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Early Archean alteration minerals in mafic-ultramafic rocks 2

- of the Barberton greenstone belt as petrological analogues 3
- for clay mineralogy on Mars 4
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15 Abstract

Characterization of terrestrial analogue sites is critical for detection and determination of clay 16 mineralogy in remote sensing studies of Mars aimed at geological, hydrological and potentially 17 18 biological investigations. In this study, we investigate a suite of hydrothermally altered early Archean rocks from the Barberton greenstone belt (BGB) of South Africa as potential 19 petrological, mineralogical and spectral analogues to hydrothermally altered metabasalts and 20 mafic-ultramafic intrusions in the martian subsurface and impact craters. We present the first 21 22 spectral imaging measurements on exceptionally well-preserved early Archean mafic-ultramafic rocks from the BGB, with the aim of studying their clay mineralogy and spectral signatures. 23 Multiple spectral analyses were conducted on different sample textures (rock powders, crushed 24 rocks, and rock slabs) appropriate for Mars rover and remote sensing exploration. Visible/near-25 infrared (VNIR) and mid-IR reflectance spectra were acquired on particulate samples, while 26 VNIR spectral imaging data were collected on rock slabs. Mid-IR emission spectra were 27 28 measured for the rock slabs and grains. Spectral features are compared from these different

spectral techniques in order to identify the minerals present in the samples and compare 29 macroscale versus microscale detections. The measured spectra reveal absorption bands that 30 correspond to clay mineralogy of the serpentine and chlorite mineral groups, consistent with 31 petrographic observations, as well as magnetite, olivine, quartz, feldspar and Al-phyllosilicate. 32 The spectral data acquired in this study expand the reference spectra dataset for remote sensing 33 studies. The implications of this study are that rocks from early Archean greenstone belts, such 34 as those of the BGB, serve as potential clay-bearing petrological analogues for hydrothermal 35 36 environments on Mars.

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38 Keywords: Barberton greenstone belt, early Earth, Archean metabasalts, Mars petrological39 analogues

40 1. Introduction

41 In the search for life on Mars, "follow the water" has been a validated and useful approach. The main indicator of surface and subsurface groundwater on Mars is the presence of low-42 temperature alteration minerals such as phyllosilicates, opal, zeolite and sulfate (e.g., Murchie et 43 al., 2009; Carter et al., 2013; Ehlmann and Edwards, 2014). Orbital remote sensing missions and 44 45 Mars rover observations have found evidence for alteration and clay formation in multiple surface and subsurface environments including a variety of hydrothermal conditions (e.g., 46 Ehlmann et al., 2009, 2010, 2011a, 2013; Marzo et al., 2010; Squyres et al., 2012; Bishop et al., 47 2013a; Michalski et al., 2013, 2017; Bridges et al., 2015; Bristow et al., 2015). These studies 48 report in situ and hyperspectral data in support of the relatively common occurrence of (Fe, Mg)-49 rich phyllosilicates (e.g., smeetite, chlorite, and serpentine) and Al-rich phyllosilicates (kaolinite, 50 montmorillonite) on the exposed surface of Mars. Lab spectra of clay-bearing rocks from 51 various hydrothermal settings on Earth (e.g., Bishop et al., 2002, 2005, 2007; Michalski et al., 52 2006; Schiffman et al., 2006; Hamilton et al., 2008; Ehlmann et al., 2011b, 2012; Cuadros et al., 53 54 2013; Yant et al., 2018) provide important constraints on geochemical conditions, temperature, type of salts and nature of water activity on Mars. Furthermore, sub-surface mafic-ultramafic 55 hydrothermal environments on early Earth and Mars may have been very similar (e.g. Cockell et 56 al., 2006; Izawa et al., 2019; Grosch and Hazen, 2015). Investigation of Fe/Mg-rich clays from 57

seafloor sites (Cuadros et al., 2013) has led to characterization of distinct types of clavs on Mars 58 (Michalski et al., 2015; Bishop et al., 2018). Consequently, clay-bearing early Archean terrestrial 59 rocks may be useful as analogue materials to extend the visible/near-infrared (VNIR) and mid-60 infrared (mid-IR) spectral databases for remote sensing studies on orbital and rover missions at 61 Mars. It is important to point out that clay mineral formation processes on early Earth and Mars 62 may not necessarily have been the same in all geological environments, and we propose that the 63 Barberton rocks are possible petrological analogues, not necessarily direct alteration process 64 analogues. However, the early sub-surface hydrothermal environments may have been similar in 65 some parts on early Earth and Mars. 66

Bioalteration of basaltic volcanic glass is a model that has been invoked to explain the 67 68 occurrence of granular and tubular microstructures in modern and ancient volcanic glass (Thorseth et al. 1995, Fisk et al. 1998, Furnes et al. 2001). In this model, chemolithoautotrophic 69 70 micro-organisms are believed to be involved in accelerating glass dissolution for nutrients and energy producing microborings (e.g. Thorseth et al., 1995). In the modern oceanic crust, the 71 72 existence of a subseafloor biosphere is supported by a range of evidence that includes microbiological data, sulphur and carbon isotopes, and detailed reviews of this body of evidence 73 74 can be found in Orcutt et al. (2011), Edwards et al. (2011), and Thorseth (2011). Titanite microstructures in ophiolite and Archean pillow lavas have also been argued to represent early 75 76 microborings by mico-organisms thriving in the Archean subseafloor environment (Furnes et al. 2004, Banerjee et al. 2007, Staudigel et al. 2008), although the biogenicity and syngenicity of 77 these structures have been brought into question by Grosch and McLoughlin (2014). However, it 78 is not impossible, that the oceanic hydrothermal subsurface was a habitat for the earliest 79 microbial life protected from harsh UV rays on early Earth. This has important astrobiological 80 implications as it would suggest that early mafic-ultramafic hydrothermal subsurface 81 environments beneath martian impact craters may have been favorable sites for microbial life to 82 thrive (e.g. Cockell et al., 2006). As a hypothesis, the hydrothermal mafic-ultramafic subsurface 83 environments on early Earth and early Mars may both have been potentially habitable sites, 84 85 particularly given the evidence for water on early Mars (e.g., Craddock and Howard, 2002; Cockell et al., 2006, Grosch and Hazen, 2015, Izawa et al., 2019). 86

