

Origin of carbonate-bearing rocks in Jezero crater: Implications for ancient habitability in subsurface environments

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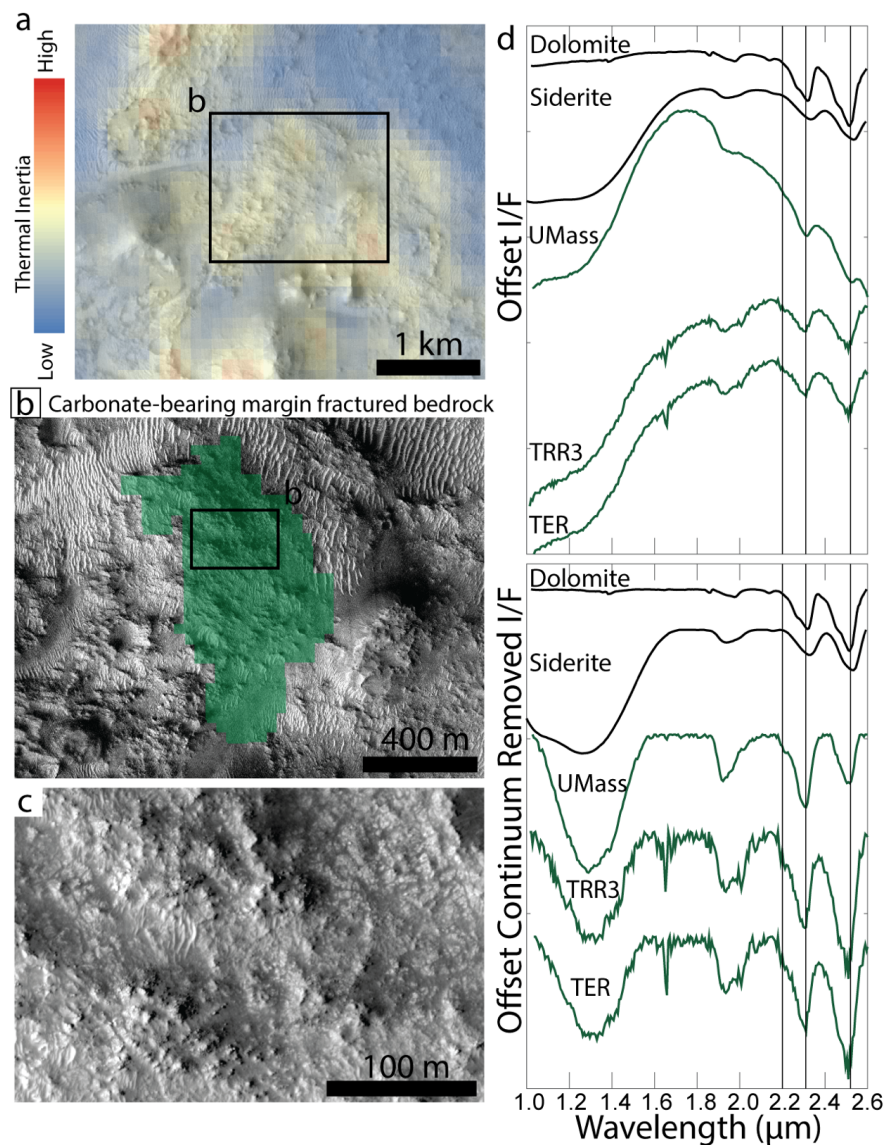
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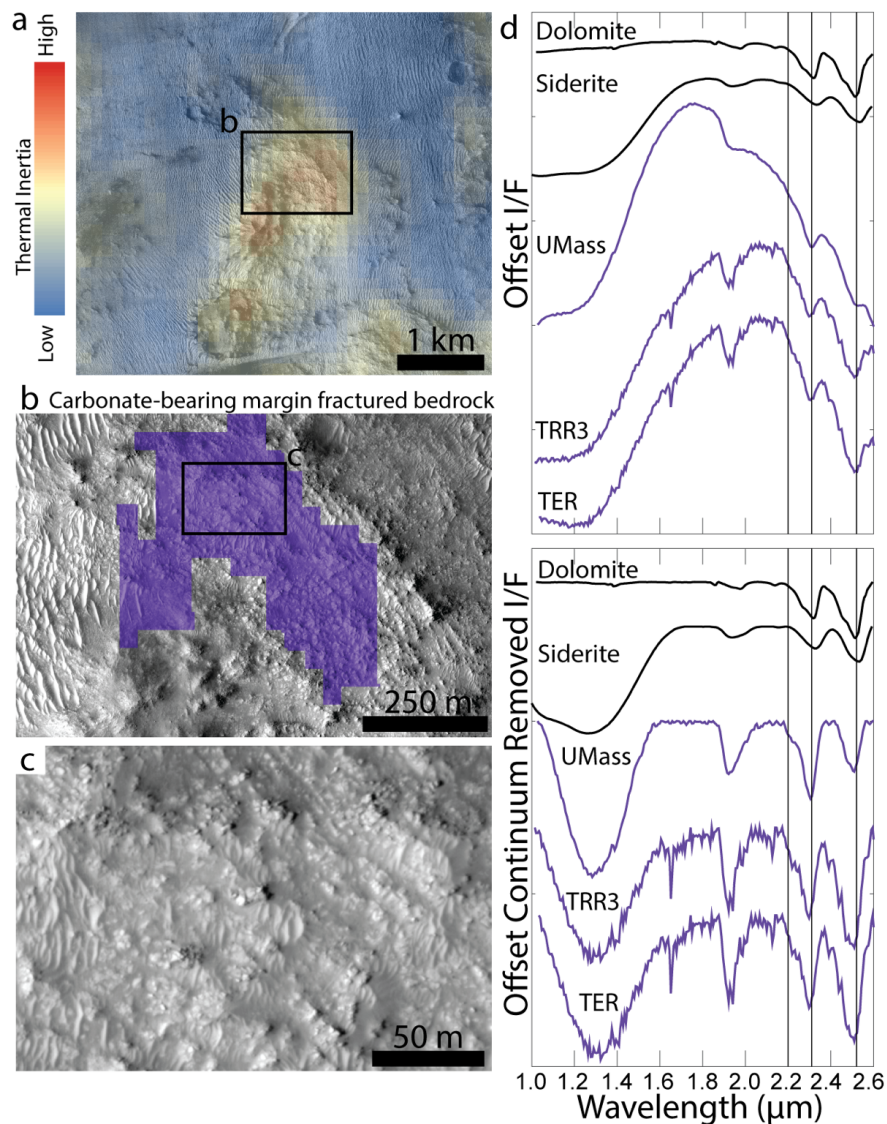
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Abstract

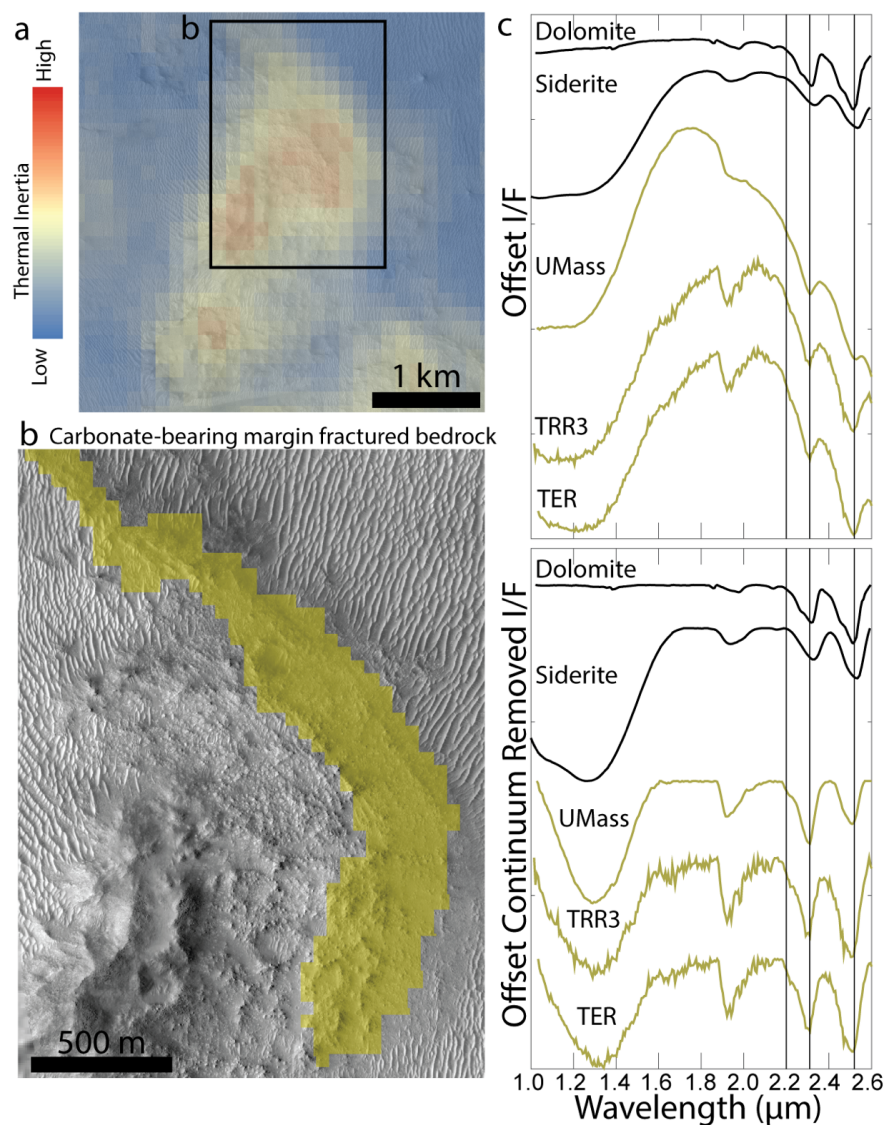
Jezero crater, an ancient lake basin that is the landing site of the Mars 2020 Perseverance rover, contains a carbonate-bearing rock unit termed the margin fractured unit. Some of the carbonates in these rocks may have formed in a fluviolacustrine environment and therefore could preserve biosignatures of paleolake-inhabiting lifeforms. Here we evaluate whether these margin fractured unit carbonates formed as authigenic precipitates in a fluviolacustrine environment or via alteration of primary minerals by groundwater. We integrate thermal inertia measurements from the Thermal Emission Imaging System (THEMIS), spectral analyses from the Compact Reconnaissance Imaging Spectrometer for Mars (CRISM), examination of stratigraphic relationships in Jezero crater using High Resolution Science Experiment (HiRISE) and Context Camera (CTX) images and digital elevation models. We also compare the Jezero crater results to observations from the Curiosity rover in Gale crater. We find that margin fractured bedrock with the deepest visible-to-near-infrared carbonate absorptions also has exceptionally high thermal inertia and thickness relative to other carbonate-bearing units in Jezero crater, consistent with enhanced cementation and crystallization by groundwater. Our results indicate that it is equally likely that carbonates in Jezero crater formed via alteration of primary minerals by alkaline groundwater rather than as authigenic precipitates in a fluviolacustrine environment. Jezero crater may have hosted ancient subsurface habitable environments related to these groundwaters, where life-sustaining redox energy was generated by water-rock interactions. The Mars 2020 Perseverance rover could encounter biosignatures preserved from this carbonate-forming environment, whether it was fluviolacustrine or in the subsurface.



