97
$\log \chi$	4.82	4.85	4.67	4.89	4.78	4.94		4.78	3.86	
$M_s$ *			6.26	9.85		9.90		9.55	0.67	3.96*
C wt. %	1.91	3.1	2.13	2.75		4.1/2.1		3.5/3.4	2.1/2.31	1.42
$\delta^{13}\text{C} \text{ ‰}$	-19.9	-0.6	-12.2			13.4/4.7		-14.7/-12.9	-2.9/-2.4	-5.27
N(wt. %)	0.064	0.137	0.095					0.149	0.105	0.075
H wt. %	0.559	0.938	0.725			0.85/0.43		1.56/0.93	1.07/0.83	0.792
$\delta\text{D} \text{ ‰}$	305	337	-57.5			374/442		83.9/138	-61.7/-74.9	-102.35
C/H (at)	0.26	0.28	0.25			0.40/0.43		0.19/0.30	0.16/0.23	0.15
$C_{10M}$ wt %	0.96	1.30			2.26	2.00		2.00	1.06	
Refs	abdio	abeijmo	c	abcf	abdhnmp	Abgkmnor	p	abeilo	abijo	anqso

References: a Greenwood et al. 2020; b Rochette et al. 2008; c This work; d Brearley 1995, 2004; e Buseck & Hua 1993; f Desnoyer et al. 1980; g Grady et al., 2002; h Hanna et al. 2020; i Hyman and Rowe 1985,1986; j Rubin et al. 2007; k Takayama and Tomeoka 2012; l Tomeoka & Buseck 1988; m Zolensky 1991, 2002; n Tonui et al. 2014; o Alexander et al. 2012/Vacher et al. 2020; p Nakamura et al. 2013 and Yamanobe et al. 2018; q King et al., 2019; r Bischoff et al. (2020); s Gattacceca et al., 2018. Abbreviations: ~alt – partly altered; pse – pseudomorphs K+T – kamacite+ taenite; phyllo – phyllosilicate; s+s – serpentine + saponite; dehyd dehydrated; sap – saponite, s+cr – serpentine + cronstedtite; TCl tochilinite + cronstedtite; fr – frambooids; fg fine grained; ros – rosettes. \* Am<sup>2</sup>/kg. The magnetic properties for CY chondrites are for Dhofar 2066.

826 The other chondrites in the C2-ung1 cluster also lack tochilinite like NWA 12563 (Table 5). WIS  
827 91600 (Brearley, 2004), Niger I, and Tagish Lake (Takayama and Tomeoka, 2012) also contain serpentine  
828 pseudomorphs after kamacite. Similar ovoids are seen in chondrule rims in CR chondrites (Weisberg et  
829 al., 1993). Fe is present as magnetite frambooids in the matrix in all four chondrites (Table 5), as well as in  
830 Bells and Essebi. The matrix in NWA 12563, CR and CI chondrites includes serpentine and saponite, as  
831 do Bells (Brearley, 1995), Essebi (Zolensky, 1991), and Tagish Lake (Zolensky et al., 2002; Takayama  
832 and Tomeoka, 2012) but that of CM chondrites contains serpentine and cronstedtite.

### 833 4.3 Alteration conditions in NWA 12563 and C2-ung1 chondrites

834 It is a truth universally acknowledged, that olivine surrounded by water must alter to serpentine.  
835 However, in C2 chondrites, where aqueous alteration is a required characteristic, chondrule olivine



836

837 Fig. 23. In terms of magnetic susceptibility and oxygen isotopic composition, the cluster of magnetite-rich  
 838 ungrouped chondrites including NWA 12563 (C2-ung1) is distinct from CM2 chondrites, while the  
 839 kamacite-bearing cluster including Acfer 094 (C2-ung2) overlaps with CM2 and CO chondrites.

840 remains fresh except in the lowest subtypes. For example, in Nogoya, a CM2.2 chondrite (Rubin et al.,  
 841 2007), chondrule phenocryst olivine is cut by veins of serpentine (Velbel et al., 2012). The lack of such  
 842 veins in chondrite matrix indicates that matrix was the source of the altering fluid. The primary phases in  
 843 NWA 12563 most vulnerable to alteration are amorphous silicate in the matrix, and metal and mesostasis  
 844 in the chondrules. As water was probably accreted within the matrix, the extent of alteration of chondrule  
 845 olivine and pyroxene may depend on the initial concentration of trapped ice, as well as on physical  
 846 conditions discussed below.

847 In NWA 12563, chondrule mesostasis has lost Si, Al, and Ca, and gained Mg and Fe when  
 848 transformed to phyllosilicates, based on Type I chondrule fresh mesostasis composition (Jones and Scott,  
 849 1989). In NWA 12563, probable sources for Mg were pyroxene when present and altered, and chondrule  
 850 metal for Fe. The altered metal inclusions show a continuum of composition between kamacite and  
 851 serpentine though speckled with Cr-, P-, S-rich phases (Fig. 11). Kamacite is abundant in Type I  
 852 chondrules (McSween, 1977) but troilite is abundant in Type IIA chondrules (Jones, 1990). Fe lost from  
 853 kamacite could be taken up by either mesostasis and by matrix, where it could eventually have contributed  
 854 to magnetite precipitation. Serpentine pseudomorphs require entry of Mg and Si into chondrule metal in  
 855 NWA 12563, as in WIS 91600 (Brearley, 2004). The serpentine S contents are higher than those in  
 856 terrestrial serpentine, except where sulfide microinclusions are present (Debret et al., 2017), and suggest a  
 857 fluid also containing S, but insufficient for forming tochilinite, unless this is strictly due to physical

858 conditions. Alteration of troilite particularly from defunct Type IIA chondrules would provide S for  
859 alteration.