The 3.55 to 3.10 billion-year-old rocks of the early Archean Barberton greenstone belt 87 (BGB) volcano-sedimentary succession represent an exceptionally well preserved rock archive 88 89 of early Earth processes. In this study, we make the case that altered, silicified, serpentinized and clay-bearing mafic-ultramafic rock samples from the early Archean BGB, may prove to be useful 90 petrological analogues for low-temperature martian environments. Hydrogen production from 91 low-temperature alteration of ultramafic and basaltic rocks has been proposed to support early 92 microbial life in Earth's earliest subsurface environments, which may have been the case for 93 early Mars (Oze and Sharma, 2005; Grosch and Hazen, 2015). Investigation of serpentines on 94 Mars has also received attention as a potential source of H₂ and methane that could have 95 supported microbial life (Ehlmann et al., 2010; Mustard and Tarnas, 2017). Archean terrestrial 96 rock environments preserved in greenstone belts may play an important role in understanding 97 early martian subsurface environments relevant to the search for extra-terrestrial life and water. 98 Previous studies have successfully provided hyperspectral maps of altered, clay-bearing outcrops 99 in the early Archean North Pole Dome of the Pilbara Craton (Brown et al., 2004; 2007). In this 100 context, the rock sequences of the Paleoarchean BGB of South Africa provide unique martian 101 102 analogues as these rocks are exceptionally well preserved and record early Earth surface and subsurface processes that could be related to processes on early Mars as well. Geological 103 104 evidence for early Archean sedimentary delta fan deposits (Drabon et al., 2019), storm-reworked volcanic deposits (Trowera and Lowe, 2016) and spherule beds marking violent meteorite 105 impacts (Lowe et al., 2003; 2014) have all been reported from the Barberton greenstone belt. In 106 situ exploration by rovers, remote sensing studies, and meteorite evidence has indicated the 107 108 presence of metabasalts, altered gabbros, olivine-/pyroxene-bearing basalts, serpentinized picritic basalts or peridotites on Mars. 109

In this study we present spectra of several 3.4 to 3.5 billion-year-old Archean mafic-110 ultramafic greenstone belt rock samples as analogue materials that include chloritized and 111 silicified tholeiitic basalts, basaltic komatiites, serpentinized ultramafic komatiites and a felsic 112 tonalite (see Table 3). These Archean BGB samples were characterized petrologically in 113 114 previous studies (e.g. Grosch et al. 2012; Bost et al., 2013). The field setting on an outcrop scale, petrography and mineralogy of the Archean BGB samples are presented here. Mineral 115 composition data on relic igneous and clay mineral phases are discussed in context with the 116 spectroscopy results. We have analyzed VNIR and mid-IR reflectance spectra and mid-IR 117

emission spectra on these early Archean samples with the aim of using these spectra for ground 118 truthing remote sensing data and performing mineral identification and characterization of 119 120 alteration environments on Mars. Specifically, the VNIR spectra cover the spectral range collected by CRISM (Compact Reconnaissance Imaging Spectrometer for Mars, Murchie et al., 121 2009) on the Mars Reconnaissance Orbiter (MRO) and the MicrOmega instrument (Bibring et 122 al., 2017) planned for ESA's ExoMars rover. The mid-IR spectra cover the spectral range 123 measured by TES (Thermal Emission Spectrometer, Christensen et al., 2001) on the Mars Global 124 Surveyor orbiter as well as the Mini-TES spectrometer on board the Mars Exploration (MER) 125 rovers (e.g., Christensen et al., 2003; Ruff et al., 2019). VNIR reflectance spectra acquired of 126 individual rock surfaces and size fractions are compared to VNIR reflectance imaging of rock 127 surfaces. This study provides an opportunity to evaluate alteration on ancient Earth rocks and 128 129 provides spectral data sets to search for related phyllosilicates and hydrous minerals on Mars.

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2. Barberton early Earth analogue sites for Mars 131

The Paleoarchean BGB hosts some of the world's best preserved rocks of the early Archean eon 132 and some of the earliest traces of microbial life preserved in shallow silicified seafloor 133 sediments, known as cherts (Walsh, 1992;) and potentially in volcanic subseafloor environments 134 135 (Mcloughlin et al., 2012 and references therein). Exceptionally well preserved tectonic slivers of altered Mesoarchean oceanic crust and associated sediments have been identified, providing a 136 137 unique mineralogical and petrological archive of information related to the evolution of early Earth (Grosch et al., 2012). For these reasons the early Archean BGB rocks have been at the 138 139 center of a number of international scientific drilling projects (e.g., Grosch et al., 2009, Philipott et al., 2009). 140

This study focuses on rocks from the 3.55 to 3.33 Ga Onverwacht Group of the BGB (see 141 simplified geological map in Figure 1 a-c with U-Pb zircon constraints). A number of early 142 Archean mafic-ultramafic hydrothermal environments have been identified, particularly in the 143 oldest part of the BGB stratigraphy related to shallow subsurface hydrothermal activity (e.g., 144 Hanor and Ducaq, 1990; Hofmann and Harris, 2008). Retrograde alteration and sheet silicate 145 formation has also been recorded in mafic-ultramafic tectonite zones (Grosch et al., 2009; 2012). 146

147 Various metabasaltic and serpentinized ultramafic field outcrops of the Onverwacht Group are 148 shown in Figure 1 (d–h) representing examples of a wide range of early Archean low-149 temperature hydrothermal settings on early Earth potentially useful as analogue sites to martian 150 environments.