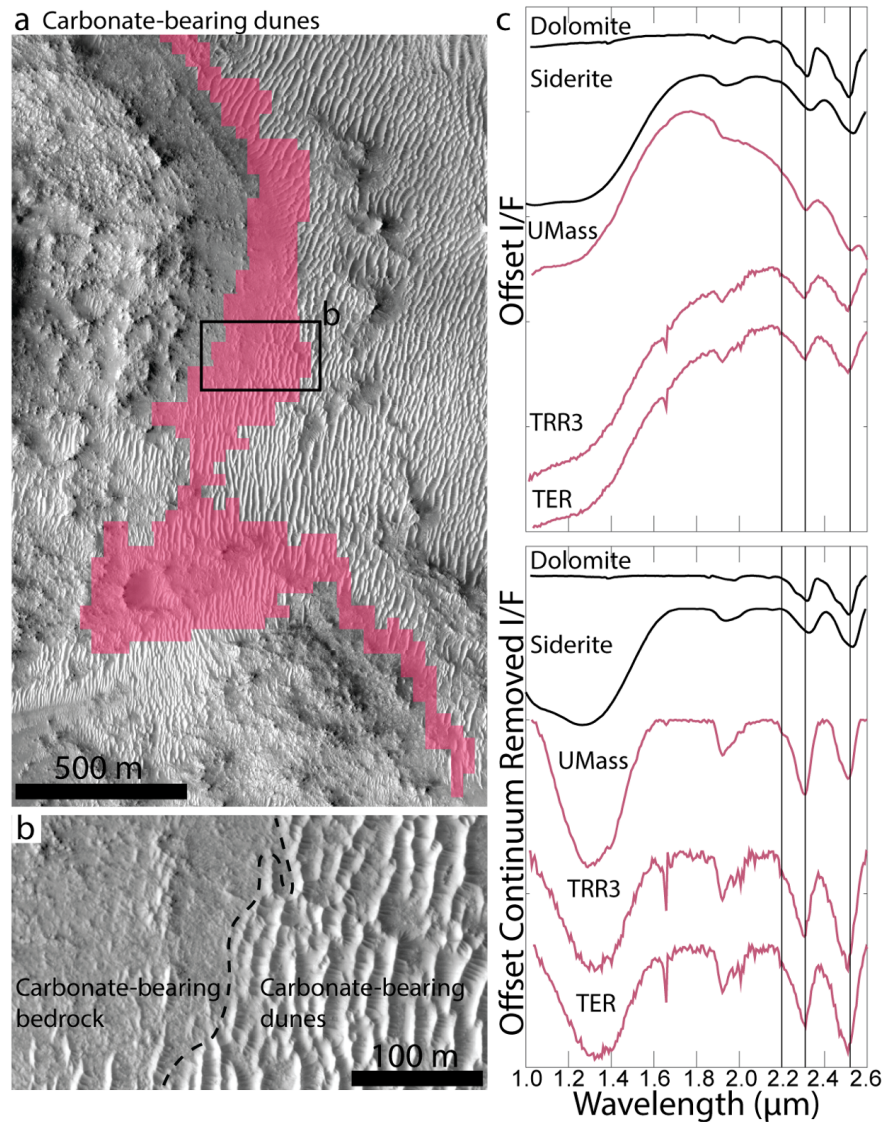
Supplementary Figure 1 | Carbonate and phyllosilicate-bearing bedrock in the margin fractured unit. (a) THEMIS qualitative thermal inertia map of region of interest. (b) CRISM pixels (dark green) for spectra shown in (e). These CRISM pixels cover light-toned fractured bedrock with notably high thermal inertia. (c) Enlarged view of light-toned margin-fractured unit bedrock and contact with dark-toned delta blocky unit material. (d) UMass, TRR3, And TER CRISM spectra of dark green pixels shown in (b). The spectral and geologic interpretation is the same as is described in Figure 4.



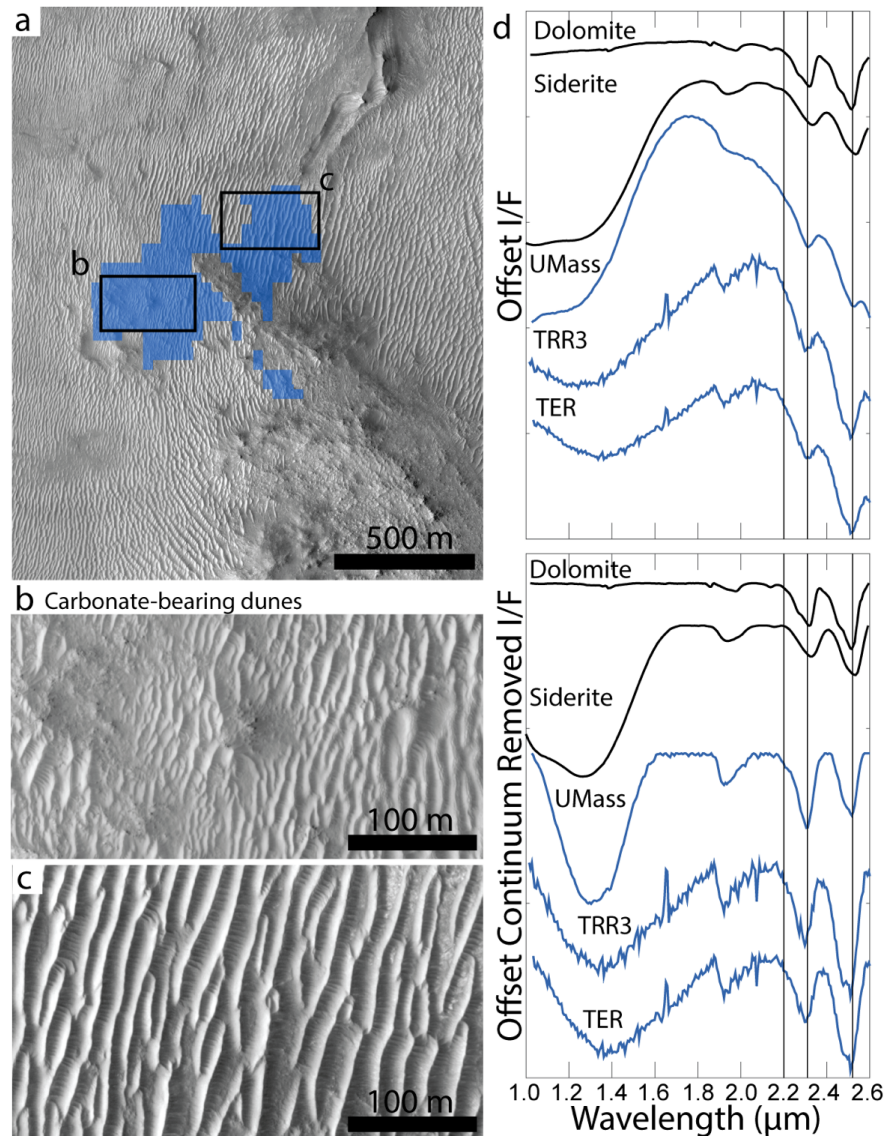
Supplementary Figure 2 | Carbonate and phyllosilicate-bearing bedrock in the margin fractured unit. (a) THEMIS qualitative thermal inertia map of region of interest. (b) CRISM pixels (purple) for spectra shown in (d). These CRISM pixels cover light-toned fractured bedrock with notably high thermal inertia. (c) Enlarged view of light-toned margin-fractured unit bedrock and contact with dark-toned delta blocky unit material. (d) UMass, TRR3, And TER CRISM spectra of purple pixels shown in (b). The spectral and geologic interpretation is the same as is described in Figure 4.



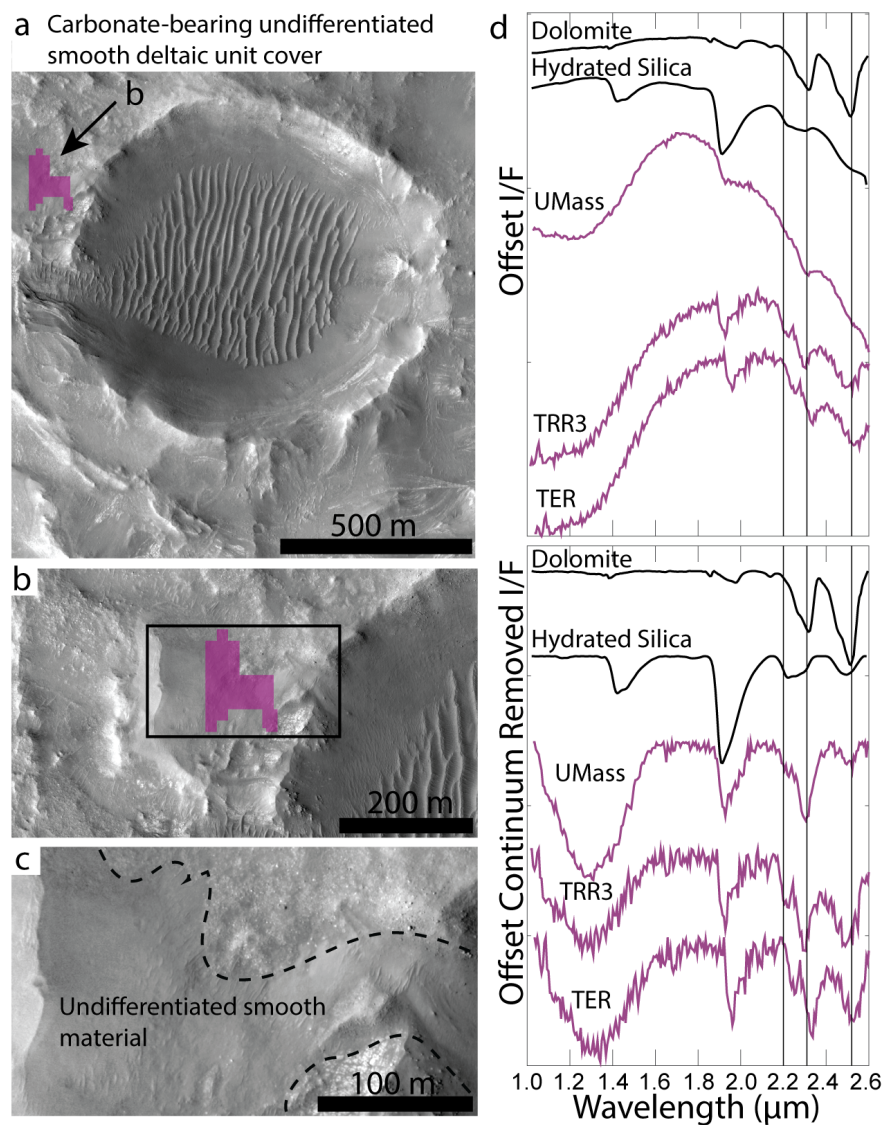
Supplementary Figure 3 | Carbonate and phyllosilicate-bearing bedrock in the margin fractured unit. (a) THEMIS qualitative thermal inertia map of region of interest. (b) CRISM pixels (yellow) for spectra shown in (c). These CRISM pixels cover light-toned fractured bedrock with notably high thermal inertia. (c) UMass, TRR3, And TER CRISM spectra of yellow pixels shown in (b). The spectral and geologic interpretation is the same as is described in Figure 4.



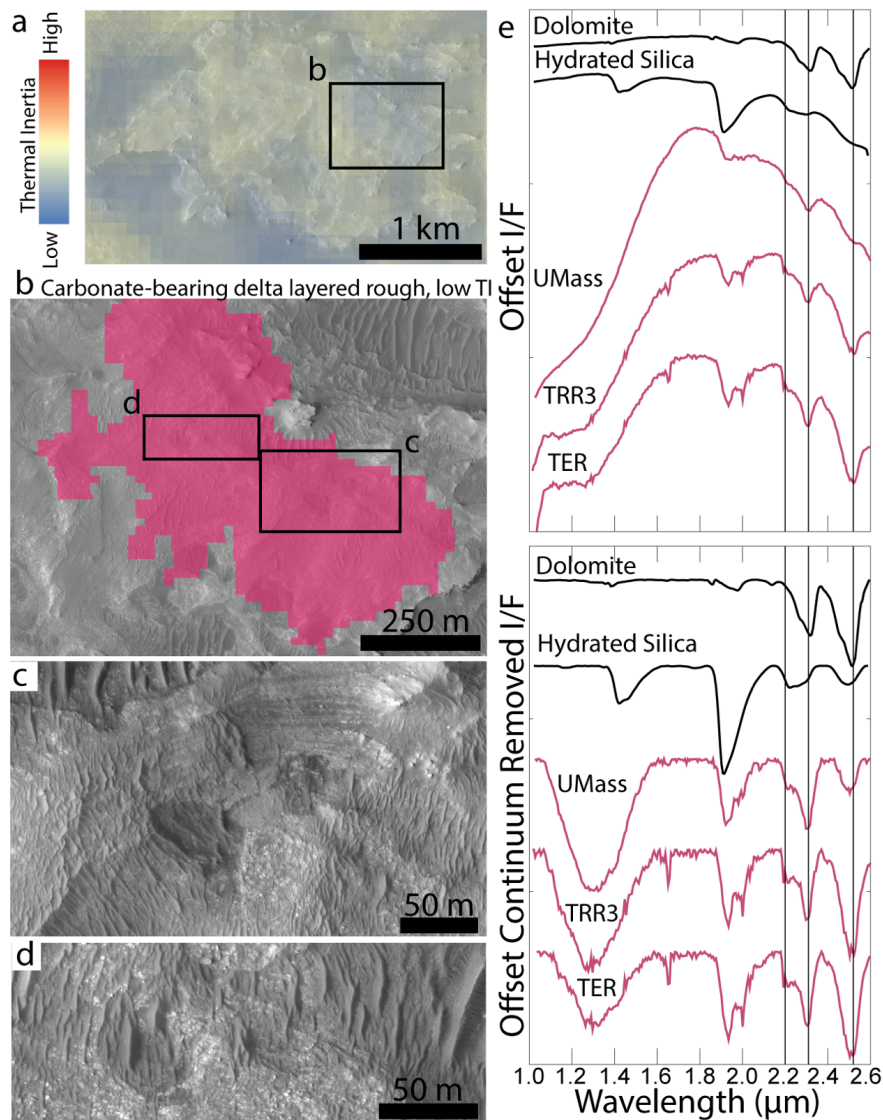
Supplementary Figure 4 | Carbonate and phyllosilicate-bearing aeolian bedforms proximal to margin fractured unit. (a) CRISM pixels (pink) for the spectra shown in (c). These pixels cover aeolian bedforms proximal to the margin fractured bedrock shown in Figure S3. (b) Enlarged views of carbonate and phyllosilicate-bearing aeolian bedforms where they contact the proximal carbonate and phyllosilicate-bearing margin fractured bedrock described in Figure S3. (c) UMass, TRR3, And TER CRISM spectra from pink pixels shown in (a). The spectral interpretation is the same as described in Figure 4. This aeolian material is therefore interpreted to derive from the proximal margin fractured unit bedrock.