860 In the weakly altered CR chondrite MET 00426, alteration has begun locally in matrix: amorphous silicate  
861 and nanosulfide are accompanied by nanophyllosilicate, explained by reaction after the melting of  
862 interstitial ice (Le Guillou and Brearley, 2014). In Acfer 094, originally considered a CM3 chondrite  
863 (Bischoff and Geiger, 1994), the matrix amorphous silicate is poor in Mg and has high  $\text{Fe}^{3+}/\Sigma\text{Fe}$  ratios  
864 (0.66–0.73; Hopp and Vollmer, 2018), suggesting aqueous alteration and oxidation after formation by  
865 condensation. In the CR chondrites and Semarkona, the amorphous silicate phase with high Fe content and  
866 high  $\text{Fe}^{3+}/\Sigma\text{Fe}$  has been transformed during alteration to a more magnesian serpentine-saponite mixture  
867 and magnetite (Le Guillou et al., 2015; Dobrica et al., 2019). In weakly altered CM chondrites the partial  
868 hydration of the matrix amorphous silicate phase produces homogeneous phyllosilicate areas (Chizmadia  
869 and Brearley, 2008; Hewins et al., 2014; Leroux et al., 2015; Vinogradoff et al., 2018). In NWA 12563  
870 this is also observed (Fig. S5 and 17 a) are not associated with nanosulfides. The composition of matrix  
871 phyllosilicate (Table 3) indicates that Fe and S were lost from amorphous silicate and nanosulfide),  
872 suggesting that the larger sulfide crystals were formed at the same time.

873 Magnetite framboids precipitate from concentrated Fe-rich fluids (Yu and Kwak, 2010; Y. Kimura  
874 et al, 2013) and possible sources for the Fe in NWA 12563 are amorphous silicate and metal pseudomorphs.  
875 The amorphous phase in NWA 12563 has a high  $\text{Fe}^{3+}/\text{Fe}^{2+}$  ratio and has partly transformed to fibrous well-  
876 ordered serpentine and saponite. Though oxidized, the amorphous silicate observed in NWA 12563 is much  
877 less iron-rich than that in CRs or CMs, e.g. the amorphous silicate in Paris (Fig. 18.). This is consistent with  
878 it being at least a partial source for oxidized solutions that deposited magnetite. Although magnetite  
879 framboids occur in all the C2-ung1 chondrites, serpentine pseudomorphs after chondrule kamacite are  
880 lacking in Bells and Essebi (Table 1) and so may not be a necessary source of the Fe in magnetite. Magnetite  
881 becomes stable relative to phyllosilicates at very low pressures under conditions of low water/rock ratios  
882 (Yu, 2012). Exhaustion of matrix water trapped by hydration and oxidation reactions would have caused  
883 late precipitation of magnetite (and probably coarse sulfide and carbonate). Magnetite precipitation indicates  
884 the exhaustion of interstitial fluid, effectively permitting the survival of amorphous silicate but also olivine.

885 The assemblages with serpentine-saponite and chlorite in NWA 12563 are reminiscent of CR  
886 chondrites (Weisberg et al., 1993) rather than CM2 chondrite with tochilinite and cronstedtite. CR  
887 chondrites also show replacement of metal by serpentine. In CM2 chondrites, metal is replaced by  
888 tochilinite, indicating an S-rich fluid, along with P- and Cr-rich phases (Palmer and Lauretta, 2011;  
889 Pignatelli et al., 2017). NWA 12563 contains magnetite framboids in chondrule rims, as well as in the  
890 matrix. Magnetite also partially replaces sulfide in the rosettes. The association of dolomite with  
891 magnetite seen in Fig. 19a suggests a simultaneous origin for these two phases, probably late. The  
892 abundance of magnetite in NWA 12563 is anomalous relative to CI chondrites as its olivine is not altered,  
893 and relative to CM chondrites as it contains no tochilinite and cronstedtite. The presence of both pyrrhotite  
894 and magnetite constrains  $f\text{S}_2$  and  $f\text{O}_2$  to conditions well above the stability fields of kamacite and  
895 tochilinite (Vacher et al., 2019). The presence of magnetite with no cronstedtite is also an indication of  
896 high  $f\text{O}_2$  and low silica activity  $a_{\text{SiO}_2}$  (Palmer and Lauretta, 2011; Zolotov, 2014).

897 Temperature is clearly important in determining phase assemblages. Both saponite and chlorite  
898 can be stable at high metamorphic temperatures (Iiyama and Roy, 1963; Fawcett and Yoder, 1966).  
899 Amorphous silicate also seems to support relatively high temperatures, at least with low  $P_{\text{H}_2\text{O}}$ , since it is  
900 only partially transformed to olivine at 700°C (Davoisne et al., 2006) and is abundant in the least  
901 metamorphosed ordinary and CO chondrites (Dobrica et al., 2019, 2020; McAdam et al, 2018).  
902 Cronstedtite is stable at low temperatures and low  $a_{\text{SiO}_2}$ : Zolotov (2014) showed that cronstedtite is



903 replaced by magnetite and greenalite, the ferroan serpentine endmember, above about 100°C, and a metal-  
904 rich micro-region will react with local acidic fluid predominantly to magnetite (Zolotov et al., 2015).  
905 Pignatelli et al. (2017) also argued that tochilinite-cronstedtite intergrowths formed at <100 °C. However,  
906 synthesis experiments (Vacher et al., 2019) indicated that tochilinite formed from metal at 120-160°C in  
907 CM chondrites under S-bearing alkaline conditions; and cronstedtite crystals could also form from  
908 amorphous silicate and metal during later cooling (50–120°C).