Coarse-grained cumulate metaperidotites are commonly found in the BGB. These include 151 ultramafic intrusions and mantle-derived rocks (Furnes et al., 2011, Grosch et al., 2012). An 152 153 example of such rock types is the layered serpentinized, olivine-rich meta-dunite of the 3.3 Ga Kromberg Formation of the mid-Onverwacht Group (see Figure 1d). Other examples of 154 serpentinized peridotitic mantle rocks include the ultramafic meta-dunite complex of the 3.47 Ga 155 upper Komati Formation (Dann, 2000). Metadunites and metaperidotites are considered in this 156 157 study as analogues for coarse-grained ultramafic cumulate rocks or mantle-derived rocks on Mars. The Onverwacht Group largely consists of tholeiitic metabasalt flows and pillow lavas. 158 Pillow lava metabasalts of the 3.53 billion-year-old Theespruit Formation have been 159 metamorphosed to upper greenschist/amphibolite facies conditions and are moderately deformed 160 161 (Figure 1f). These pillow metabasalts also have mica and amphibole as part of their metamorphic assemblage. Other oceanic pillow lava flows in the structurally overlying 3.47 Ga Hooggoenoeg 162 163 Formation and 3.3 Ga Kromberg Formation are exceptionally well-preserved, recording early subseafloor magmatic and hydrothermal conditions (Figure 1e). Metamorphic alteration 164 165 temperature conditions in these rocks could be as low as ca. T = 140 °C (Grosch et al. 2012). Metabasaltic flows in the Hooggenoeg, Kromberg and Mendon Formations also record evidence 166 for low-temperature ca. 80 – 150 °C subseafloor silicification beneath overlying oceanic 167 sediments (Hanor and Ducaq, 1990; Hofmann and Harris, 2008). Metabasaltic alteration 168 environments on Mars similar to these silicified basalts would be acid-sulphate leaching by 169 sulphate-rich groundwater brines at low-temperature conditions. Most of the metabasaltic flows 170 in the BGB consist of chloritized basalts (Figure 1e) and altered basaltic komatilites, possibly 171 similar to hydrothermally produced crustal chlorite in the subsurface of Mars (see Ehlmann et al., 172 2011a,). The typical major element bulk rock composition of a metabasalt from the BGB is 173 174 provided in **Table 1**. This composition is compared to those of the martian metabasalts Chesterlake and Deadwood measured by the Mars Exploration Rover (MER) Opportunity at 175 176 Endeavour Crater (Squyres et al., 2012, cf. Table 1). A strong similarity in whole-rock metabasalt composition is observed between these rocks. For this reason, further investigation of 177

the metabasalts and serpentinites from the BGB were undertaken with the goal of providinggeochemical constraints on the formation environments for related metabasalts on Mars.

The BGB is also the site of ultramafic volcanic rocks known as komatiites (Viljoen and 180 Viljoen, 1969b), which are generally considered to represent hotter mantle conditions on the 181 early Earth. These rocks can be spinifex-textured due to the presence of elongated olivine or 182 pyroxene hopper crystals. Ultramafic komatiites can be olivine- or pyroxene-rich. An olivine-183 184 spinifex komatilte from the 3.48 Ga Komati Formation of the BGB is shown in Figure 1g. Komatiites are typically altered by silicification or serpentinization. Serpentinized komatiites and 185 komatiitic basalts typically contain the alteration assemblage chlorite + antigorite + brucite. 186 Other serpentine minerals can also be present such as chrysotile. We propose that serpentinized 187 188 komatiites from the BGB region may help understand serpentinized ultramafic rocks exposed on Mars. Serpentine has been observed in mélange terrains with olivine and other phyllosilicates at 189 190 Claritas Rise and Nili Fossae, in small outcrops of the central peaks, walls or ejecta of a few impact craters, and associated with olivine and Mg-carbonate in Noachian rocks underneath a 191 192 Hesperian flow in northern Syrtis Major west of the Isidis impact basin (Ehlmann et al., 2010). Carbonate + quartz + fuchsite (Cr-rich mica) + chlorite alteration horizons (Figure 1h) have also 193 194 been identified in komatiites, basaltic komatiites, metagabbros and metaperidotites (e.g., Hanor and Ducag, 1990; Hofmann and Harris, 2008, Grosch et al., 2012). These typically occur in 195 196 retrograde tectonic shear zones (Grosch et al., 2012). Given the high CO₂ activity in martian groundwater brines (Michalski et al., 2013), these carbonate and sheet silicate BGB 197 hydrothermal alteration sites may also prove to be useful analogues for altered olivine-bearing 198 rocks on Mars such as those across the Nili Fossae region, in the flows of Syrtis Major, 199 surrounding the Isidis impact basin, and associated with smaller craters (e.g., Hamilton and 200 Christensen, 2005; Mustard et al., 2005, 2009; Bishop et al., 2013; Michalski et al., 2019). 201

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3. Methods

3.1 Sample Preparation

A list of the 11 rock samples analyzed in this study is provided in **Table 2**. This includes the BGB rock formation where the sample was collected (see Map in Figure 1) and the

207 corresponding GPS co-ordinates. Multiple samples were prepared for each rock of interest in the rock preparation lab at the University of Bergen, Norway. For each hand specimen sample, a thin 208 209 section (24 mm wide by 48 mm long and ca. 38 micron thick) was prepared for petrographic analysis and mineral identification (Grosch et al., 2012). Portions of each sample were crushed to 210 prepare coarse-grained samples by using a jaw crusher, which produced rock chips 211 approximately 0.3 - 0.8 mm in diameter. Additional samples were further pulverized into rock 212 powers using a clean agate mill. Using a rock saw, rock slabs were cut for selected samples with 213 dimensions of ca. 25 by 50 mm for the HySpex VNIR spectral scanning. 214

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216 **3.2 Sample Petrography**