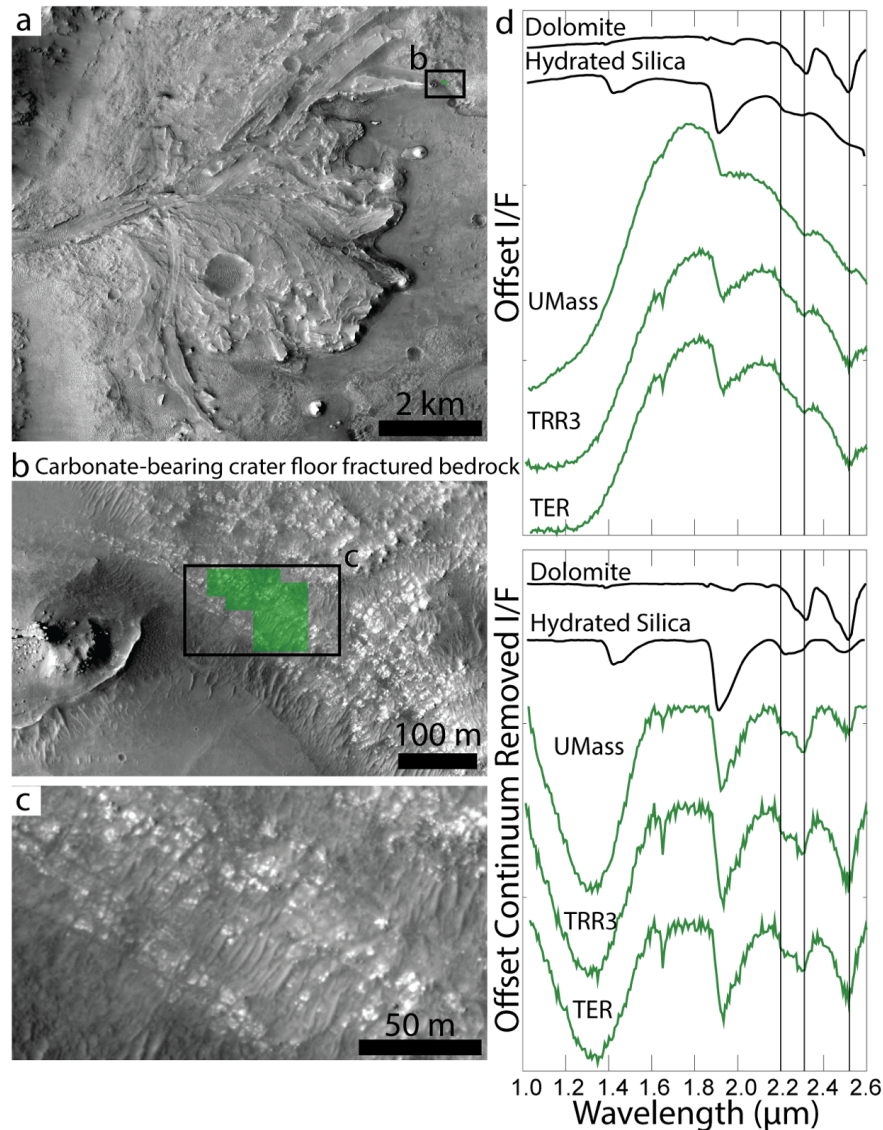
Supplementary Figure 5 | Carbonate and phyllosilicate-bearing aeolian bedforms proximal to margin fractured unit. (a) CRISM pixels (blue) for the spectra shown in (d). These pixels cover aeolian bedforms proximal to the margin fractured bedrock shown in Figure S3. (b-c) Enlarged views of carbonate and phyllosilicate-bearing aeolian bedforms where they contact the proximal carbonate and phyllosilicate-bearing margin fractured bedrock described in Figure S3. (d) UMass, TRR3, And TER CRISM spectra from blue pixels shown in (a). The spectral interpretation is the same as described in Figure 4. This aeolian material is therefore interpreted to derive from the proximal margin fractured unit bedrock.



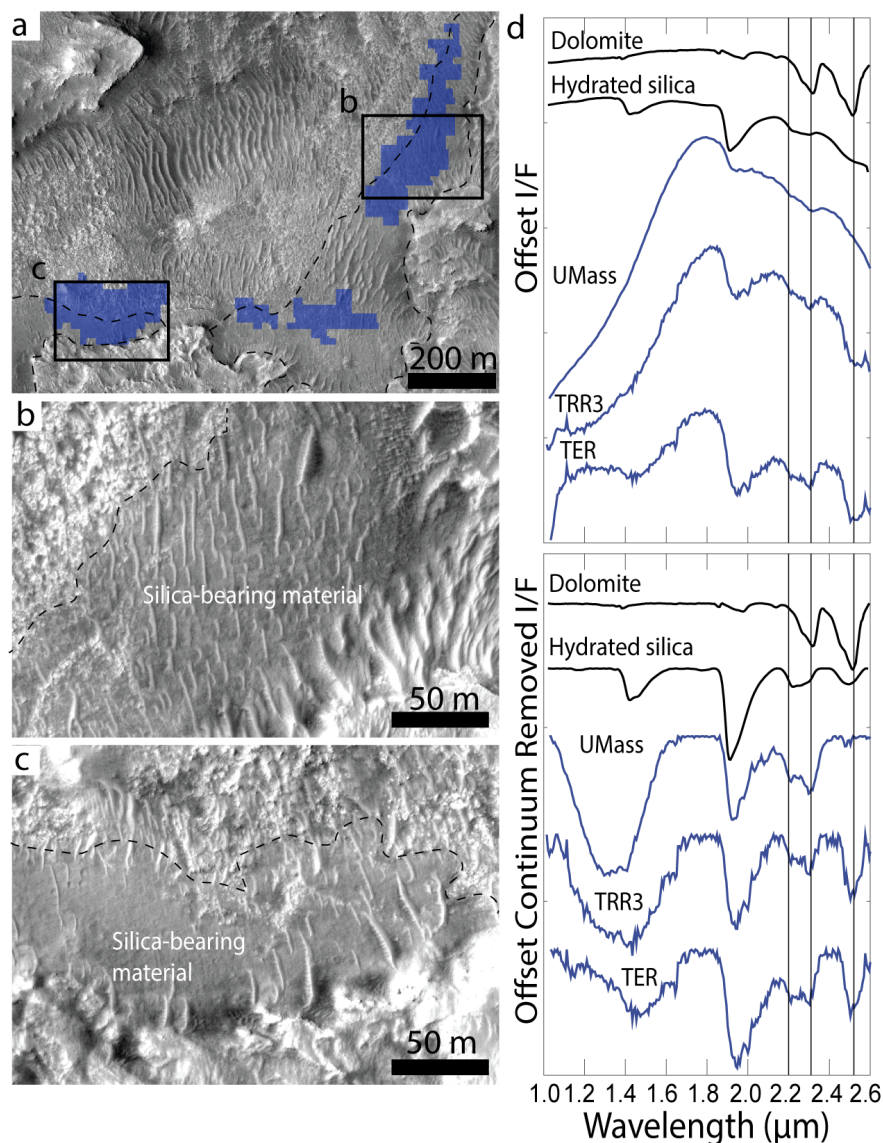
Supplementary Figure 6 | Carbonate and phyllosilicate-bearing rock in the western Jezero delta undifferentiated smooth unit. (a) Regional view of reported compositional detections from CRISM pixels shown in blue. (b) CRISM pixels (purple) for spectra shown in (c), which cover primarily dark-toned bedrock in the delta undifferentiated smooth unit. (c) UMass, TRR3, And TER CRISM spectra from blue pixels shown in (a & b). The spectral interpretation is the same as described in Figure 8, but is present on a different deltaic unit.



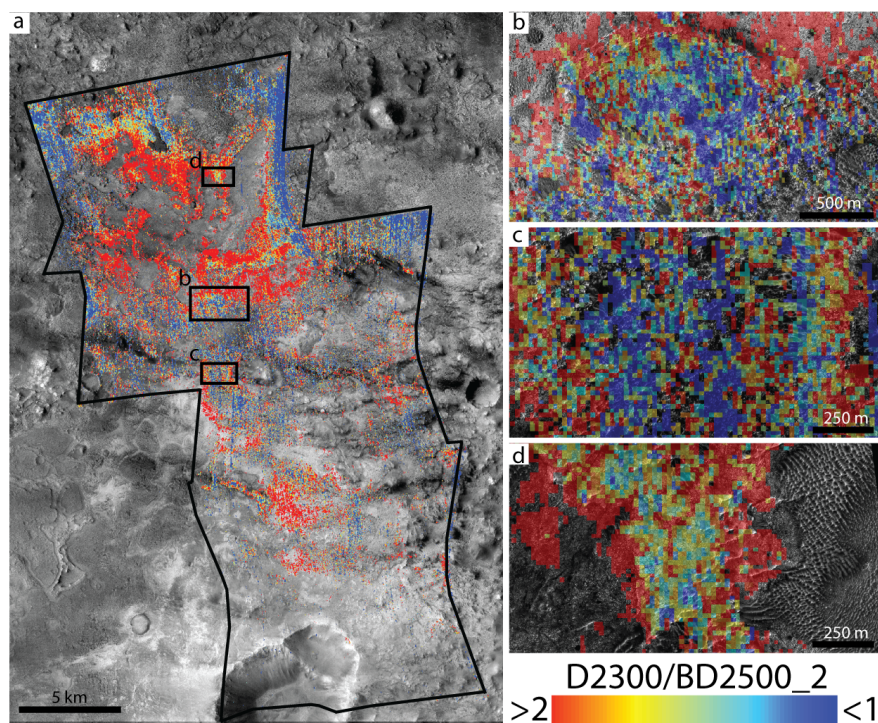
Supplementary Figure 7 | Carbonate and phyllosilicate-bearing rock in the northern Jezero delta layered rough unit with low thermal inertia. (a) THEMIS qualitative thermal inertia values over region of interest. (b) Regional view of reported compositional detections from CRISM pixels shown in pink, which cover the northern delta low thermal inertia surface component. (c) Enlarged view of layered rocks in low thermal inertia northern delta, covered by aeolian bedforms. (d) Enlarged view of light-toned rocks in low thermal inertia northern delta, covered by aeolian bedforms. (e) UMass, TRR3, And TER CRISM spectra from pink pixels shown in (b). The spectral interpretation is the same as described in Figure 8, but is present on a different deltaic unit.



Supplementary Figure 8 | Carbonate, hydrated silica, phyllosilicate, and olivine-bearing rock in the crater floor fractured 2 unit. (a) Overview figure showing regional context of extracted CRISM spectra. (b) CRISM pixels (green) for spectra in (d) are shown in green. These pixels cover the crater floor fractured 2 unit. (c) Enlarged view of crater floor fractured 2 unit bedrock overlain by dark-toned material and aeolian bedforms. (d) UMass, TRR3, And TER CRISM spectra of pixels shown in (b). The absorption features are consistent with those described in Figure 12, and the compositional interpretation is the same. The geologic context is similar to that described in Figure 14, where dark-toned unconsolidated material and aeolian bedforms comprise the majority of the area covered by the CRISM data. This dark-toned unconsolidated material may be locally derived, in which case it has the same composition as the bedrock. Alternatively, this material may be the silica-rich material—described by Tarnas et al. (2019) and reported in Figures 15, 16, and S9 in this work—spectrally mixing with olivine, carbonate, and phyllosilicate-bearing bedrock in the crater floor fractured 2 unit.



Supplementary Figure 9 | Smooth dark-toned silica-bearing material overlying crater floor fractured unit. (a) CRISM pixels (blue) for the spectra shown in (d). These pixels cover the smooth dark-toned material reported to be silica-bearing by Tarnas et al. (2019). (b) Enlarged view of smooth dark-toned silica-bearing material overlying the crater floor fractured unit. (c) Another enlarged view of smooth dark-toned silica-bearing material overlying the crater floor fractured unit. (d) UMass, TRR3, and TER CRISM spectra from blue pixels shown in (a). These spectra are consistent with hydrated silica and the Jezero crater hydrated silica detections reported in this material in different locations by Tarnas et al. (2019).



Συμπλεγμενταρψ Φιγυρε 10 | Ωιδεσπρεαδ ουτσροπς οφ ςαρβονατες ιν Νιλι Φοσσας ωιτη λωω 2.3/2.5 μm band depth ratios. (a) Ratio of the 2.3 and 2.5 μm absorptions (D2300/BD2500_2) in CRISM pixels of images FRT000028BA, FRT00009D96, FRT0000C256, and FRT000095FE with BD2500_2 ≥ 0.005 . The combined extent of all four CRISM images is shown as a black outline. This shows an even more widespread distribution of carbonates with similar 2.3/2.5 μm band depth ratios as are found in the Jezero crater margin fractured unit. All pixels shown here are associated with the ROB unit. (b-d) Enlarged views of boxes shown in (a).