909  
910 Higher temperatures for NWA 12563 would be consistent with arguments that the saponite-  
911 bearing CI and C2-ung1 chondrites were altered at higher temperatures than CM chondrites considering  
912 modeling of phase equilibria (Zolensky et al., 1989) and oxygen isotope data (Clayton and Mayeda,  
913 1999a,b). Raman spectra of organic material (Fig.21) for NWA 12563 and the CM2 chondrites Paris and  
914 Murchison (Vinogradoff et al., 2018) and therefore are not indicative of a higher temperature that could  
915 demonstrate that the stability limits for cronstedtite and tochilinite were exceeded in NWA 12563.  
916 However, temperatures based on Raman data for the organics in these meteorites are in any case poorly  
917 constrained below 220°C (Busemann et al., 2007; Quirico et al., 2018). In a similar fashion, the organic  
918 XANES data are also comparable with Paris and Murchison, which reinforces the conclusion that  
919 temperature was not significantly higher in NWA 12563. Lacking strong evidence for higher temperature,  
920 it would appear that higher fO<sub>2</sub>, low pressure and water/rock ratio, and lower a<sub>SiO<sub>2</sub></sub> were key factors in  
921 producing abundant magnetite (Brearley, 2004; Yu, 2012; Vacher et al., 2018) in NWA 12563, as in WIS  
922 91600, but not in CM2 chondrites.

#### 923 **4.4 NWA 12563 and C2-ung1 chondrite parent bodies**

924 The textures in NWA 12563 give some clues as to the nature of its (grand) parent body. NWA 12563  
925 is primarily very fine grained matrix, with broken, deformed, and partially rimmed chondrule fragments. It  
926 resembles regolith material. Many clusters of ferroan olivine crystals in NWA 12563 appear to be defunct  
927 Type II porphyritic chondrules (Fig. 9d) that suffered replacement of mesostasis without pseudomorphing  
928 of the chondrule. Some dispersion of the debris is possible, as suggested by Tomeoka and Buseck (1985),  
929 but the concentrations of phosphate in some defunct chondrules indicate that P from the original glass has  
930 not been transported away. The combination of small clasts (crystal clasts of unaltered olivine and chondrule  
931 fragments) with large chondrule entities (including deformed and partly disbarred chondrule shards, olivine-  
932 cluster defunct chondrules, and gerrymandered chondrules altered after fragmentation), suggests a complex  
933 history. The olivine bar segments scattered into adjacent matrix (Fig. 9b), appear consistent with chondrule  
934 fragments plunging into an unlithified, either muddy (Bland and Travis, 2017) or powdery debris matrix. If  
935 the chondrule were deformed by shocks reducing the porosity of a lithified body to the 0.7% value, the  
936 scattering of bar fragments would have been less likely. The rare isolated olivine grains in Orgueil could  
937 similarly be attributed to some kind of regolith process. The low porosity of NWA 12563 and its fractures  
938 suggest weak shocks. Could a multi-stage history, with gentle collisions during the continuing assembly of  
939 an already warmed up body, explain the unusual mineral assemblages of NWA 12563 and WIS 91600? The  
940 incorporation of unaltered chondrule debris into a warm muddy matrix would give an unusual set of  
941 conditions for alteration of high temperature phases, possibly compatible with the preservation of olivine  
942 and the serpentinization of kamacite as well as of mesostasis.

943 The presence of saponite suggests alteration of silica-rich phases such as glass and pyroxene,  
944 higher SiO<sub>2</sub> in the amorphous precursor, or higher temperature for NWA 12563 and other C2-ung1  
945 chondrites than for CM chondrites. However, the weak 3 μm absorption and the low H abundance are not  
946 consistent with a high water content. The absence of tochilinite and cronstedtite in NWA and other C2-  
947 ung1 chondrites also suggests higher temperature. However, a higher oxygen fugacity as indicated by  
948 magnetite abundance would prevent formation of the CM chondrite phases. A combination of high  
949 temperature permitting saponite and chlorite growth coupled with exhaustion of the water and its silica  
950 content could explain the mineral assemblages in C2-ung1 chondrites.

951 NWA 12563 falls in the C2-ung1 cluster in Fig. 22 (Bells, Essebi, NWA 12563, Niger I, WIS  
952 91600, and Tagish Lake, in order of increasing  $\delta^{18}\text{O}$ ), with O isotopic compositions ranging from those of  
953  $^{18}\text{O}$ -rich CMs to near those of CI chondrites, but fall short of those of the heated CY (or Belgica)  
954 chondrites (Fig. 2). MET 00432 (Nakamura et al., 2013; Yamanobe et al., 2018) also has the matrix  
955 mineral assemblage and oxygen isotopic composition of this cluster, but few details are available. Among  
956 the C2-ung1 chondrites in Table 5, Bells appears to be the least altered, with fresh olivine in matrix (clasts  
957 a few tens of microns in size) and chondrules, only partly altered pyroxene, partly altered kamacite and  
958 very fine grained phyllosilicate (<1 nm to 200 nm) in the matrix (Brearley, 1995). It contains only 3.3%  
959  $\text{H}_2\text{O}$  (Alexander et al., 2010). Despite this, Bells has been considered to be highly altered, because it is  
960 matrix-rich (McSween, 1979) and phyllosilicate-rich (Howard et al., 2015). Bells is followed by Essebi,  
961 with partially altered kamacite and pyroxene (Zolensky et al., 1991; Buseck and Hua, 1993) and WIS  
962 91600, places third, based on kamacite pseudomorphed by serpentine though having unaltered pyroxene.  
963 NWA 12563 is like both WIS 91600 and Tagish Lake in having metal pseudomorphs, magnetite  
964 framboids, matrix saponite and also sulfide rosettes (Zolensky et al., 2002; Brearley, 2004; Tonui et al.,  
965 2014) but Tagish Lake is more altered. The gerrymandered Type I chondrules in NWA 12563 resemble  
966 chondrule cores with phyllosilicate-rich outer zones in Tagish Lake (Takayama and Tomeoka, 2012).  
967 NWA 12563 is also similar to Niger I (Table 5) but it most resembles WIS 91600.