Photomicrographs of representative samples and associated alteration assemblages are shown in 217 Figure 2. The metadunite sample (KM-dun) from the ca. 3.49 Ga upper Komati Formation 218 displays a coarse-grained texture consisting mainly of subhedral olivine grains replaced by low-219 220 temperature serpentine alteration minerals (Figure 2a). A two-stage alteration pattern is present with the cores of former olivine grains altered to fine-grained serpentine and magnetite that is, in 221 turn, surrounded by outer rims of chlorite (clinochlore). Relic igneous phases could not be 222 identified and the sample displays a high degree of chloritization. The serpentinized spinifex 223 224 komatiite sample (KM-spin18) consists of elongated, olivine hopper crystals as well as dendritic and plumose olivine crystal aggregates in the matrix between elongated spinifex blades (Figure 225 226 2b). The olivine hopper crystals are extensively altered mainly to chlorite and magnetite with minor serpentine, whereas the finer-grained crystals in the matrix are largely replaced by 227 228 serpentine. The altered komatiite sample (KM-2c) consists mainly of olivine replaced by brucite and antigorite (high temperature serpentine), with a rim of carbonate (dolomite) and quartz 229 230 (Figure 2c). Antigorite veins in this sample display extensional cross-fibers (Figure 2c).

The serpentinized ultramafic komatiite sample (KM-K6) contains olivine replaced by fine-grained antigorite and minor lizardite (low temperature serpentine), cross-cut by chlorite and lizardite veins (Figure 2d). Magnetite, carbonate (dolomite) and quartz are present with chlorite in the altered interstitial groundmass. The metabasaltic sample from the ca. 3.47 Ga Hooggenoeg Formation (HC3) features chloritized volcanic glass from a chilled pillow lava rim. The low-

temperature alteration assemblage in this sample consists of green chlorite plus epidote, with 236 accessory titanite and quartz (Figure 2e). Sample HC3 contains the typical subgreenschist facies 237 238 assemblage found in metabasalts from the BGB, although actinolitic amphibole is also frequently present in pillow lava metabasalts that have experienced greenschist facies conditions (Grosch et 239 al., 2012). An extensively altered metabasalt from the ca. 3.55 Ga Theespruit Formation in the 240 oldest part of the BGB (THS-4) consists mainly of coarse tabular grains of Cr-rich chlorite, 241 magnetite and sulphides (Figure 2f). An altered felsic rock from the Kaap Valley Tonalite 242 intrusion (KV1) is included as a control sample for comparison with the mafic-ultramafic rock 243 types and enables the study of sericitized plagioclase and chloritized amphibole. 244

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246 **3.3.** *Reflectance spectra of BGB samples*

Reflectance spectra were acquired using two systems. VNIR spectra were measured from 0.35-247 2.5 µm under ambient conditions on rock surfaces, coarse-grained rock fragments, and rock 248 249 powders (ground with a ball mill system to $\sim 125 \,\mu$ m) using an ASD FieldSpecPro spectrometer at the SETI Institute. Reflectance spectra were also measured on the particulate samples at 250 RELAB at Brown University from 0.3 to 50 µm as in past studies (e.g. Bishop et al., 2008). The 251 spectra are a composite of bidirectional spectra collected under ambient conditions at 5 nm 252 spectral resolution from 0.3-1.3 µm relative to Halon and biconical FTIR spectra collected using 253 a Nicolet spectrometer under a dehydrated environment (purged of H_2O overnight) at 4 cm⁻¹ 254 255 spectral resolution from 1-50 µm relative to a rough gold surface.

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257 3.4. Mid-IR emission spectra of BGB samples

Emissivity measurements were conducted in the Planetary Emissivity Laboratory (PEL) at the German Aerospace Center (DLR) in Berlin, Germany. Details on the laboratory set-up, equipment, measurements methods and calibration procedures can be found in Maturilli et al. (2019). To measure emissivity, samples were heated in two custom-designed external emissivity chambers, each of them attached to a Bruker Vertex80V FTIR spectrometer and spectra were collected from ~630-4000 cm⁻¹ (or 2.5 to 16 μ m) for these experiments. The samples were

264 heated under purged air in an emissivity chamber attached to one of the FTIR spectrometers at PEL. Each sample was positioned in an aluminum cup and heated for one hour to reach the 265 266 temperatures steps of 20 °C, 50 °C, and 100 °C. The emissivity chamber walls were watercooled to 5 °C during the sample measurements to minimize the spectral signature of the 267 chamber measured at the detector. Successively, the same samples have been heated under 268 vacuum in a second emissivity chamber, attached to another FTIR spectrometer. The whole 269 optical path between the sample and the detector was under vacuum (0.7 mbar). A thermocouple 270 was in contact with the sample surface to allow heating of the samples at the desired 271 temperatures of 50 and 100 °C. All the samples measured in emissivity have been calibrated 272 against an Acktar Fractal BlackTM blackbody, measured under exactly the same conditions as the 273 samples. 274

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276 3.5. Spectral imaging of rock slabs using HySpex

The rock slabs were scanned using the hyperspectral imaging spectrometer system (HySpex) at 277 the GFZ Potsdam (Baarstad et al., 2005). The HySpex system uses two cameras: the VNIR 1600 278 covers the range 400 - 1000 nm and the 320m-e covers the range1000 nm - 2500 nm. The 279 samples are moved through the field of view of both cameras via a precision translation stage. 280 281 This allows recording an image scene over the range 400-2500 nm. Both line scan cameras, the translation stage, and the light source (Hedler studio light with 1600 W) are mounted in a 282 283 laboratory rack that allows the use of different foreoptics and measurement distances for the HySpex cameras. For this study we selected lenses optimized for a distance of 30 cm above the 284 285 sample. The geometric correction and reflectance retrieval is carried out with an in-house preprocessing chain that combines fast Fourier transform techniques (Xie et al., 2003) with scale 286 287 invariant feature transformation (SIFT) techniques (Vedaldi et al., 2008) for image registration and a manual delineation of the reflectance panel for irradiance normalization and reflectance 288 289 retrieval.

Automated mineral mapping from large imaging spectroscopy datasets was first proposed with the USGS Tetracorder (Clark et al., 2003) and MICA (Kokaly et al. 2012; Material Identification and Characterization Algorithm) algorithms in an operational and scalable way.