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Origin of carbonate-bearing rocks in Jezero crater: Implications for ancient habitability in subsurface environments

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Abstract

Jezero crater, an ancient lake basin that is the landing site of the Mars 2020 Perseverance rover, contains a carbonate-bearing rock unit termed the margin fractured unit. Some of the carbonates in these rocks may have formed in a fluviolacustrine environment and therefore could preserve biosignatures of paleolake-inhabiting lifeforms. Here we evaluate whether these

margin fractured unit carbonates formed as authigenic precipitates in a fluviolacustrine environment or via alteration of primary minerals by groundwater. We integrate thermal inertia measurements from the Thermal Emission Imaging System (THEMIS), spectral analyses from the Compact Reconnaissance Imaging Spectrometer for Mars (CRISM), examination of stratigraphic relationships in Jezero crater using High Resolution Science Experiment (HiRISE) and Context Camera (CTX) images and digital elevation models. We also compare the Jezero crater results to observations from the Curiosity rover in Gale crater. We find that margin fractured bedrock with the deepest visible-to-near-infrared carbonate absorptions also has exceptionally high thermal inertia and thickness relative to other carbonate-bearing units in Jezero crater, consistent with enhanced cementation and crystallization by groundwater. Our results indicate that it is equally likely that carbonates in Jezero crater formed via alteration of primary minerals by alkaline groundwater rather than as authigenic precipitates in a fluviolacustrine environment. Jezero crater may have hosted ancient subsurface habitable environments related to these groundwaters, where life-sustaining redox energy was generated by water-rock interactions. The Mars 2020 Perseverance rover could encounter biosignatures preserved from this carbonate-forming environment, whether it was fluviolacustrine or in the subsurface.

Plain Language Summary

Spacecraft orbiting Mars can measure the composition of rocks that make up its surface. Understanding rock composition allows us to interpret past environmental conditions on Mars, including their likelihood to be habitable. Using data acquired from orbit, researchers have

found carbonate minerals in Jezero crater and the surrounding region—called the Nili Fossae region. Jezero crater is the landing site of NASA’s Mars 2020 Perseverance rover and once contained a lake. The discovery of carbonates is exciting because on Earth they sometimes form in habitable environments and preserve fossils. In this study, we show that it is equally likely that carbonates in Jezero crater formed by groundwater rather than forming in Jezero crater’s ancient lake. We also find that contact with past groundwater likely changed the physical properties of some rocks in Jezero crater that contain carbonate, which made the carbonate composition more obvious from orbit. Many carbonates that form by groundwater on Earth preserve fossils from underground habitable environments, where organisms obtain energy from chemicals dissolved in water rather than from sunlight. We propose that fossils from similar underground ancient habitable environments could exist in the Jezero carbonates, exposed at the surface by rock erosion over billions of years.

1.0 Introduction

The Nili Fossae region lies immediately west of the ~3.96 Gyr old (Werner, 2008) Isidis impact structure on Mars and north of the Late Hesperian age (Greely and Guest, 1987) Syrtis Major volcano. Widespread occurrences of olivine were first reported in Nili Fossae using data from the Thermal Emission Spectrometer (TES; Christensen et al., 2001) aboard the Mars Global Surveyor spacecraft (Hoefen et al., 2003). Nili Fossae is one of the most mineralogically diverse exposures of the martian surface, with evidence for olivine, pyroxene (Mustard et al., 2009), phyllosilicates (Ehlmann et al., 2009; Mustard et al., 2008), carbonates (Ehlmann et al., 2008b), and sulfates (Ehlmann and Mustard, 2012; Quinn and Ehlmann, 2019) observed in orbital

spectroscopic data. Here we focus primarily on the olivine-bearing unit in Jezero crater and the surrounding Nili Fossae region, which we term the regional olivine-bearing unit (ROB unit).

Previously proposed origins for the $\sim 3.82 \pm 0.07$ Gyr age (Mandon et al., 2020) ROB unit include an ultramafic ashfall deposit (Kremer et al., 2019; Mandon et al., 2020), an impact spherule deposit (Palumbo and Head, 2018) from an impact younger than Isidis (Mandon et al., 2020), or detrital sedimentary rock (Rogers et al., 2018). Spectral data interpreted by modeling suggest ~ 20 - 25% modal abundance of olivine in this unit (Edwards and Ehlmann, 2015) and average olivine grain sizes of 0.5 mm or larger (Brown et al., 2020). It has been variably altered to carbonate, phyllosilicate(s), and possibly hydrated silica (Bramble et al., 2017; Brown et al., 2010; Ehlmann et al., 2008b; Goudge et al., 2015; Mandon et al., 2020; Tarnas et al., 2019; Viviano et al., 2013) and has a fractured-to-rubby texture (Bramble et al., 2017; Kremer et al., 2019). Its compositional and thermal inertia properties are similar to the Algonquin olivine-rich tephra deposit and Comanche carbonate investigated by NASA's Spirit rover (Ruff et al., 2019), as well as other high thermal inertia clastic rocks on Mars (Rogers et al., 2018). The ROB unit drapes into the Jezero crater rim and is present on the modern-day crater floor (Goudge et al., 2015; Kremer et al., 2019; Mandon et al., 2020; Sun and Stack, 2020). It is also incised by rivers that flowed into Jezero crater (Goudge et al., 2015) and deposited the deltaic outcrops there (Fassett and Head, 2005; Goudge et al., 2017; Schon et al., 2012). The Jezero delta deposits contain low calcium pyroxene, olivine, phyllosilicate(s), carbonate, and possible hydrated silica (Ehlmann et al., 2009; Goudge et al., 2018, 2015; Horgan et al., 2020a), much of which is likely detrital (Goudge et al., 2015), but some of which may be authigenic (Bristow and Milliken, 2011).

Recent photogeologic mapping has defined 15 bedrock units in western Jezero crater (Stack et al., 2020). Five of these units are carbonate-bearing: the delta blocky unit, delta truncated curvilinear layered unit, delta layered rough unit, crater floor fractured 2 unit, and margin fractured unit (Figure 1). Goudge et al., (2015) and Ehlmann et al., (2008) grouped the carbonate-bearing units exposed along the inner margin (margin fractured) and on the crater floor (crater floor fractured 2) of Jezero, and correlated these deposits within the crater with the surrounding ROB unit, due to their morphological and spectral similarities. Later work presented the evidence consistent with a fluviolacustrine origin for carbonates in the margin fractured unit, highlighting in particular the association of deeper 2.5 μm absorptions and low 2.3/2.5 μm band depth ratios with the approximate maximum height of the Jezero paleolake (Horgan et al., 2020a). These authors also highlighted possible spectral trends with respect to distance from the inlet valley. An unknown amount of fluviolacustrine sediment was deposited along the crater rim during highstanding periods of the Jezero paleolake, and it is possible that these sediments were carbonate-bearing and have since been lithified, as is common along margins of alkaline lakes on Earth (Horgan et al., 2020a). The observation of strong VNIR spectral signals of carbonate near the former margin of the Jezero paleolake is therefore consistent with formation of these carbonates in a near-shore fluviolacustrine environment. Here we compare the evidence for the carbonate formation scenario presented by Goudge et al., (2015) and Ehlmann et al., (2008)—where carbonate formation in the margin fractured unit and ROB occurs via the same processes—versus the fluviolacustrine carbonate formation scenario proposed by Horgan et al., (2020a).

Determining the conditions of carbonate formation in Jezero crater is key to evaluating past habitability and potential for biosignature preservation in rocks explored by the Perseverance rover. This must be undertaken using orbital data until the Perseverance rover encounters carbonate-bearing rocks, which it will be guided to via interpretations of orbital data. The Mars Science Laboratory Curiosity rover recently groundtruthed CRISM hematite detections in Gale crater (Fraeman et al., 2020b), providing valuable context for how interpretations of orbitally-acquired ~12-32 m/pixel VNIR hyperspectral images (Fraeman et al., 2013) compare to the true compositional nature of rocks determined in-situ (David et al., 2020; Fraeman et al., 2020a, 2020b; Frydenvang et al., 2020; Horgan et al., 2020b; Jacob et al., 2020; L'Haridon et al., 2020; McAdam et al., 2020; Morris et al., 2020; Rampe et al., 2020; Thompson et al., 2020). To evaluate the most likely origin scenarios for carbonate-bearing rocks in Jezero crater, we integrated data from the Compact Reconnaissance Imaging Spectrometer for Mars (CRISM; Murchie et al., 2007), images and digital elevation models (DEMs; Beyer et al., 2018) from the High Resolution Imaging Science Experiment (HiRISE; McEwen et al., 2007) and Context Camera (CTX; Malin et al., 2007), as well as data from the Thermal Emission Imaging System (THEMIS; Christensen et al., 2004).

2.0 Background

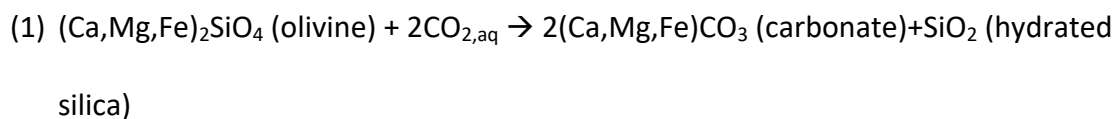
Phyllosilicates in the Nili Fossae region were first discovered using data from the Observatoire pour la Minéralogie, l'Eau, les Glaces et l'Activité (OMEGA; Bibring et al., 2004) instrument aboard Mars Express (Poulet et al., 2005), indicating that this landscape preserves evidence of past aqueous activity. Higher spatial resolution data from CRISM aboard the Mars

Reconnaissance Orbiter (MRO) found evidence for phyllosilicates (Ehlmann et al., 2009; Mustard et al., 2008), carbonates (Ehlmann et al., 2008b), sulfates (Ehlmann and Mustard, 2012; Quinn and Ehlmann, 2019), and mafic minerals (Mustard et al., 2009) in Nili Fossae and showed that these minerals were correlated with specific units in a regionally extensive stratigraphy. From oldest to youngest, the Nili Fossae region includes a basement sequence, an olivine-bearing unit, a capping unit, and a sulfate-bearing unit (Ehlmann and Mustard, 2012). The basement sequence has been further subdivided into the Stratified Basement Unit, Blue Fractured Unit, Mixed Lithology Plains Unit, LCP-bearing Plateaus Unit, and Fe/Mg-smectite-bearing Mounds Unit (Scheller and Ehlmann, 2020) and contains well-preserved martian crust older than 3.9 Gyr (Mustard et al., 2009), which has no comparably well-preserved analog in Earth's geologic record. The basement sequence contains widespread low calcium pyroxene and phyllosilicate VNIR spectral signatures (Scheller and Ehlmann, 2020), as well as outcrops containing kaolinite (Bramble et al., 2017; Ehlmann et al., 2009) and hydrated silica (Tarnas et al., 2019). The capping unit is associated with spectra consistent with mafic minerals (Bramble et al., 2017; Mustard et al., 2009) and glass (Cannon et al., 2017) and is interpreted to be volcanoclastic (Bramble et al., 2017; Hundal et al., 2020). The sulfate-bearing unit records evidence for standing bodies of water, diagenesis under acidic conditions, and fluvial activity (Quinn and Ehlmann, 2019).

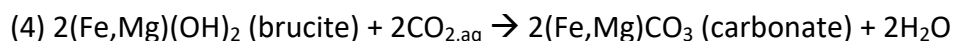
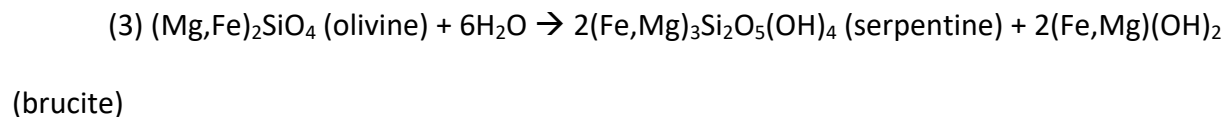
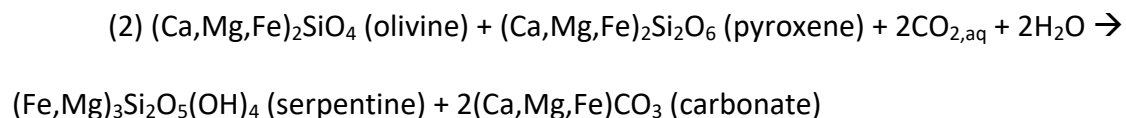
Alteration hypotheses for the ROB unit, including the exposures on the Jezero crater floor and margin, include groundwater percolation under geothermal gradient-driven temperatures (Ehlmann et al., 2009), alteration under a thick CO₂ atmosphere (Edwards and Ehlmann, 2015; van Berk and Fu, 2011), simultaneous serpentinization and carbonation that

occurred during dehydration of the underlying phyllosilicate basement (Brown et al., 2010; Viviano et al., 2013), carbonation of partially serpentinized olivine (Viviano et al., 2013), alteration under the conditions of the modern day martian atmosphere (Kelemen et al., 2020a), low temperature water-rock alteration (Edwards and Ehlmann, 2015), low temperature carbonate rind formation (Ehlmann et al., 2008b; Jull et al., 1988), or surface weathering to carbonate (Ehlmann et al., 2008b). Distinguishing between these hypotheses using orbital data alone is challenging, but mineral identifications and geologic context observed by the Mars 2020 Perseverance rover should be sufficient to constrain the causes of carbonation and other alteration, as mineral assemblages and their distributions can be used to constrain reaction conditions.