968 WIS 91600 is distinct among C2-ung1 chondrites, having evidence of slight reheating. The criteria  
969 for a heating event after aqueous alteration include dehydration of phyllosilicates seen in low EMPA totals  
970 or structural changes observed in TEM, plus modification to light elements and organic material. The CY  
971 chondrite group that includes Belgica and meteorites like Yamato 86720 with very high  $\delta^{18}\text{O}$  values  
972 (Tomeoka et al., 1989; Clayton and Mayeda, 1999a; King et al., 2019) has experienced some form of brief  
973 heating that caused dehydration but not recrystallization. An important question is whether heating of the  
974 parent body is due to impacts or to internal radioactivity (Sugita et al., 2019). These chondrites could  
975 alternatively have been heated by passing close to the Sun (Chaumard et al., 2012). However, peak  
976 temperature estimates for some meteorites are far above those which could be achieved by solar heating  
977 (Nakato et al., 2013).

978 WIS 91600 contains dehydrated saponite in the matrix (Brearley, 2004; Tonui et al., 2014). This is  
979 interpreted to be due to a milder reheating than that which produced the CY or Belgica chondrites  
980 (Tomeoka et al., 1989; Tonui et al., 2014). The bulk H values of NWA 12563 suggest affinities with  
981 heated CM chondrites (Alexander et al. (2013) though the matrix shows no evidence for desiccation of  
982 phyllosilicates (Table 3). Olivine Cr composition data show that NWA 12563 has not been reheated above  
983 stage 3.0 (Grossman and Brearley, 2005) though this method is not sensitive to heating of short duration.  
984 It is possible that NWA 12563 represents the precursor material that was slightly heated to become WIS  
985 91600.

986 Harries and Langenhorst (2013) considered that Bells and Essebi are CM/CI transitional types,  
987 though this is inconsistent with  $\delta\text{D}$  and  $\delta^{15}\text{N}$  values. Bells is also completely different from common CC  
988 chondrites except CRs in Mg isotopic composition (Van Kooten et al., 2020). NWA 12563 differs from  
989 Bells and Essebi in having sulfide rosettes, and pseudomorphed metal (Table 5) and its  $\delta\text{D}$  is much lower  
990 than theirs, ruling out a direct relationship. There are clearly different types of C2-ung1 chondrites, in part  
991 distinguished by different oxygen isotope compositions (Greenwood et al., 2020) plus magnetite  
992 abundance or magnetic susceptibility (Fig. 22) and none of them need be closely related to any of the CM,  
993 CI and CY chondrites. Vinogradoff et al. (2018) suggested that organic evolution before accretion into  
994 meteorite parent bodies might yield diverse organic-silicate-ice mixtures which may lead to various  
995 groups of chondrites.

#### 996 4.5 NWA 12563 C2-ung1 chondrites and asteroids

997 Greenwood et al. (2020) have estimated the number of asteroids represented by meteorites and  
998 proposed two or three parent bodies for those in the C2-ung1 cluster: Tagish Lake plus Dho 1988 (see Fig.  
999 2) are assigned to one parent body, with Bells and Essebi representing either one or two bodies. This  
1000 approach is consistent with the large number of C-complex asteroids observed (Vernazza et al., 2017)  
1001 even though many of them may not have generated NEO and probable sources of meteorites. WIS 91600  
1002 and NWA 12563 were not discussed by Greenwood et al. (2020) and could represent one or two  
1003 additional parent bodies for this cluster.

1004 Asteroid spectra resembling those of carbonaceous chondrites have a wide range of shapes, with  
1005 slopes varying from negative in the visible (B class blue) to the steeply positive D class positive, with C-  
1006 complex spectra having patterns between these extremes (DeMeo et al., 2009). Ungrouped carbonaceous  
1007 chondrites are clearly of great interest for the interpretation of asteroid compositions, but quite similar  
1008 meteorites appear to correspond to different asteroid spectral types. Direct correlation of meteorite and  
1009 asteroid types may be difficult because infrared spectra are very sensitive to differences in grain size  
1010 (Vernazza et al., 2016), heterogeneity (Gilmour et al., 2019), and space weathering (Brunetto et al., 2020)  
1011 that may obscure details of the geologic history of the grandparent and parent bodies.

1012 We compared the Vis-NIR spectrum of NWA 12563 with the spectral database RELAB of Brown  
1013 University ([www.planetary.brown.edu/rehab/](http://www.planetary.brown.edu/rehab/)), using the M4AST tool at IMCCE-France  
1014 (<http://spectre.imcce.fr/m4ast/index.php/index/home>). In terms of general spectral shape and albedo, the  
1015 Vis-NIR spectrum of NWA 12563 resembles the spectra of some “bright” carbonaceous chondrites such  
1016 as CO3 Frontier Mountain 95002 (see spectra in Lantz et al., 2017). The M4AST tool also provides a best  
1017 match with the asteroid taxonomic class K (DeMeo et al. 2009).