293 Both algorithms use spectral reference libraries and manual expert- and user-defined feature databases (Clark et al., 2007) to characterize surface material from an image spectroscopy 294 295 dataset. This work uses the EnMAP Geological Mapper (EnGeoMAP) in its second version (Mielke et al., 2016). The main difference to the aforementioned algorithms is the automated 296 definition of characteristic absorption bands in both the unknown image spectroscopy dataset and 297 the reference library, assuming no a priori expert knowledge on the shape and position of the 298 characteristic absorption features. The absorption band retrieval is carried out using the 299 geometric hull absorption band retrieval algorithm (Mielke et al., 2015) via continuum removal 300 (Clark et al., 1987). 301

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303 **4. Results**

304 4.1. VNIR and mid-IR spectral measurements on rock powders and fractions

305 VNIR reflectance spectra measured under ambient conditions from 0.35 to 2.5 μ m of the rock powders and coarse grained material indicate the presence of alteration minerals (Figure 3). 306 307 Spectra of the KM-27 and KM-Dun samples (Figure 3b) have features consistent with serpentine including strong, narrow bands at 1.39 and 2.33 µm (e.g. King and Clark, 1989; Bishop et al., 308 309 2008). Additional features due to serpentine are present in spectra of these samples near 1.91, 1.98, 2.11, and 2.45 µm (Figure 3d). Spectra of the KM-spin 4 and KM-spin 18 samples exhibit 310 similar spectral features, although they are much weaker, indicating some serpentine is present. 311 Spectra of the THS-4 samples (Figure 3a) have bands near 1.39, 1.91, 1.98, 2.11, 2.25, 2.32 and 312 2.38 µm that are consistent with a mixture of Mg-rich chlorite and serpentine. Spectra of the 313 KV1 samples (Figure 3a) have bands near 1.41 and 2.21 µm that are consistent with Al-OH in 314 montmorillonite or mica, as well as a broad water band with a minimum at 1.91 µm consistent 315 with smectite minerals (e.g. Bishop et al., 2008). Variance in the shape of the Fe bands near 316 0.65-0.75, 0.88-0.93, and 1.03-1.13 µm, the shape of the water band near 1.9-2.0 µm and the 317 position of the OH bands near 2.25-2.5 µm are attributed to changes in the Fe/Mg-rich 318 phyllosilicates. Spectra of several Fe/Mg-clays and olivine (forsterite) are included in Figure 3d 319 for comparison. 320

VNIR spectra of the coarse-grained samples measured under a H_2O -scrubbed environment from 0.3-4 µm are shown in Figure 4a,c. These data show shifts in the OH and H_2O stretching vibrations near 1.4, 1.9, 2.2-2.3, and 2.7-3 µm. KV1 has features consistent with serpentine and quartz, while the THS-4 and KM-Dun-b spectra are darker and likely contain some basalt in addition to the chlorite and serpentine. The KM-27, KM-spin-4, and KM-spin-18 spectra are dominated by a downward slope characteristic of coatings or mixtures of grain size (e.g., Bishop, 2019).

Mid-IR reflectance spectra of the coarse-grained samples are shown in Figure 4b, d. The 328 reflectance features near 1000-1200 cm⁻¹ and from \sim 350-650 cm⁻¹ are characteristic of silicate 329 minerals (e.g. Salisbury et al., 1991; Lane and Bishop, 2019). The KV1 spectrum again appears 330 to have the strongest spectral component due to quartz based on features near 1100-1200 cm⁻¹ 331 (Figure 4b). The spectra of THS-4 and KM-Dun-b exhibit a basaltic type shape with additional 332 features consistent with chlorite (Figure 4b). The KM-spin 4 spectrum contains features due to 333 serpentine and chlorite near 1100, 1050, 870, 650, 500-470 cm⁻¹, while the KM-27 and KM-spin 334 18 spectra include features due to serpentine and chlorite, but also contain a broader band near 335 950-1000 cm⁻¹ that could be due to forsteritic olivine and a shoulder near 550 cm⁻¹ consistent with 336 magnetite (Figure 4d). 337

Mid-IR emission spectra from $\sim 600-1800 \text{ cm}^{-1}$ of the samples are presented in Figure 5. 338 For KV1 spectra were recorded for the slab, coarse grains and powder (Figure 5a). The KV1 slab 339 spectrum includes some features consistent with a mixture of basalt and chlorite, but spectra of 340 the particulate samples are more consistent with a mixture containing quartz (Figure 5b). The 341 quartz grains in this sample are apparently more dominant in the particulate samples. Emission 342 spectra of quartz include features due to Si-O stretching vibrations near 1100-1200 cm⁻¹ and 343 bending vibrations near 500 cm⁻¹ (e.g. Wenrich and Christensen, 1996; Christensen et al., 2000; 344 Land and Bishop, 2019). Basalt emission spectra in this region exhibit a broader band in the 345 silicate stretching region that has been important in modeling the spectra of Mars (e.g. 346 347 Christensen et al., 2001). Spectra of plagioclase (albite) and hornblende are also shown for comparison as they were found in petrologic analyses and could be present at lower abundances 348 349 based on the emission spectra. The THS-4 emission spectra are dominated by features due to the chlorite mineral chamosite (Figure 5b) as also observed in the VNIR spectra. The chlorite-type 350

bands near 950, 1030, and 1070 cm⁻¹ are observed in both grain sizes of this sample, although the water band near1600 cm⁻¹ is more pronounced in the spectrum of the fine-grained sample, as expected. Emission spectra of KM-27 and KM-Dun-b (Figure 5c) include features consistent with serpentine and chamosite, but also contain a broader emission features spanning 900-1200 cm⁻¹ that is consistent with basalt. Emission spectra of KM-spin-4 and KM-spin-18 (Figure 5d) are even more characteristic of basalt, but could have smaller components of serpentine or olivine, as found in the petrologic analyses.