Carbonation of Fe, Mg, and Ca-bearing silicate material on Earth—including mafic and ultramafic rock—occurs via water-rock-gas reactions with high CO₂ activity/partial pressure in both high and low temperature and pressure conditions. Direct carbonation of olivine occurs via the reaction



while carbonation of pyroxene and olivine can occur via the reaction series



176 (5) $2(\text{Fe,Mg})_3\text{Si}_2\text{O}_5(\text{OH})_4$ (serpentine) + $3\text{CO}_{2,\text{aq}} \rightarrow \text{Mg}_3\text{Si}_4\text{O}_{10}(\text{OH})_2$ (talc) + $3(\text{Mg,Fe})\text{CO}_3$
177 (carbonate) + $5\text{H}_2\text{O}$

178 (6) $\text{Mg}_3\text{Si}_4\text{O}_{10}(\text{OH})_2$ (talc) + $3\text{CO}_{2,\text{aq}} \rightarrow 3(\text{Mg,Fe})\text{CO}_3$ (carbonate) + 4SiO_2 (hydrated silica) +
179 $6\text{H}_2\text{O}$

180 which forms carbonate, hydrated silica, serpentine, talc, and brucite (Kelemen et al., 2011;
181 Matter and Kelemen, 2009; Moody, 1976; Oelkers et al., 2008). If reactions 1-6 all proceed to
182 completion, the resulting minerals are carbonates and hydrated silica. This is what has occurred
183 in listvenite deposits where all primary minerals have been altered to form carbonate and
184 hydrated silica (e.g., Falk and Kelemen, 2015). Carbonation of olivine always results in
185 precipitation of additional silicate minerals that contain the Si atoms from olivine and pyroxene
186 that lose their hosted cations to more thermodynamically stable carbonate minerals (e.g., Falk
187 and Kelemen, 2015; Kelemen et al., 2020b). The rate of olivine carbonation is maximized at
188 $\sim 185^\circ\text{C}$, but also occurs at slower rates at temperatures between $\sim 1\text{--}350^\circ\text{C}$ (Kelemen and
189 Hirth, 2012; O'Connor et al., 2005). The rate of carbonation is more sensitive to temperature
190 than pressure (Kelemen and Hirth, 2012) and is most sensitive to CO_2 partial pressure (P_{CO_2}),
191 with carbonation rates increasing roughly linearly as a function of increasing P_{CO_2} (O'Connor et
192 al., 2005). Fluids associated with carbonation of olivine typically have high pH, high alkalinity,
193 and low oxygen fugacity (Kelemen et al., 2011). Fluid chemistries associated with carbonation
194 of silicates are habitable when temperatures are less than 122°C , the currently known high
195 temperature limit for life (Takai et al., 2008), resulting in microbial inhabitation of many
196 peridotites actively experiencing serpentinization/carbonation (e.g., Kraus et al., 2021;
197 Rempfert et al., 2017; Schrenk et al., 2013).

During carbonation of silicates, reaction-driven cracking due to volume expansion exposes fresh primary mineral surfaces (Kelemen and Hirth, 2012; Klein and Le Roux, 2020) and produces fracture networks filled with mineralized carbonate veins (Evans et al., 2020; Kelemen and Matter, 2008). Carbonation of ultramafic rocks occurs in submarine oceanic crust as well as in obducted oceanic crust –ophiolites (Falk and Kelemen, 2015; Kelemen et al., 2020b). Subaerial and submarine ultramafic volcanics, including komatiites, kimberlites, and boninites, are also typically carbonated (Mitchell et al., 2019). Some of this carbonate is igneous, erupting from the mantle as carbonatite-ultramafic melts/tuffs, which have the lowest SiO₂ activity of any known volcanic material (Agashev et al., 2008; Dongre and Tappe, 2019; Doroshkevich et al., 2019; Duke et al., 2014; Melluso et al., 2010; Russell et al., 2012; Shavers et al., 2016; Sparks et al., 2009; White et al., 2012). Kimberlites erupt from >150 km depth, have high CO₂ activity, and typically contain carbonates of secondary origins, in addition to primary carbonates from carbonatite melt components (Sparks, 2013). Some aqueous alteration/carbonation during emplacement likely occurs via alteration by volatiles hosted in melts during their eruption (Mitchell, 2013, 2008; Mitchell et al., 2019, 2009), while some aqueous alteration likely occurs via assimilation of surface/near-surface fluids in a hydrothermal system within the hot ultramafic tuff or solidifying lava (Sparks et al., 2009).

Carbonate can also abiotically precipitate independently of the presence of ultramafic materials, including in evaporite deposits (e.g., Kah et al., 2001), within carbonate-saturated water columns (e.g., Given and Wilkinson, 1985), and as pedogenic carbonate in soils (Zamanian et al., 2016). Biomediated precipitation of carbonate occurs in soils (e.g., Whiffin et al., 2007) and within water columns (e.g., Dupraz et al., 2004), or in structures such as microbial

220 mats (e.g., Dupraz et al., 2009), shells, and reefs (e.g., Webb, 1996). Evaporite deposits often
221 contain other minerals in assemblage with carbonate, depending on the initial fluid
222 composition, including various sulfates and chlorides (e.g., Kah et al., 2001). Biomediated
223 carbonate precipitation in the water column is not necessarily accompanied by precipitation of
224 other minerals (e.g., Dupraz et al., 2009, 2004; Webb, 1996). Both abiotically and biotically
225 precipitated carbonates preserve biosignatures on Earth. The habitability of conditions under
226 which carbonates precipitate is therefore the most important control on carbonate
227 biosignature preservation potential.

228 Martian carbonates have been identified in meteorites ALH 84001, Nakhla, Governor
229 Valadares, Lafayette, and EETA 79001 (McSween, 1994), in the Comanche carbonate
230 investigated by the Spirit rover (Morris et al., 2010), in soil surrounding the Phoenix lander
231 (Boynton et al., 2009), possibly globally in Mars dust (Bandfield et al., 2003) using TES—though
232 sulfates could also explain this spectral signature (Lane et al., 2004), and in bedrock in multiple
233 localities on the planet using CRISM (Amador et al., 2017; Bultel et al., 2019; Carrozzo et al.,
234 2017; Carter et al., 2015; Ehlmann et al., 2008b; Jain and Chauhan, 2015; Michalski et al., 2017,
235 2013; Michalski and Niles, 2010; Wray et al., 2016) and TES (Glotch and Rogers, 2013).

236 Carbonate detections using CRISM are typically associated with mixed or proximal
237 phyllosilicates (Bultel et al., 2019; Carrozzo et al., 2017; Michalski et al., 2017; Wray et al.,
238 2016). Assuming equilibrium conditions, carbonates in ALH 84001 formed at 18 ± 4 °C (Halevy
239 et al., 2011) 3.9-4.0 Gyr ago (Borg et al., 1999) and are present as concretions or as massive
240 carbonate intergrown with feldspathic glass and orthopyroxene (Corrigan and Harvey, 2004;
241 Steele et al., 2007), while Nakhla and Governor Valadares meteorites contain vein-filling

carbonate (Gooding et al., 1991) and carbonate associated with silicate alteration zones (Bridges and Grady, 2000). The Comanche carbonate has been proposed to form via hydrothermal alteration (Morris et al., 2010) or evaporite precipitation following alteration of the Algonquin volcanic tephra deposits (Ruff et al., 2014).

3.0 Methodology

To characterize the properties of carbonate-bearing rocks in Jezero crater to the greatest extent possible from orbit, we integrated all orbital datasets that are at sufficiently high spatial resolution to investigate individual carbonate-bearing outcrops. We used CRISM to identify VNIR spectra consistent with the presence of carbonate and minerals in assemblage with carbonate, images and DEMs from HiRISE and CTX to characterize morphology and place detections in their geologic and, if possible, stratigraphic contexts, as well as thermal inertia (TI) estimates from THEMIS measurements to evaluate dust cover and rock competency, which we then linked to identified VNIR spectral absorption features. We also used results from recent photogeologic mapping of Jezero crater by the Mars 2020 Rover Science Team (Stack et al., 2020) to determine which of these units are carbonate-bearing. This integrated approach permits evaluation of hypotheses for the origins of carbonate-bearing rocks in Jezero crater.

3.1 CRISM analysis

For this analysis, we focused on CRISM images HRL000040FF (32 m/pixel), FRT000047A3 (18 m/pixel), and FRT00005C5E (18 m/pixel), which cover the Perseverance rover landing site, the geologic units mapped by Stack et al. (2020), and previously identified carbonates. We used

CRISM data produced via three different processing techniques to identify spectral features consistent with the presence of carbonate and additional assemblage minerals. These included (1) the CRISM TRR3 pipeline, (2) the CRISM TER pipeline, and (3) the University of Massachusetts at Amherst (UMass) pipeline (Itoh and Parente, 2021). To identify regions with spectral signatures consistent with carbonate, we combined three separate mineral mapping approaches: (1) Band parameters (Viviano-Beck et al., 2014), (2) Dynamic aperture factor analysis/target transformation (DAFA/TT; Lin et al., 2021), (3) a Generative Adversarial Network based feature extraction technique (GAN; Saranathan and Parente, 2021). We compared the results from each of these techniques to test their validity. We also used DAFA/TT to locate unique spectra that are not present in the Minerals Identified through CRISM Analysis (MICA; Viviano-Beck et al., 2014) library that is employed during typical GAN mapping of CRISM data, then used those DAFA/TT-discovered spectra as prototypes for more detailed GAN mapping. After characterizing the spatial distribution of spectral signals in the three CRISM images analyzed, and interpreting the spectrally-dominant mineralogy consistent with those signals, we isolated CRISM pixels with similar spectral signals that outcropped over defined geologic units (e.g., dunes, differently textured bedrock, low thermal inertia material, high thermal inertia material, and specific geologic units) based on morphology, thermal inertia, and the mapping results from Stack et al. (2020). We then interpreted the mineral assemblages consistent with spectra from these pixels in all three CRISM data types (TRR3, TER, and UMass) via comparison to library spectra. Carbonate minerals are identified by the presence of absorptions centered at 2.3 and 2.5 μm , Fe/Mg-phylosilicates by a narrow absorption centered at 2.3 μm , Al-phylosilicate by a narrow absorption centered at 2.2 μm , hydrated silica by a

broad absorption centered at 2.2 μm , and olivine and siderite by a broad absorption centered at 1.0 μm . Integrating this information, we characterized the mineral composition of specific rock units or debris in Jezero crater to the greatest extent currently possible using orbitally-acquired VNIR hyperspectral data.

We compared carbonate spectral signals in Jezero crater with those in the ROB unit elsewhere in the Nili Fossae region using CRISM TRR3 images FRT0000C256, FRT00009D96, FRT000095FE, and FRT000028BA. We chose these images based on the strong carbonate absorption features in this area that were reported by Mandon et al., (2020). Following Horgan et al. (2020a), we mapped the value of the 2.3/2.5 μm absorption band depths in these images to determine whether the carbonate absorption strength seen in the margin fractured unit is unique to Jezero, or is consistent with carbonate spectral signals in the ROB unit throughout the Nili Fossae region. We also used the Hyperspectral Subspace Identification (HySime) algorithm (Bioucas-Dias and Nascimento, 2008) to calculate endmembers imparting spectral variance in CRISM pixels covering outcrops of carbonate-bearing rocks in Jezero crater as well as in the CRISM images covering the ROB in the greater Nili Fossae region. We used information from band parameter mapping (Viviano-Beck et al., 2014) to isolate CRISM pixels used for spectral endmember extraction. If the endmembers resembled spectra characteristic of specific minerals, they were interpreted to reflect the composition of the surface covered by those CRISM pixels. This spectral endmember extraction and interpretation process is analogous to methods applied in past studies (e.g., Bandfield et al., 2002, 2000; Fischer et al., 2015; Glotch and Bandfield, 2006; Smith et al., 2000). It allowed us to extract clean mineral assemblage spectra from both Jezero crater and the surrounding Nili Fossae region CRISM images, even

though the images from the surrounding region lacked spectrally bland pixels that would be required for spectral ratioing (e.g., Mustard et al., 2008). This allowed us to directly compare the spectra of carbonate-bearing mineral assemblages in Jezero crater with those in the ROB unit outside of Jezero crater, while also comparing how relative absorption strengths varied across each image.

3.2 HiRISE and CTX images and DEMs

We used the HiRISE and CTX image/DEM mosaics of Jezero crater generated by Fergason et al., (2020), as well as HiRISE image/DEM mosaics and a global CTX mosaic generated by Dickson et al., (2020). HiRISE images were used to characterize unit morphology, distinguish between differently textured bedrock units, and differentiate between bedrock and aeolian bedforms or other clearly unconsolidated materials. This information was used to isolate specific CRISM pixels for spectral analysis and mineral assemblage interpretation (Section 3.1). We also used HiRISE DEMs to extract cross sections of CRISM detections and broader-scale geologic units for spectral and stratigraphic interpretations.