1018  
1019 We compared the MIR-FIR reflectance spectrum of NWA 12563 with the spectra of Bells and  
1020 Essebi, and other carbonaceous chondrite spectra available from our previous studies (Brunetto et al.  
1021 2014, Lantz et al. 2015, Lantz et al. 2017, Brunetto et al. 2018, Noun et al. 2019) and from the literature  
1022 (Hanna et al., 2020, for CM WIS 91600). In Fig. 25 we show the result of this comparison in the surface  
1023 scattering range around the so called “10- $\mu\text{m}$ ” and “20- $\mu\text{m}$ ” bands (Si-O stretching and bending modes),  
1024 where we also include the spectrum of asteroid Bennu (Hamilton et al. 2019). Although the peak positions  
1025 of hydrated and anhydrous silicates are similar to those observed for other chondrites, the spectrum of  
1026 NWA 12563 is significantly different than the spectra of the other chondrites considered here, both in  
1027 terms of general shape and relative ratio between the two main peaks. It is interesting to observe how the  
1028 spectral signatures of NWA 12563 and the CO3 chondrite Frontier Mountain 95002 are different, even if  
1029 the two meteorites look similar in terms of Vis-NIR diffuse reflectance. This confirms that the MIR  
1030 spectral range is much more diagnostic than the Vis-NIR range in terms of mineral detection and  
1031 characterization. In the FIR we see that NWA12563 and WIS 91600 have similar features at 22.4  $\mu\text{m}$ , and  
1032 all the C2-ung1 analyzed have similar 28  $\mu\text{m}$  features (Fig. 25). Generally speaking, Bells and WIS 91600  
1033 are spectrally the closest meteorites to NWA12563, since they share many similarities in peak position  
1034 and overall shape.

1035  
1036  
1037 Magnetite changes spectra from red to blue (Clark et al., 2011), yet both magnetite-rich Tagish  
1038 Lake and WIS 91600 have spectra very similar to those of D-type asteroids (Hiroi et al., 2001, 2005). We

1039 find that dispersed fine-grained magnetite may have its spectral signature obscured by other phases (Fig.  
1040 23). Tagish Lake specimens vary and match those of C-, X-, Xc-, and D-type asteroids, though not B-type  
1041 (Gilmour et al., 2019). The 3  $\mu\text{m}$  absorption band of WIS 91600 suggests mild heating (Tonui et al.,  
1042 2014). CM2 chondrites have Ch- and Cgh-type VIR spectra, while the Bennu spectra (Hamilton et al.,  
1043 2019; de Léon et al., 2018) have the negative blue B-type spectra. The Bennu spectra are spectrally  
1044 “blue” in the VIR as in carbonaceous chondrites containing magnetite (Clark et al., 2011) and show  
1045 features of phyllosilicate at  $\sim 2.7$  and  $23 \mu\text{m}$ , and of magnetite at  $18$  and  $29 \mu\text{m}$  (Hamilton et al., 2019; de  
1046 Léon et al., 2018). There are similarities between Bennu and some chondrites at longer wave lengths. It is  
1047 significant that the  $18 \mu\text{m}$  feature is also seen in Orgueil (Hamilton et al., 2019) and WIS 91600 (Hanna et  
1048 al. 2020), but not in the CM2 chondrites. Thus, considering specifically magnetite, CI and C2-ung1 are  
1049 possible constituents of Bennu. Our measurement on NWA 12563 indicates that the fraction of magnetite  
1050 on an asteroid surface has to be extremely high to be detected in remote sensing data. If the fraction of  
1051 asteroid magnetite is not as high as in NWA 12563, an unambiguous in situ detection of magnetite in  
1052 localized regions of an asteroid surface would require very small footprints, difficult to be achieved by  
1053 spacecraft in typical global characterization orbits. More extensive IR spectroscopy of C2-ung1 chondrites  
1054 and more remote sensing spectra with smaller footprints on Bennu are needed to gain more information on  
1055 asteroid properties.

1056

1057  
1058 Fig. 24. Comparison of the MIR and FIR reflectance spectrum of NWA 12563 with other  
1059 carbonaceous chondrite spectra measured at SOLEIL (Brunetto et al. 2014, Lantz et al. 2015, Lantz et al.  
1060 2017, Brunetto et al. 2018, Noun et al. 2019) except for WIS 91600 (Hanna et al., 2020), and the spectrum  
1061 of asteroid Bennu (Hamilton et al. 2019). This shows the surface scattering range around the so called  
1062 “10- $\mu\text{m}$ ” band (Si-O stretching), with a vertical line at  $1000\text{ cm}^{-1}$  ( $10\ \mu\text{m}$ ) to stress the differences in main  
1063 peak position.

1064  
1065 The spectrum of NWA 12563 also looks relatively different from the spectrum of Benu, although  
1066 some general similarities can be pointed out, such as a main peak position below  $1000\text{ cm}^{-1}$  (above  $10\text{ }\mu\text{m}$ )  
1067 and a quite large width of the Si-O complex. However, the spectra of NWA 12563 and WIS 91600 in Fig.  
1068 24 give among the best matches to Benu's spectrum. Previous experiments on irradiated chondrites  
1069 (Brunetto et al. 2020) have shown that space weathering may be responsible for the spectral mismatch  
1070 between Benu and chondrites rich in hydrated matrix. We cannot exclude that irradiating NWA 12563  
1071 with low energy ions would produce a better match to Benu's spectrum, especially because space  
1072 weathering has been shown to induce a shift of the Si-O peak position towards longer wavelengths and an  
1073 increase in band width.