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359 4.2. Spectral imaging of rock slabs

False color composite images of the rock slabs are shown in Figure 6a,b and point spectra of 360 locations marked on Figure 6a,b are shown in Figure 6f,g together with mineral reference spectra 361 from the United States Geological Survey (USGS) digital spectral library (Clark et al., 2007). 362 The extracted characteristic absorption bands of the unknown image pixel spectrum and the 363 reference library standards are compared via a weighted fitting technique similar to the 364 Tetracorder (Clark et al., 2003) and MICA (Kokaly et al. 2012) algorithms. Figure 6c,d,e shows 365 366 a best weighted fit material map using sensor specific thresholds, which are lower than the user defined threshold values considering the high Signal to Noise Performance (SNR) of the HySpex 367 368 system (Baarstad et al., 2005).

Samples such as KMK-6 and KR1 are too dark (low overall albedo) to be analyzed with 369 reflectance values below the dark cellular-rubber background, as shown in Figure 6f. The 370 hyperspectral scanning of the samples indicates the presence of clay minerals, in particular 371 chlorite and serpentine. The reflectance spectra of samples KM-Dun and THS-4 indicate the 372 presence of both chlorite and serpentine, consistent with the petrographic identification from thin 373 section microscopy (see Figure 2). VNIR spectral imaging of sample KM-2c revealed the 374 presence of antigorite veins, also consistent with the petrographic observations (Figure 2c). 375 Collectively, the HySpex VNIR imaging method tested here succeeded in mapping chlorite and 376 serpentine clay mineralogy identified in our samples through petrographic thin-section analyses, 377 and bulk VNIR and mid-IR spectral measurements. 378

379 Spectra of a few surface spots from the HySpex imaging spectrometer are compared with bulk spectra of the crushed samples in Figure 7. These data illustrate how the bulk spectra 380 381 contain features present in one or two spot spectra that are focused on individual mineral grains in the rock. For example, the surface spot measured on rock slab KM 2C contains features due to 382 chlorite that are also found in the spectra of the THS-4 bulk grains sample. The HC3A surface 383 spot spectrum contains a broad Fe band near 0.9-1.2 µm that is consistent with olivine, bands 384 near 2.0, 2.25 and 2.35 µm that are consistent with chamosite, and a band at 2.20 µm due to Al-385 OH in phyllosilicates. Spectra of the KV1 surface spot and KV1 bulk grains also contain 386 phyllosilicate OH features from 2.2 to 2.35 µm, but show variations in the abundance of the 387 chamosite and Al-smectite. The KM-Dun spectra exhibit basalt-type signatures and the basalt 388 band is more clearly defined in the surface spot spectrum. A small shift in the wavelength of the 389 390 OH combination band near 2.33-2.35 µm represents changes in the Fe/Mg abundance in the chlorite measured by each technique. 391

392

393 **5. Discussion**

Based on orbital data, a very wide range of aqueous environments of Noachian age has been 394 recognized on Mars, and their occurrence implies that hydrothermal sub-surface alteration of 395 396 mafic-ultramafic rocks was widespread during the Noachian Period on ancient Mars (Wray et al., 2009; Bishop et al., 2013a, 2018). It is likely that some of the low-temperature clay minerals 397 398 observed on Mars are due to hydrothermal processes, particularly those associated with large impact craters. Smectite, kaolinite, and illite are common clay minerals on Mars that form at low 399 400 temperatures below ~100 °C. However, moderate to higher temperature clay minerals (~100-400 °C) such as serpentine, chlorite, and talc have been confirmed in Leighton Crater and other sites 401 402 using hyperspectral analysis in crater ejecta, knobby terrains, or as part of discontinuous layers in craters or valley walls of the Nili Fossae region (Amador et al., 2018; Brown et al., 2020). 403 404 Furthermore, olivine (precursor to serpentine) fractured rock units have been identified in deep regions of craters in the Nili Fossae region (e.g. Bramble et al., 2017) of similar age to early 405 Archean rocks on Earth and serpentine has commonly been found as a possible higher 406 temperature mineral in other regions of Mars (e.g. Ehlmann et al., 2010). Amador et al., (2018) 407 have argued that serpentinization processes were pervasive in the early part of Mars' history as it 408

was more geologically active then. Hence early Earth and early Mars may have had similarserpentinization processes in their early geological histories.

Even though not all alteration processes would have been the same on early Earth and 411 Mars, the current study proposes that some subsurface mafic-ultramafic hydrothermal 412 413 environments on ancient Mars and early Archean Earth may have been similar. In this context, the rock sequences of the Paleoarchean Barberton greenstone belt of South Africa provide 414 415 unique martian analogues as these rocks are exceptionally well-preserved and record early Earth (and perhaps martian-type) subsurface hydrothermal/alteration processes. *In-situ* exploration by 416 rovers, remote sensing studies, and meteorite evidence has indicated the presence of altered 417 gabbros, olivine-/pyroxene-bearing metabasalts and possible serpentinites on Mars (e.g., Ehlman 418 419 and Edwards, 2014; Ruff et al., 2019; Hamilton et al., 2019; Murchie et al., 2019; McSween, 2015). We present selected 3.5 to 3.3 billion-year-old Archean greenstone belt analogue samples 420 421 that include altered tholeiitic metabasalts and serpentinized ultramafic rocks. Spectroscopic measurements were conducted on different scales, namely bulk analyses on rock fragments and 422 423 rock powders, and microscale analyses on rock slabs by HySpex in-situ analysis. VNIR reflectance, mid-IR emission and VNIR spectral imaging have successfully identified serpentine 424 425 and chlorite alteration mineralogy irrespective of scale or material form. The petrography and mineralogy of the samples were analyzed in terms of relic igneous phases and clay mineral 426 427 alteration and are in good agreement with spectral identification of minerals in the samples. We expanded past studies by acquiring VNIR reflectance and mid-IR emission spectra on these early 428 Archean samples for comparison with orbital spectral data of Mars. 429