3.3 THEMIS thermal inertia mapping

We first used the qualitative THEMIS thermal inertia mosaic from Fergason et al., (2006), which the Dickson et al. (2020) HiRISE image/DEM and CTX image mosaics are coregistered to, in order to differentiate bedrock exposures from unconsolidated material. This information was used to isolate CRISM pixel clusters for spectral analysis and mineral assemblage interpretation (Section 3.1). After characterizing the spectra of different bedrock

units and unconsolidated materials in Jezero crater, we noticed a possible correlation between the band depth of the 2.5 μm absorption features—consistent with and attributed to the presence of carbonate—with thermal inertia. To further investigate this relationship, we used a quantitative thermal inertia map to characterize the relationship between thermal inertia and VNIR spectral features in Jezero crater.

Quantitative thermal inertia maps of Gale crater were generated by inputting nighttime THEMIS images (~ 100 m/px) (Christensen et al., 2004b) taken during the southern hemisphere summer into the KRC thermal model (Edwards et al., 2018a; Fergason et al., 2006b; Kieffer, 2013), which fits surface temperature estimates at each pixel to seasonally stable thermal inertia values. Additional inputs include observation time parameters (e.g., season, local time, solar azimuth, dust opacity, and modeled air temperatures), surface geometry (e.g., slope angle, aspect, and elevation) and estimates of material properties (e.g., albedo, emissivity, specific heat, and thermal conductivity) (Kieffer, 2013; Putzig et al., 2005).

3.4 THEMIS thermal inertia and CRISM band parameter correlation

Using a THEMIS quantitative thermal inertia mosaic combined with CRISM MTRDR data of image HRL000040FF, we evaluated the relationship between quantitative thermal inertia and other band parameters in Jezero crater. In our CRISM and qualitative thermal inertia analysis, we noticed a similarity between thermal inertia and the absorption depth of BD860—attributed to hematite—in Gale crater and thermal inertia and the absorption depth of BD2500_2—attributed to carbonate—in Jezero crater. In Gale crater, Vera Rubin ridge (VRR) has the strongest and most widespread BD860 feature of hematite in northern Mount Sharp (Fraeman

et al., 2013), as well as high thermal inertia relative to the surrounding terrain (Edwards et al., 2018b). In Jezero crater, the margin fractured unit has the strongest BD2500_2 feature of carbonate (Horgan et al., 2020a), as well as high thermal inertia relative to the surrounding terrain (Figure 2). After a detailed campaign by Curiosity, the deeper VNIR hematite absorptions in VRR were attributed to hematite crystallinity enhanced by groundwater and less coverage by unconsolidated material due to cementation that gives VRR its topographic prominence (David et al., 2020; Fraeman et al., 2020a, 2020b; Frydenvang et al., 2020; Horgan et al., 2020b; Jacob et al., 2020; L'Haridon et al., 2020; McAdam et al., 2020; Morris et al., 2020; Rampe et al., 2020; Thompson et al., 2020). Because of the qualitative correlation between CRISM signal and thermal inertia in these localities, we quantitatively compared CRISM VNIR band parameters and quantitative thermal inertia in Gale crater and Jezero crater, interpreting that a similarity in their relationship may be indicative of similar groundwater alteration histories impacting the modern-day physical and spectral properties of VRR and the margin fractured unit. The 1 μm absorption of olivine has previously been shown to not correlate with thermal inertia (Brown et al., 2020).

The algorithm for comparing CRISM band parameters and quantitative thermal inertia data uses Principal Components Analysis to identify which band parameters contribute to the largest variability in the data and reveals how linked each parameter is to the variability in thermal inertia. Using thermal inertia maps resampled to CRISM spatial resolution, data points are then clustered into Gaussian mixtures using only key high-variability band parameters and thermal inertia as inputs. Correlation coefficients between band parameters and a representative average thermal inertia can then be calculated for individual geologic units.

Further details of the classification method can be found in Koepfel et al., (In Review). While we found interesting relationships between multiple CRISM band parameters and thermal inertia, here we focus on BD860 in Gale crater and BD2500_2 in Jezero crater to constrain possible hypotheses for margin fractured unit emplacement, mechanisms for carbonation, and the relationship between orbital observations and anticipated landed observations in Jezero crater.

4.0 Results

Combining the results of our mineral mapping with the photogeologic map of Jezero crater from Stack et al. (2020), we found VNIR spectra consistent with the presence of carbonate (Figure 3) in (1) high TI bedrock in the margin fractured unit (Figures 4-6, S1-S3), (2) large aeolian bedforms proximal to the margin fractured unit (Figures 7, S4-S5), (3) the delta blocky unit (Figure 8), (4) the delta truncated curvilinear unit (Figure 9), (5) undifferentiated smooth material on the western delta (Figures 10, S6), (6) the delta layered rough unit (Figures 11, S7), (7) moderate TI bedrock in the crater floor fractured 2 (CFF 2) unit (Figures 12-13, S8), and (8) low TI dunes and unconsolidated dark-toned material in the CFF 2 unit (Figure 14). We also report five newly discovered occurrences of hydrated silica in the same dark-toned material that Tarnas et al. (2019) and Dundar et al. (2019) reported hydrated silica in (Figures 15-16, S9). Below we describe the VNIR spectral features of these outcrops and associated mineral assemblage interpretations based on these spectral features (Figure 17). We also describe their morphology, thermal inertia properties, geologic context, and if possible stratigraphic relationship to other units in Jezero crater.

4.1 Crater floor fractured 2 unit

4.1.1 VNIR spectra

We find VNIR spectra consistent with the presence of carbonate in bedrock within the crater floor fractured 2 unit (Figures 12, S8), as well as dunes and dark-toned unconsolidated material directly overlying crater floor fractured 2 unit bedrock (Figure 14). The spectra are consistent with a mixture of carbonate, phyllosilicate(s), olivine, and possibly hydrated silica. The presence of carbonate is indicated by absorptions centered at 2.3 and 2.5 μm and has previously been mapped in this unit by Ehlmann et al., (2009) and Goudge et al., (2015). The higher 2.3/2.5 μm absorption band depth ratio of these spectra compared to spectra of pure carbonate, as well as the narrowing of the 2.3 μm feature relative to that of pure carbonate, indicates mixing with phyllosilicate (Figures 12, 14, 17, S8). The broad absorption feature centered at 1.0 μm is consistent with the presence of olivine and/or siderite (Figure 17). Because the crater floor fractured 2 unit contains widespread spectra consistent with olivine, we interpret olivine to be the more likely cause of the 1.0 μm centered broad absorption.

The presence of olivine, carbonate, and phyllosilicate(s) in the crater floor fractured 2 unit is consistent with past interpretations of the mineral assemblage in this unit in Jezero crater (Horgan et al., 2020b) and the ROB unit in Nili Fossae (Brown et al., 2010). Based on the presence of a 2.2 μm feature (e.g., Figure 17 hydrated silica and montmorillonite spectra) that combines with the narrow 2.3 μm absorption (e.g., Figure 17 saponite, talc, and serpentine spectra), we interpret the presence of hydrated silica and/or Al-phyllosilicate, consistent with the interpretation of this same feature in a similar spectrum from the ROB unit in Tarnas et al.

(2019). We reason that this feature is more likely to be caused by presence of hydrated silica rather than Al-phylosilicate if the protolith material is ultramafic, as hydrated silica is ubiquitous in 100% carbonated olivine deposits (listvenites). However, if the protolith composition of this unit is mafic rather than ultramafic, Al-phylosilicates may have formed via alteration of plagioclase during carbonation of olivine, as occurs during carbonation of mafic rocks on Earth (Matter and Kelemen, 2009).

We find five new outcrops of the smooth dark-toned hydrated-silica-bearing material reported by Tarnas et al. (2019) and Dundar et al. (2019) (Figures 15-16, S9). As reported by Tarnas et al. (2019), this smooth dark-toned material always outcrops immediately above the crater floor fractured unit. We also find an outcrop where smooth dark-toned material partially covers crater floor fractured unit bedrock, but does not cover it entirely, and over this area we find a stronger 2.2 μm absorption consistent with hydrated silica relative to the 2.5 μm absorption consistent with carbonate (Figure 16). This indicates that at least some of the 2.2 μm absorption signal from the crater floor fractured 2 unit is due to coverage of crater floor fractured 2 unit bedrock by hydrated-silica-bearing smooth dark-toned material. However, there is likely some material in crater floor fractured 2 bedrock bearing a 2.2 μm absorption feature consistent with hydrated silica and/or Al-phylosilicate, as this absorption feature is present even when there is no obvious dark-toned material covering the outcrop (e.g., Figure 12). The origin and stratigraphic position of this smooth dark-toned hydrated-silica-bearing material remains ambiguous.

4.1.2 Morphology, thermal inertia, and geologic context

Crater floor fractured unit 2 bedrock outcrops containing carbonate-bearing mineral assemblages have fractures and linear ridges that are characteristic of this unit both within Jezero crater and regionally (Bramble et al., 2017; Goudge et al., 2017, 2015). This unit also contains larger-scale topographic features including broader ridges (Figure 13). Carbonate-bearing bedrock textures also include aeolian bedforms and dark-toned rubble overlying light-toned bedrock, which protrudes through this unconsolidated material (Figure 14). The morphology of crater floor fractured 2 unit bedrock with VNIR spectra consistent with carbonate at CRISM spatial scales is not appreciably different from outcrops of this unit with no clear VNIR carbonate spectral absorptions, nor is it appreciably different from the surface textures in much of the margin fractured unit. Parts of the crater floor fractured 2 unit with VNIR spectra consistent with the presence of carbonate are also not topographically higher than unit components that do not have clear VNIR spectral signatures of carbonate. Qualitative thermal inertia estimates (Fergason et al., 2006a) of these outcrops are also consistent with both bedrock and overlying unconsolidated material containing VNIR spectra consistent with the presence of carbonate. The ROB unit is banded (Kremer et al., 2019) and some of this banding is visible in Jezero crater, but only in the crater floor fractured 1 unit. None of these banded outcrops have VNIR spectra at CRISM spatial scales consistent with the presence of carbonate. HiRISE images show no clear contacts between the crater floor fractured 1 and 2 units and the margin fractured unit. A cross section from the ROB unit outside of the crater to the edge of the largest northernmost outcrop of crater floor fractured 1 and 2 units shows a gradational relationship as this unit drapes into Jezero crater (Figure 18). Including the margin fractured unit in this cross section results in the same relationship (Figure 18).

4.2 Deltaic units

4.2.1 VNIR spectra

There are CRISM VNIR spectra consistent with the presence of carbonate in the delta blocky unit (Figure 8), the delta truncated curvilinear unit (Figure 9; Goudge et al., 2017), undifferentiated smooth material on the western delta (Figures 10, S6), and the delta layered rough unit (Figures 11, S7). The spectra are consistent with a mixture of carbonate, phyllosilicate, and either hydrated silica and/or Al-phyllosilicate. The presence of carbonate is indicated by absorptions centered at 2.3 and 2.5 μm . All VNIR spectra containing carbonate absorptions in the deltaic units have higher 2.3/2.5 μm ratios compared to carbonate-bearing VNIR spectra in the crater floor fractured 2 and margin fractured units, indicating a higher spectral abundance of Fe/Mg-phyllosilicates mixed with carbonate in the deltas (Figure 17). The 2.2/2.3 μm absorption depth ratios are also lower in the deltaic outcrops of carbonate-bearing assemblages compared to those in the crater floor fractured 2 unit, indicating a higher spectral abundance of Fe/Mg-phyllosilicate relative to the phase imparting the 2.2 μm absorption (Figure 17). It is unclear if the 2.2 μm absorptions in VNIR spectra of carbonate-bearing assemblages are caused by hydrated silica and/or Al-phyllosilicate (Figure 17). As noted by previous studies (Goudge et al., 2015) carbonate spectral absorptions are more spatially widespread in the northern fan deposit relative to the western delta deposit (Figures 8-10 and S6-S7 compared to Figures 11 and S7).

4.2.2 Morphology, thermal inertia, and geologic context

The morphological properties and stratigraphic relationships between the individual units in the Jezero delta are detailed in Stack et al. (2020). The units are, from stratigraphically youngest to oldest, the delta blocky, thickly layered and truncated curvilinear layered, thinly layered, and layered rough units. In the western delta, clear bedrock exposures of carbonate-bearing material at spatial resolutions measurable using CRISM are present in the delta blocky (Figure 8) and delta truncated curvilinear (Figure 9) units. These carbonate-bearing outcrops have previously been reported by (Ehlmann et al., 2009, 2008b; Goudge et al., 2015). Other locations with VNIR spectra consistent with the presence of carbonate are undifferentiated smooth, dark-toned material (Figures 10, S6). Light-toned bedrock exposures of carbonate-bearing material also have moderate thermal inertia (Figures 8-9) while carbonate-bearing undifferentiated smooth material has low thermal inertia (Figures 10, S6), consistent with the interpretation that the undifferentiated smooth material is unconsolidated relative to the light-toned bedrock. In the northern fan deposit, carbonate-bearing rock is widespread, both in moderate thermal inertia (Figure 11) and low thermal inertia (Figure S7) material. All outcrops of deltaic carbonate-bearing rocks are not morphologically distinct from other rocks within their units, nor do they show notable stratigraphic relationships with other rocks in their units. It is unclear whether the undifferentiated smooth material is eroded unconsolidated material from the deltaic bedrock units, or if it has experienced a different authigenic or detrital formation history compared to those deltaic bedrock units.