1074 Magnetite has been suggested to be present at the surface of Benu (Hamilton et al. 2019), but  
1075 magnetite IR features lie at higher wave lengths than the investigated spectral range for NWA 12563.  
1076

1077 Ryugu has a smoothly rising red spectrum of the Cb-type, requiring an origin in the Eulalia or  
1078 Polana family (Sugita et al., 2019; Kitazato et al., 2019). The match with the CY chondrite Y-86029 and  
1079 laboratory-heated chondrites is evidence of mild heating (Sugita et al., 2019; King et al., 2019; Kitazato et  
1080 al., 2019). The albedo of Ryugu is much darker than that of NWA 12563. Recently, the Hayabusa2  
1081 mission has measured the NIR spectrum of Ryugu's subsurface material within the artificial crater  
1082 produced by the SCI impact experiments (Arakawa et al. 2020). The spectra collected by the NIRS3  
1083 instrument (Kitazato et al. 2020) exhibit an OH feature suggesting that the subsurface material  
1084 experienced heating above  $300^\circ\text{C}$ , similar to the surface. These results support the hypothesis that primary  
1085 thermal alteration occurred due to radiogenic and/or impact heating on Ryugu's parent body.  
1086 Unfortunately, an MIR spectrum of Ryugu is not available for now.

1087 NEO sources of C2 chondrites are rubble piles, and it has recently been confirmed that Ryugu and  
1088 Benu are not monomict assemblages, but polymict like Almahata Sitta (Bischoff et al., 2010; Sugita et  
1089 al., 2019; Tatsumi et al., 2020; Dellagiustina et al, 2020). Even if there are no foreign inclusions in an  
1090 asteroid there might be fragments of different parts of the grandparent asteroid with different alteration of  
1091 metamorphic histories. Asteroids may have separated into icy crusts and zones with different amounts of  
1092 aqueous alteration (Castillo-Rodriguez and Schmidt (2010). Individual meteorites with different IR  
1093 spectra could be derived from the same main belt asteroid, if it had a cold surface layer over a uniformly  
1094 heated interior. We have concluded that NWA 12563 is a proxy for the unheated precursor to WIS 91600  
1095 and we suggest that NWA 12563 and WIS 91600 could be derived from the same grandparent body.

1096

## 1097 5. CONCLUSIONS

1098  
1099 NWA 12563 contains 14% Type I and II chondrules similar in primary mineralogy to those in  
1100 CM2 chondrites. However, its alteration differs: chondrule mesostasis contains serpentine-saponite and  
1101 chlorite, metal is pseudomorphed by serpentine; matrix contains amorphous silicate, serpentine-saponite,  
1102 plus 6.8% magnetite framboids. Cronstedtite and tochilinite are absent, possibly because of higher  $f\text{O}_2$ ,  
1103 lower  $A_{\text{SiO}_2}$ , and higher temperature than for CM2 chondrites.

1104

1105 The 86% matrix contains amorphous silicate and phyllosilicate with an  $\text{Fe}^{3+}/\Sigma\text{Fe}$  ratio measured  
1106 using STXM of  $\sim 75\%$  ( $\pm 5$ ), as in most CR chondrites, Acfer 094 and Semarkona. The amorphous silicate  
1107 phase, about half transformed to more magnesian serpentine-saponite, contains pores and pyrrhotite

1108 inclusions. It is more magnesian than amorphous silicate in CR chondrites and the Paris CM2 chondrite,  
1109 possibly because Fe was extracted to deposit magnetite.

1110 NWA 12563 has similarities to a number of ungrouped magnetite-rich and  $^{18}\text{O}$ -rich chondrites  
1111 (Bells, Essebi, NWA 12563, Niger I, WIS 91600, Tagish Lake, Dho 1988 and MET 00432). We call this  
1112  $^{18}\text{O}$ -rich cluster C2-ung1 as opposed to a second  $^{18}\text{O}$ -poor cluster of C2 ungrouped chondrites. The C2-  
1113 ung1 meteorites fall in a field on a plot of magnetic susceptibility  $\log \chi$  against  $\delta^{18}\text{O}$  that is separated from  
1114 the field for CM2 chondrites. Alteration is similar for chondrites in this cluster: there is evidence of  
1115 saponite in the matrix, they lack tochilinite and cronstedtite, and magnetite is abundant; in many sulfide  
1116 forms rosettes and metal is pseudomorphed by serpentine. Bells and Essebi differ slightly in having some  
1117 fresh metal and no sulfide rosettes, and they also have significantly different  $\delta^{13}\text{C}$ .

1118 We agree with Greenwood et al. (2020) that the C2-ung1 chondrites require several different  
1119 parent bodies. NWA 12563 and WIS 91600, however, are very similar and may represent one such body.  
1120 WIS 91600 has suffered dehydration of phyllosilicates and comes from the more heated portion of this  
1121 body.

1122 The Vis-NIR spectrum of NWA 12563 matches the asteroid taxonomic class K and resembles that  
1123 of CO3 FRO 95002. The MIR reflectance spectrum is significantly different those of other chondrites  
1124 including FRO 95002. This confirms that the MIR spectral range is much more diagnostic for mineral  
1125 detection and characterization than the Vis-NIR range. Bells and WIS 91600 are spectrally the closest  
1126 meteorites to NWA12563, since they share many similarities in peak position and overall shape. The  
1127 identification of magnetite in Bennu means that more extensive IR spectroscopy of the magnetite-rich C2-  
1128 ung1 chondrites is desirable, though an unambiguous in-situ detection of magnetite in localized regions of  
1129 an asteroid surface would require very small footprints