The HySpex instrument hyperspectral data used to 'map' the mineralogy of the rock 430 431 samples has proven to be useful in mineral identification and the results compare well with bulk spectroscopic analyses. This technique has potential to provide useful information for both 432 433 terrestrial studies and studies seeking to understand hydrothermal alteration of rocks from current or future Mars missions. The power of imaging spectroscopy is that it coordinates textural and 434 435 spatial information with mineralogy, information that is not gained from bulk analysis of rock powders or rock chips. This spatial and textural-related data are needed to demonstrate that a 436 437 clay-bearing deposit on Mars may be hydrothermal in origin and also that a terrestrial deposit may indeed be a useful Mars analogue. These kinds of analyses will be particularly useful in 438

preparing for the micromega analyses to be collected by the ExoMars rover (Bibring et al., 2017). The relationships between alteration phases, primary igneous minerals and textural characteristics are critical for piecing together the process history of metamorphosed terrestrial and extra-terrestrial mafic and ultramafic rocks. In theory, *in situ* imaging spectroscopy of relevant rock surfaces has the power to aid in this type of direct comparative study. A HySpextype instrument such as micromega attached to a Mars rover could also provide useful *in situ* scans of rock texture, mineralogy, and the alteration processes producing clay minerals.

446

447 6. Implications

The Archean Barberton greenstone belt rocks hold significant potential for future petrological 448 analogue studies for Mars. Remote sensing hyperspectral studies on Mars have revealed 449 evidence for hydrothermal activity involving, not only chloritization, but also carbonate 450 formation in altered mafic-ultramafic rocks (e.g., Viviano et al. 2013; Ehlmann and Edwards, 451 452 2014; Lane et al., 1997; Hamilton et al., 2019; Brown et al., 2020), sulphates (Bishop et al., 2013a) and hematite on Mars (Christensen et al., 2000, 2001; Lane et al., 2002). A number of 453 454 early Archean rocks from the Barberton greenstone belt could serve as hyperspectral petrological analogues for martian environments in future analogue studies: Banded Iron Formations (BIF) 455 456 for hematite and magnetite layers; hydrothermal and sedimentary barite units for sulphate analogues; as well as carbonitized and silicified mafic-ultramafic rocks for siderite, dolomite and 457 458 calcite hyperspectral studies. Silicified metabasalts and serpentinites of the Barberton greenstone belt may also contribute to understanding acid leaching processes on Mars (c.f. Yant et al., 459 460 2016). The upcoming Mars2020 rover analyses of Jezero crater will investigate phyllosilicate and carbonate-bearing outcrops in delta sediments, as well as basaltic bedrock containing olivine, 461 462 chlorite, and serpentine (e.g., Goudge et al. 2017, Bramble et al., 2017; Amador et al., 2018; Brown et al., 2020). If chlorite- and serpentine-bearing rocks are encountered by Mars2020, the 463 464 rock textures and spectral analyses in this study will provide ground truth for understanding the potential formation conditions for those rocks. 465

466

468 Figures

Figure 1. Simplified geological map of the Barberton greenstone belt (BGB). (a-b) Geographical location of the BGB on the boundary between South Africa and Swaziland. (c). Geological map with U-Pb age constraints of various rock types of the Onverwacht Group of the Barberton Supergroup (see Grosch et al., 2011 and references therein). (d - h) Various field outcrops showing altered serpentinized ultramafic rocks and metabasalts as potential analogue sites for hydrothermal alteration of crust on Mars.

475

Figure 2. Petrographic images of relevant mafic and ultramafic rock types with associated lowtemperature alteration assemblages (white scale bar is 200 μ m). (a) KM-Dun = chloritized metadunite, (b) serpentinized olivine spinifex komatiite, (c) KM-2c = serpentinized komatiite with serpentine extensional veins (d) KM-K6 = serpentinized peridotitic komatiite with minor alteration veins (e) = HC3 chlorite – epidote – quartz assemblage in metabasaltic pillow lava chilled margin (f) altered komatiitic basalt with Cr- chlorite, serpentine and accessory opaques.

482

Figure 3. VNIR reflectance spectra from 0.35 to 2.5 μm measured with the ASD spectrometer
under ambient conditions. (a) KV1 and THS-4 spectra, (b) KM-27 and Km-Dun-b spectra, (c)
KM-spin 4 and KM-spin 18 spectra, (d) fine-grained BGB samples compared with minerals from
the Bishop library (e.g. Bishop, 2019) and synthetic olivine (Fo90) from Isaacson et al. (2014).
Spectral features due to electronic or vibrational absorptions are marked on the plots to facilitate
comparison of the BGB samples with each other and their mineral components.

489

490 *Figure 4*.

Reflectance spectra across the VNIR and mid-IR range measured under an H₂O- and CO₂-purged 491 environment. Spectra of the coarse-grained samples KV1, THS-4, and ZKM-Dun-b are shown in 492 (a) from 0.3-4 μ m as a function of wavelength and (b) from 1400 to 300 cm⁻¹ as a function of 493 wavenumber, while spectra of KM-27, KM-spin-4, and KM-spin-18 are shown in (c) and (d). 494 495 Reflectance spectra of basalt and selected minerals (Bishop, 2019) are provided for comparison. 496 Spectra of the clay minerals chamosite (Fe-rich chlorite, solid line), clinochlore (Mg-rich chlorite, dotted line), lizardite (serpentine, solid line), chrysotile (serpentine, dotted line) and 497 montmorillonite SAz-1 (Al-rich smectite) are from Bishop et al. (2008), and the basalt spectrum 498 of sample G2 from Bishop et al. (2002) is described in Harloff et al. (2001). Vertical lines mark 499 features due to minerals in the spectra. 500