4.3 Margin fractured unit and proximal aeolian bedforms

4.3.1 VNIR spectra

Outcrops of bedrock in the margin fractured unit have VNIR absorption features consistent with a mixture of carbonate, phyllosilicate, and possible olivine, as has been reported by previous studies (Goudge et al., 2015; Horgan et al., 2020a). The presence of carbonate is indicated by absorptions centered at 2.3 and 2.5 μm . While spectra of the margin fractured unit have the lowest 2.3/2.5 μm absorption band depth ratios within Jezero crater, their relative strength is consistent with a mixture of carbonate and phyllosilicate (Figures 4-6, S1-S3). This is further supported by the narrow shape of the 2.3 μm feature relative to that typical of pure carbonate (Bishop et al., 2013) (Figure 17). The low 2.3/2.5 μm band depth ratio values in the margin fractured unit are also found in the ROB unit elsewhere in Nili Fossae (Figures 19, S10), including in the ROB unit immediately outside of Jezero (Figure 19a), indicating that the carbonate spectral strength in the margin fractured unit is nonunique in a regionally context. As indicated by Horgan et al., (2020a), the 1.0 μm feature in the margin fractured unit could either be caused by olivine and/or siderite (Figures 4-6, 17, S1-S3) and without clear absorption features of olivine in other outcrops of this unit, inferring that this mineral is the cause of the 1.0 μm absorption is potentially less valid than in the crater floor fractured 2 unit. However, the lack of clear contacts between the ROB unit draping into Jezero crater and the margin fractured unit does favor the interpretation that the 1.0 μm absorption in the margin fractured unit is due to olivine, which is present throughout the ROB unit.

The large aeolian bedforms immediately proximal to the margin fractured unit contain very similar VNIR spectra to margin fractured bedrock (Figures 7, S4-S5), indicating that they may be locally sourced sediments composed of the same carbonate-phyllosilicate-possible olivine mixture as the margin fractured unit. Many aeolian bedforms in Jezero have similar VNIR

spectral properties as proximal bedrock, indicating that they are often locally sourced from erosion of that bedrock (Arvidson and Christian, 2020).

4.3.2 Morphology, thermal inertia, and geologic context

The margin fractured unit has a fractured, sometimes rubbly, and occasionally ridged appearance similar to the crater floor fractured 2 unit (Figures 4-6, S1-S3). It has high thermal inertia compared to other units in Jezero crater (Figure 2b-c) and rocks with the strongest carbonate VNIR absorption features also have high thermal inertia (Figure 2a). There are no visible contacts between the margin fractured unit and the crater floor fractured 2 unit, nor the margin fractured unit and the ROB unit that drapes into Jezero crater (Goudge et al., 2015), with cross sections showing a gradational relationship in all cases (Figure 18). The morphology, high thermal inertia, and VNIR spectral signatures resemble the ROB unit (Goudge et al., 2015), including the ROB component immediately north of the crater rim from the margin fractured unit, and the component of the ROB that drapes into Jezero crater (Figure 2b-c). Many of the strongest carbonate absorption features in this unit are associated with elevation of the estimated maximum Jezero lake level (Horgan et al., 2020a). The delta blocky unit outcrops stratigraphically above the margin fractured unit and there are clear contacts in many locations (e.g., Figure 4c). In one area, a cross-section exposure of the margin fractured unit shows possible banding similar to that seen in the crater floor fractured 1 unit (Figure 3b), though these may actually be diagenetically-generated ridges rather than beds. The largest margin fractured unit outcrops contain either plateaus (e.g., Figures 5, S1-S2) or mounds of bedrock bearing the characteristic fractured morphology of the unit (e.g., Figures 4, 6, S3).

The large aeolian bedforms proximal to the margin fractured unit have low thermal inertia—which is expected for unconsolidated material—and overly margin fractured bedrock in many locations (Figure S5c). These aeolian bedforms are longer wavelength than those in lower elevations of western Jezero crater, likely due to a combination of physical properties of the unconsolidated grains and the predominant wind patterns in Jezero crater (Day and Dorn, 2019).

4.4 Carbonate-bearing rocks outside of Jezero

We find outcrops of carbonate-bearing rocks in the ROB unit with strong spectral similarities to carbonates in the crater floor fractured and margin fractured units in Jezero crater (Figure 19, S10). The 2.3/2.5 μm band depth ratios in carbonate-bearing rocks outside of Jezero crater are equivalent to those found in the margin fractured unit (Figure 19, S10). This is true in outcrops of the ROB 100-200 km northeast of Jezero crater (Figure 19b-d), outcrops of the ROB immediately northwards of the Jezero crater rim from the margin fractured unit (Figure 19a), and outcrops of ROB that drape into Jezero crater (Figure 19a). These low 2.3/2.5 μm band depth ratios span extensive amounts of outcrop regionally (Figure 19b, S10), and indeed may be present in many outcrops without CRISM FRT, HRL, FRS, HRS, or ATO image coverage. The spectral endmembers associated with CRISM pixels with BD2500_2 values greater than 0.005 inside and outside of Jezero crater are similar and contain absorptions consistent with the presence of carbonate, Fe/Mg-phyllosilicate, and olivine (Figure 19e). In summary, we find extensive carbonate-bearing rocks in the ROB unit that resemble the carbonate spectral signatures of the margin fractured unit.

5.0 Discussion

Our results indicate that it is equally likely that carbonates in Jezero crater formed via groundwater alteration rather than fluviolacustrine precipitation, but neither of these hypotheses can be definitively proven using orbital data. We include pedogenic carbonate formation in the groundwater formation scenario, as the fluids causing carbonate formation would be present in the subsurface, though they may have originally precipitated on the surface and percolated downwards. It is also possible that a hypothesis intermediate to groundwater and fluviolacustrine carbonate formation may be true, whereby some carbonate formed in the subsurface and some formed as fluviolacustrine sediment, as discussed by Horgan et al., (2020a).

In this section, we evaluate the observations that favor carbonate formation by groundwater (Sections 5.1 and 5.2) and carbonate formation in a fluviolacustrine environment (Sections 5.1 and 5.3). We discuss the facies that could preserve biosignatures in either scenario based on Earth analogs (Section 5.4). Finally, we discuss the possibility that some carbonate in Jezero crater and the surrounding Nili Fossae region may have formed under the conditions of the modern martian atmosphere (Section 5.5; Kelemen et al., 2020a).

5.1 Thickness of the margin fractured unit

The margin fractured unit is topographically higher than the crater floor fractured 2 unit (Horgan et al., 2020a; Figure 18). If the margin fractured and crater floor fractured 2 units are both part of the ROB unit, then this topographic difference could be caused by ROB unit

thickness variations (Scenario 1) or topographic variations in the underlying unit draped by the ROB unit (Scenario 2). If unit thickness variations are the cause of topographic variations, then the margin fractured unit would be ~10-100 times thicker than the ROB is elsewhere in Nili Fossae and ~3-6 times thicker than the ROB is in Libya Montes (Kremer et al., 2019). Another possible explanation for the observed topographic difference is that 100s of meters of lithified fluviolacustrine sediments have been emplaced atop the ROB unit (Scenario 3).

5.1.1 Scenario 1: The margin fractured unit is a thick outcrop of ROB unit

Our observations favor Scenario 1 over Scenario 2, as some margin fractured outcrops show possible banding (e.g., Figure S3b, S4a) and are ~200 meters thick (e.g., Figure 18 Cross Section E). Perhaps the ROB unit was originally 100s of meters thick when originally emplaced, similar to its less-eroded outcrops in Libya Montes (Kremer et al., 2019), and has been substantially eroded to <10% of its original thickness in the Nili Fossae region. This interpretation is consistent with the typical material properties of high TI clastic rocks on Mars, which are interpreted to be friable, eroding faster than surrounding rocks to expose relatively dust-free surfaces (Rogers et al., 2018).

If Scenario 1 (Figure 18) is the correct interpretation for topographic variability between the margin fractured and crater floor fractured units, then the margin fractured unit component of the ROB unit was more cemented and/or protected from erosion compared to the ROB elsewhere in Jezero crater and the Nili Fossae region. Because the delta blocky unit is locally stratigraphically above the margin fractured unit (e.g., Figure 4c), it is possible that the delta originally covered part of the margin fractured unit, protecting it from erosion.

Furthermore, coverage by the delta and/or any since-eroded overlying units would likely provide a pathway for later groundwater flow. This is similar to the proposed setting for past enhanced groundwater flow in VRR, Gale crater, where the contact between the Murray Formation and the Stimson Formation is thought to have provided a pathway for groundwater, which cemented VRR relative to surrounding rocks and also protected it from physical erosion while the overlying material was being removed (Bryk et al., 2019; Rampe et al., 2020). This constitutes another possible similarity between the origins of VRR in Gale crater and the margin fractured unit in Jezero crater. The position of the margin fractured unit immediately interior to Jezero crater's rim also increases the likelihood that this material experienced enhanced groundwater alteration relative to the ROB unit further interior to Jezero crater, as well as outside of Jezero crater, because bedrock surrounding impact craters is fractured and more permeable than non-impacted bedrock (Collins, 2014). Such enhanced permeability would increase groundwater flow around and into Jezero crater, possibly resulting in enhanced mineral dissolution, disequilibria from fluid mixing, and more favorable redox conditions for chemolithotrophic microorganisms. These observations are consistent with enhanced groundwater alteration producing the topographic (Figure 19), VNIR spectral, and thermal inertia properties of the margin fractured unit, similar to hypotheses proposed for the formation of VRR (Bryk et al., 2019; Fraeman et al., 2020b, 2020a; Jacob et al., 2020; Rampe et al., 2020), but under less acidic aqueous conditions. Differences in fluid chemistry could cause groundwater-driven formation of hematite in the Murray Formation under oxidizing conditions, but cause groundwater-driven formation of carbonate in Jezero crater given high dissolved CO₂ concentrations.

638
639 *5.1.2 Scenario 2: The margin fractured unit is the ROB unit draping underlying topography*

640 Our observations do not rule out Scenario 2 (Figure 19), where topographic differences
641 between the margin fractured and crater floor fractured units are controlled by topographic
642 variations of an underlying unit. Fresh ~40 km diameter impact craters do not typically have
643 shelves immediately interior to their rims, therefore this is unlikely to be the cause of any
644 underlying topographic variability. Furthermore, if there was a thick unit emplaced beneath the
645 margin fractured unit, it would likely be present within the Nili Fossae regional stratigraphy
646 (Ehlmann and Mustard, 2012) between the basement and ROB units; no such unit is observed
647 outside of Jezero crater. Still, Jezero crater has been filled by ~1 km of material relative to the
648 shape expected of a fresh impact crater of its size (Fassett and Head, 2005; Schon et al., 2012)
649 and the nature of that material remains ambiguous, as it would also likely outcrop between the
650 basement and ROB units if it was emplaced across the entire Nili Fossae region. The ROB drapes
651 topography across the entire Nili Fossae region, as well as in Libya Montes (Kremer et al., 2019),
652 with outcrop thicknesses from ~1-25 meters in Nili Fossae and ~50-100 meters in Libya Montes.
653 It is therefore possible that the topography of the margin fractured unit compared to the crater
654 floor fractured unit is entirely dictated by underlying topography, without any thickness
655 variations greater than ~100 meters in the unit itself. If this is the case, and the unit is only ~10
656 meters thick, it is possible that the Radar Imager for Mars' Subsurface Exploration (RIMFAX;
657 Hamran et al., 2020) might see an unconformable contact between it and the underlying unit
658 that controls topography.

5.1.3 Scenario 3: The margin fractured unit is lithified fluviolacustrine sediment

Scenario 3, where thickness differences between the margin fractured and crater floor fractured units are caused by deposition of fluviolacustrine sediments, is another possible explanation for the observations reported here (Figure 18). As proposed in the study detailing a fluviolacustrine origin hypothesis for the margin fractured unit carbonates (Horgan et al., 2020a), these sediments could be deposited in a shallow water setting during the highstanding period of ancient Lake Jezero. Such carbonate-bearing fluviolacustrine sediments form topographic highs at the former margins of lakes on Earth (e.g., Antalya region, SW Turkey; Glover and Robertson, 2003), therefore this could be the cause of the thickness variations reported here (Figure 18). In this scenario, hundreds of meters of lithified fluviolacustrine sediment could be protected from erosion by overlying deltaic material and/or a since eroded overlying unit, similar to the argument presented in Scenario 1. This fluviolacustrine sediment would be deposited before the outflow channel in eastern Jezero crater was breached. Authigenic and detrital sedimentation rates on early Mars are unconstrained, therefore we do not estimate the amount of time required for deposition of 100s of meters of lithified fluviolacustrine sediments in the Jezero paleolake before the breach event occurred. However, the Curiosity rover has traversed ~300 meters of a fluvial, deltaic, lacustrine, and aeolian sediments in Gale crater, which is also a crater lake basin, therefore a paleolacustrine sedimentary sequences 100s of meters thick is not unprecedented in the martian geologic record.