1130  
1131  
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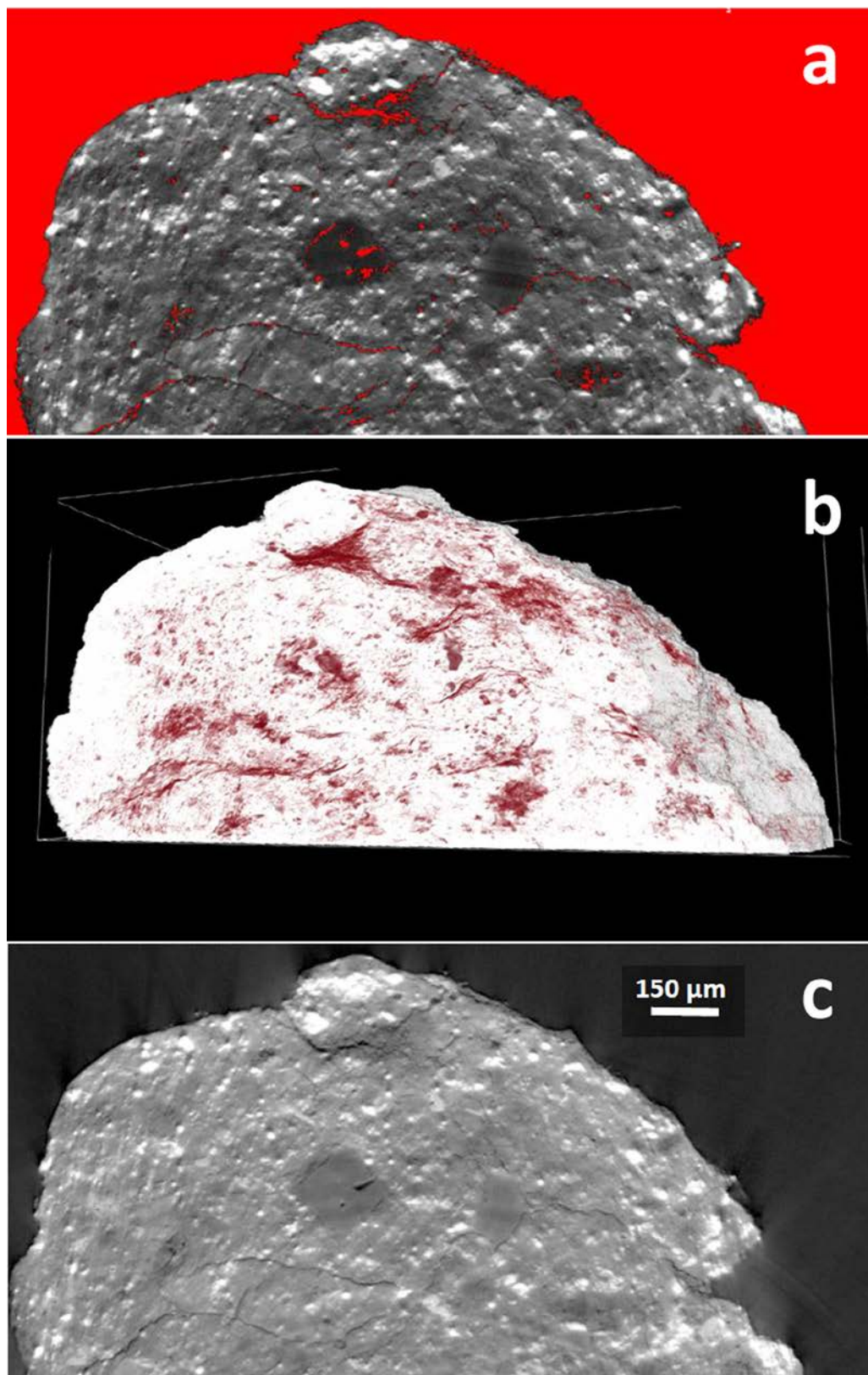
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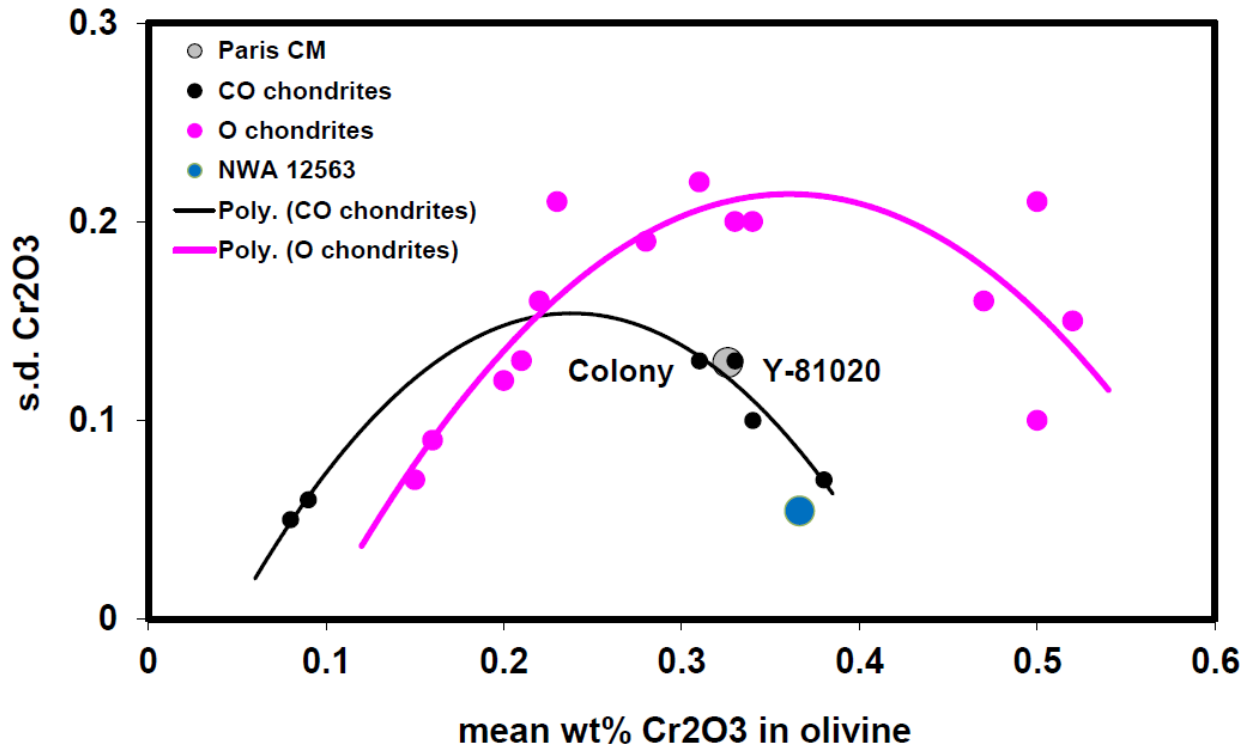