502 *Figure 5.*

503 Mid-IR emission spectra of BGB samples (a) KV1, (b) THS-4, (c) KM-Dun-b and KM-27, and (d) KM-spin-4 and KM-spin-18. Spectra are shown for multiple sample textures for samples 504 505 KV1 and THS-4 to illustrate the influence of grain size and surface type on the spectra. Emission spectra of selected minerals and basalt are shown for comparison. Emission spectra of the 506 507 chamosite (Fe-rich chlorite) is from Bishop et al. (2008), the basalt spectrum is sample G2 from Bishop et al. (2002) that is described in Harloff et al. (2001), the serpentine, hornblende and 508 forsterite are from Lane and Bishop (2019), and the quartz and albite spectra are from the ASU 509 library (Christensen et al., 2000). Vertical lines mark features in the BGB spectra associated with 510 the rock and mineral spectra. 511

512

Figure 6. VNIR spectral imaging of rock slabs KMK-6, KD-Pyrox, KR1, KM-2C, THS4, KM-DUN and HC3A. (a) color maps of the rock slabs using R: 640 nm, G: 549 nm, B: 458 nm, (b) color maps of the rock slabs using R: 2222 nm, G: 890 nm, B: 553 nm, (c) maps of correlation values, (d) maps of the best fit plus gradient, (e) maps of the best fit, (f) VNIR spectra collected from regions on the rock slabs marked in (a) and (b) that were selected to highlight the observed clay mineralogy in the samples, and (g) lab spectra of minerals from the USGS spectral library for comparison.

520

521 *Figure 7.* Spectra of a few surface spots from the HySpex imaging spectrometer compared in the same diagram with bulk spectra of the selected crushed samples.

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524

525

526 **Table Captions**

527 *Table 1.* Whole rock compositions in wt.% element oxides of a typical BGB metabasalt528 compared to altered metabasalts on Mars.

529

Table 2. List of samples in the study and their locations in the BGB stratigraphy. For further
information on rock age see Grosch et al., (2011)

532

533 *Table 3.* List of samples and analytical technique

534

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Figure 3







Figure 6



Figure 7



		•	
	Barberton	^(a) Mars	^(a) Mars
	BG-196	Chesterlake	Deadwood
	metabasalt	metabasalt	metabasalt
	(this study)		
SiO ₂	45.23	45.5	44
TiO ₂	1.04	1.09	0.98
AI_2O_3	12.18	8.8	8.3
FeOt	20.01	20.10	19.4
MnO	0.31	0.48	0.22
MgO	9.96	8.8	5.7
CaO	8.1	6.8	6.7
Na ₂ O	0.68	2.7	2.2
K ₂ 0	0.81	0.41	0.62
P_2O_5	0.06	1	0.78
L.O.I	2.2	na	na
SUM	100.58	na	na

Table 1. Whole rock compositions of metabasalts

^(a) rock compositional data from Squyres et al., 2012

Acronym	Rock type/Mineralogy
THS-4	Greenschist facies pillow metabasite
KR1	Serpentinized komatiitic basalt
KM-2C	Serpentine veined komatiitic basalt
KMK-6	Serpentinized massive komatiite
KM-Spin 4	Serpentinized Olivine Spinifex Komatiite
KM-Spin 18	Serpentinized Olivine Spinifex Komatiite
KD-Pyrox	Altered Pyroxene Komatiite
KM-Dun-b	Serpentinized Metadunite
KM-27	Serpentinized Massive Komatiite
HC3A	Greenschist facies thoeliitic basalt
KV1	Altered felsic tonalite

Table 2. List of analoge samples used in the study and there $\mathbf{l}_{^{l}}$

Formation	AGE (Ga)	S coordinates	E coordinates
Middle Theespruit Formation	3.53	25°59.854'	030°50.191'
Lowermost Komati Formation	3.48	25°59.586	030°51.094'
Lowermost Komati Formation	3.48	25°59.582'	030°51.098'
Lower Komati Formation	3.48	25°59.557'	030°51.093'
Lower Komati Formation	3.48	25°59.569'	030°51.088'
Lower Komati Formation	3.48	25°59.319'	030°51.507'
Middle Komati Formation	3.48	25°59.264'	030°51.564'
Upper Komati Formation	3.48	25°58.912'	030°50.677'
Upper Komati Formation	3.48	25°58.837'	030°52.035'
Lower Hooggenoeg Formation	3.47	25°56.712'	030°53.064'
TTG pluton	3.23	25°44.398'	030°59.902'
*			

Table 3. Spectral measurements of samples

Sample ID	JB sample #	sample type	mineralogy	HySpex imagery	ASD	RELAB	Emission
KV1	JB1570	rock slab	plagioclase, quartz	Х	Х		Х
KV1	JB1571	coarse grains (~1 mm)	hornblende &		Х	Х	Х
KV1	JB1572	powder (~<125 μm)	chlorite		Х		Х
THS-4		rock slab	chlorite &	Х			
THS-4	JB1573	coarse grains (~1 mm)	fuchsite mica		Х	Х	Х
THS-4	JB1574	powder (~<125 μm)			Х		Х
KM-Dun		rock slab	serpentine & magnetite	e X			
KM-Dun-b-(D1 JB1676	coarse grains (~1 mm)			Х	Х	Х
KM-Dun-b-(D1 JB1677	powder (~<125 μm)			Х		
KM-27	JB1678	coarse grains (~1 mm)	serpentine & chlorite		Х	Х	Х
KM-27	JB1679	powder (~<125 μm)			Х		
KM-spin-4	JB1680	coarse grains (~1 mm)	serpentine, magnetite		х	Х	Х
KM-spin-4	JB1681	powder (~<125 μm)	olivine		Х		
KM-spin-18	JB1682	coarse grains (~1 mm)	serpentine, magnetite		х	х	Х
KM-spin-18	JB1683	powder (~<125 μm)	olivine		Х		
KMK-6		rock slab	serpentine, peridotite	х			
KD-pyrox		rock slab	serpentine	х			
KR1		rock slab	serpentine, chlorite	Х			
KM-2C		rock slab	serpentine,	Х			
HC3A		rock slab	chlorite, epidote, quart	z X			