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- 1504
- 1505 **Electronic Annex** Supporting Material for "NWA 12563 and ungrouped C2 chondrites ..."
- 1506 Table S1. Analyses of olivine and pyroxene in AOA, chondrules and matrix.
- 1507 Table S2. Analyses of altered mesostasis and metal in chondrules.
- 1508 Table S3. ATEM analyses (atomic percent) of matrix silicate in NWA12563
- 1509 Figures S1-S5.



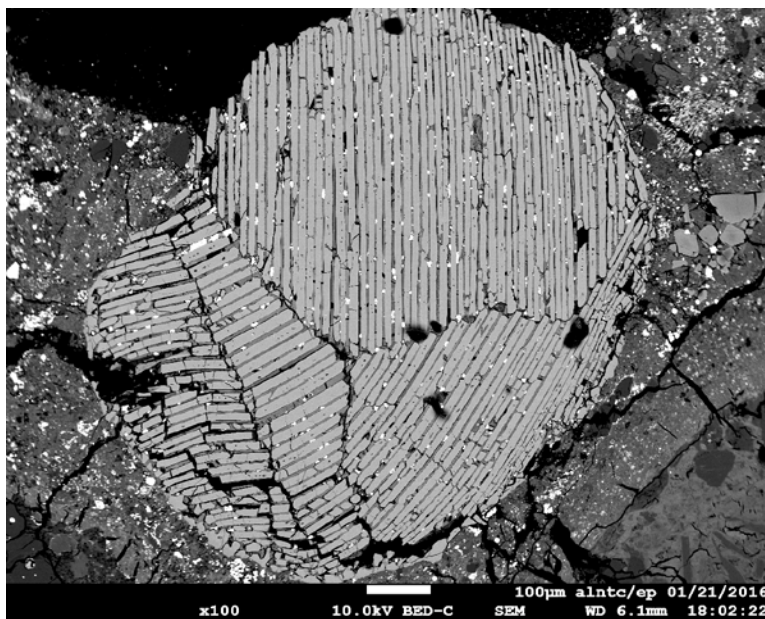
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1511 Fig. S1. X-CT images of a matrix chip showing (a) porosity in red (b) fractures in red (c) magnetite in  
1512 white.



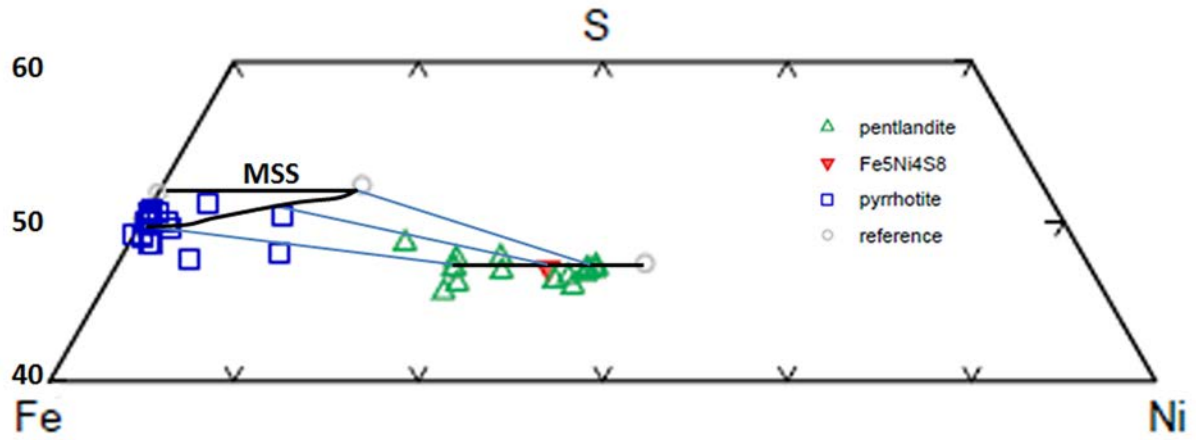
1513  
 1514 Fig. S2. Chromium diagram for ferroan olivine in NWA 12563, after Grossman and Brearley (2005). The  
 1515 lack of heterogeneity due to exsolution of chromite in olivine shows that the chondrules have not been  
 1516 significantly reheated.

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 1522 Fig. S3. Type IIA barred olivine chondrule with fractures, microfaults, and kinking.

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Fig. S4. Compositions of sulfides in atom % on part of the 200°C section of the Fe-NiS system, based on reference compositions from Craig (1973) and Harries and Langenhorst. (2013).



1528  
1529 Fig. S5. An Mg-Fe-Ni-Ca-S x-ray map of the area of the matrix of NWA 12563 in Fig. 16, showing  
1530 phyllosilicate-rich patches (green) and porous multi-phase regions with amorphous silicate and sulfide  
1531 inclusions. Width of image  $\sim 9 \mu\text{m}$ .

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