In summary, the topographic difference between the margin fractured and crater floor fractured units favors either cementation by groundwater, draping of underlying topography,

or fluviolacustrine sediment deposition, depending on the preferred interpretation of the cross sections presented here (Figure 18). Both the groundwater alteration and fluviolacustrine scenarios (Scenarios 1 & 3) have high potential to preserve biosignatures. In the groundwater scenario (Scenario 1), biosignatures would be entombed in mineralized veins where previously habitable alkaline groundwater flowed, whereas in the fluviolacustrine scenario (Scenario 3), biosignatures would be entombed in fine-grain sedimentary carbonates that authigenically formed in the lake water column.

5.2 Formation of carbonates by alkaline groundwater

It is possible that the margin fractured and crater floor fractured 2 units are both part of the ROB unit. This interpretation is based on the morphologic, VNIR spectral, and stratigraphic properties shared by these units. Furthermore, the ROB unit drapes into Jezero crater and has no visible contact or morphologic differences with the margin fractured unit (Figure 18), implying that they are the same unit. The ROB unit also has carbonate absorptions of similar strength to those found in the margin fractured unit both immediately outside of Jezero crater (Figure 19a), which are contiguous with the margin fractured unit, and in other locations of this unit in Nili Fossae (Figure 19b-d). 2.3/2.5 μm band depth ratios also show no significant differences between carbonate-bearing mineral assemblage spectra in the margin fractured unit, the ROB unit immediately outside of Jezero crater, the ROB unit draping into the Jezero crater rim, or the ROB unit 100-200 km northeast of Jezero crater (Figure 19). The lack of any contacts between the margin fractured and crater floor fractured units, as well as the VNIR spectral and morphological similarities between these units, implies a shared carbonate

formation process. This interpretation is consistent with past interpretations of the relationship between the margin fractured, crater floor fractured 2, and ROB units (Ehlmann et al., 2009; Goudge et al., 2015).

Spectral variations between the margin fractured and crater floor fractured 2 units could be attributed to groundwater alteration and cementation. Groundtruthing of orbitally-measured VNIR spectra by Curiosity in Gale crater demonstrates that variations in groundwater activity can substantially alter VNIR spectral properties of rocks without appreciably changing their modal mineralogy (Fraeman et al., 2020b, 2020a; Rampe et al., 2020). The Curiosity rover recently completed a campaign to characterize the Vera Rubin ridge (VRR), which had originally been called the hematite ridge based on its strong 0.86 μm absorption band in CRISM data, which was attributed to hematite (Fraeman et al., 2013). This extensive campaign indicated that VRR appears to be a component of the more widespread Murray Formation, but has experienced episodes of groundwater alteration that cemented the rocks in the ridge to make them more resistant to erosion relative to other rocks in the Murray Formation (David et al., 2020; Fraeman et al., 2020a, 2020b; Frydenvang et al., 2020; Horgan et al., 2020b; Jacob et al., 2020; L'Haridon et al., 2020; McAdam et al., 2020; Morris et al., 2020; Rampe et al., 2020; Thompson et al., 2020). This has resulted in less coverage by unconsolidated material and higher thermal inertia relative to the majority of the Murray Formation (Edwards et al., 2018). This same groundwater alteration also increased the crystallinity of hematite in the ridge, producing a strong 0.86 μm absorption attributed to hematite without substantially changing rock modal mineralogy from that of the rest of the Murray Formation (Rampe et al., 2020). Indeed, hematite modal abundances in drill cores in and near VRR ranged from 3-15% (Rampe

et al., 2020), compared to 1-14% elsewhere in the Murray Formation (Bristow et al., 2018; Rampe et al., 2017). Stoer, the drill core sampled during the VRR campaign with the highest hematite modal abundance, was not located on a part of VRR with a particularly strong 0.86 μm hematite absorption in CRISM data (Fraeman et al., 2020b, 2020a; Rampe et al., 2020). Based on these results, physical and spectral mixing of phases, variations in cementation and grain sizes of rock components, variability of coverage by unconsolidated material, and differences in spatial resolutions between datasets can manifest in substantial differences between orbital interpretations and groundtruth.

A similar groundwater-driven history would explain the properties of the margin fractured unit relative to the crater floor fractured 2 unit, though under different fluid chemistries compared to groundwater in the Murray Formation. The margin fractured unit is high-standing compared to the crater floor fractured 2 unit (Figure 18), indicating that it is cemented and more resistant to erosion. It also has high thermal inertia relative to the crater floor fractured unit (Figure 2b-c). Margin fractured unit rocks with the strongest VNIR carbonate absorptions also have high thermal inertia (Figure 2a-c), just as VRR rocks with the strongest VNIR hematite absorptions have high thermal inertia (Figure 2d-f). The ROB unit has outcrops of high thermal inertia rocks with strong VNIR carbonate absorptions both inside and outside of Jezero crater (Figure 19). Because there is no clear contact between these rocks and those with identical properties in the margin fractured unit, it is likely that they constitute the same unit. In sedimentary rocks, high thermal inertia can be attributed to lower coverage by unconsolidated material as well as increased rock cementation. The topographically-highstanding nature of the margin fractured unit indicates that cementation is likely a factor

affecting its thermal inertia, but the particularly light-toned nature of these rocks compared to others in Jezero crater indicates that they are also less covered by unconsolidated material. These properties are shared by VRR in Gale crater. As such, a simple explanation for our observations is that the margin fractured unit is the same as the crater floor fractured 2 and ROB units, but has experienced more cementation and crystallinity enhancement by groundwater alteration. This, combined with less coverage by unconsolidated material, is likely what generates its strong VNIR absorptions consistent with carbonate. It is possible that increased crystallization by groundwater could erase or overprint biosignatures in these rocks (McMahon et al., 2018), but increased cementation of the rocks could also make them resistant to physical weathering and decrease their permeability, lowering susceptibility to later groundwater infiltration episodes that could overprint biosignatures. The cementing material could also be authigenic carbonate precipitated from alkaline groundwater with high dissolved carbonate concentrations, which could preserve biosignatures of microorganisms inhabiting the groundwater (McMahon et al., 2018; Summons et al., 2011).

5.3 Formation of fluviolacustrine carbonates

A fluviolacustrine origin for carbonates in the margin fractured unit of Jezero crater (Horgan et al., 2020a) is equally as likely as groundwater carbonate formation given orbital observations. If the 1.0 μm absorption in this unit is caused by presence of olivine rather than presence of siderite, as seems probable based on the continuity between the margin fractured and ROB units, then authigenic lacustrine carbonates may be mixed with detrital mafic material that is present in the Jezero delta (Brown et al., 2020; Ehlmann et al., 2008a; Goudge et al.,

2017; Horgan et al., 2020a; Parente et al., 2019). Evidence for a fluviolacustrine origin for carbonates in the margin fractured unit includes the association of strong 2.5 μm absorptions and low 2.3/2.5 μm band depth ratios near the estimated shoreline of the Jezero paleolake at its highstand (Horgan et al., 2020a). However, similar carbonate spectral features can be seen in CRISM data or the ROB unit elsewhere in Nili Fossae (Figures 19, S10). Horgan et al., (2020a) also present evidence for spectral variability in the margin fractured unit as a function of distance from the inlet channel, but these observations could also be explained by variations in groundwater activity, cementation, and coverage by unconsolidated material. Based on these observations, as well as similarities in the relationship between VNIR spectral properties and thermal inertia in the margin fractured unit in Jezero crater and VRR in Gale crater (Figure 2), we consider a groundwater origin for enhanced VNIR carbonate absorptions to be equally as likely as a fluviolacustrine origin. In this scenario, the presence of deep VNIR carbonate absorptions between the highstand and breach levels of the Jezero paleolake is coincidental.

Deposition of ~200-300 meters of fluvial, deltaic, and lacustrine sediments during the highstand of the paleolake in Jezero crater could explain the greater thickness of the margin fractured unit relative to other carbonate-bearing rocks in Jezero crater and the Nili Fossae region (Section 5.1.3). In this scenario, the Jezero paleolake must have existed at its highstand level for sufficient amounts of time for this sediment to be deposited. Sedimentation rates vary widely on Earth and are largely controlled by climate and substrate properties. Since both of these parameters are largely unconstrained for Noachian Mars, it is difficult to accurately assess the amount of time required for ~200-300 meters of sediment to be deposited in the paleolake during its highstand. However, the Curiosity rover has traversed ~300 meters of

fluvial, deltaic, aeolian, and lacustrine sediments at Mount Sharp, therefore it is plausible that a similarly thick carbonate-bearing sedimentary sequence could have been deposited in Jezero crater during continuous or intermittent lake-forming eras. We conclude that a fluviolacustrine sedimentary origin for carbonates in the margin fractured unit is plausible based on orbital observations, but that carbonate formation by alkaline groundwater is equally likely based on similarities between carbonates within and outside of Jezero crater, as well as similarities between the rock properties of VRR and the margin fractured unit as observed from orbit.

5.4 Possible biosignatures from subsurface habitable environments

Our results suggest multiple possible formation mechanisms for the carbonates in Jezero crater, some of which may have created taphonomic windows conducive to biosignature preservation. It has been suggested that the stronger VNIR absorptions of carbonate in the margin fractured unit represent a carbonate deposit that formed in a fluviolacustrine environment (Horgan et al., 2020a). In this scenario, carbonate precipitation would have a high likelihood of preserving biosignatures like stromatolites and organic matter if the Jezero paleolake was inhabited at the time of carbonate formation. Our results do not preclude this formation mechanism, but indicate that it is equally likely that these carbonates formed from alkaline groundwaters, which commonly form subsurface habitable environments on Earth. While the surface of Mars was sporadically habitable (Wordsworth et al., 2021), deep subsurface environments likely constituted the longest-lived habitable environments on Mars (Michalski et al., 2018, 2013; Tarnas et al., 2018), providing a refugia for martian life. Near-subsurface groundwaters that connect to deep subsurface groundwater through liquid veins that cross-cut the cryosphere would introduce life from deep subsurface refugia into near-surface

environments, potentially entombing it in mineralized veins. This possible connectivity to deep subsurface refugia makes groundwater-derived carbonates particularly compelling targets for biosignature preservation in Jezero crater. Minerals precipitated around surface springs from subsurface fluid systems also preserve biosignatures on Earth (Hays et al., 2017), and spring-fed carbonate precipitants in Jezero crater would have high biosignature preservation potential.

If the margin fractured, crater floor fractured 2, and ROB units are the same rock unit, then carbonates in the margin fractured unit would have effectively the same material biosignature preservation potential as carbonates in the crater floor fractured or ROB units, although local conditions would dictate the likelihood for biosignatures to exist within different carbonate-bearing outcrops. The likelihood that these units contain biosignatures is therefore dictated by the temperature of any alteration by groundwater, as well as redox compound availability in those possible aqueous solutions. Mafic and ultramafic rock is carbonated on Earth under both habitable and uninhabitable conditions. Carbonate-bearing facies in subseafloor altered mafic and ultramafic rocks preserve biosignatures (Ivarsson et al., 2018, 2012; Klein et al., 2015), as do carbonates in terrestrial serpentinites formed by groundwater alteration of ophiolites (Newman et al., 2020), and cross-cutting alteration veins in terrestrial mafic and granitic sedimentary rocks that have been altered by groundwater (Drake et al., 2017; McKinley, 2000; Onstott et al., 2019).

5.5 Possibility of carbonate formation in modern day Mars atmospheric conditions

Kelemen et al., (2020a) presented the hypothesis that carbonation of olivine, particularly in the Nili Fossae region, could occur under the conditions of the modern day

martian atmosphere at temperatures $> 0^{\circ}\text{C}$. Under these conditions, carbonation of olivine would occur without precipitation of serpentine, brucite, or talc, which have not been unambiguously confirmed to exist in the ROB unit. However, there is spectral evidence of either talc or saponite in the ROB unit, with some authors favoring the interpretation of the presence of talc (Brown et al., 2010; Viviano et al., 2013). Small outcrops of Mg-rich serpentine have been reported in the ROB unit (Ehlmann et al., 2010; Leask et al., 2018). Furthermore, if more widespread Fe-rich serpentine were present, it could not be differentiated from many other phyllosilicates in the wavelength range measured by CRISM (Figure 17; Calvin and King, 1997; Ehlmann et al., 2010). Additionally, groundtruthings of orbital compositional measurements on Mars (Arvidson et al., 2008, 2006; Bristow et al., 2018; Carter and Poulet, 2012; Clark et al., 2007; Dobrea et al., 2012; Fox et al., 2016; Fraeman et al., 2016, 2013; Glotch et al., 2006; Morris et al., 2010, 2006; Rampe et al., 2020, 2017; Rice et al., 2010; Ruff et al., 2011; Squyres et al., 2008; Wang et al., 2006; Wray et al., 2009) indicate that CRISM analysis can accurately identify the *presence* of minerals in rocks on Mars, but cannot be used as strong evidence for the *absence* of minerals due to phase mixing effects and obscuring by dust.

While orbital evidence cannot unambiguously confirm or deny this hypothesis, it may be inconsistent with the presence of widespread olivine on the surface of Mars (Ody et al., 2013) with no reported associated secondary minerals. Many of these unaltered olivine provinces are at more equatorial latitudes than the Nili Fossae region, where temperatures are greater than 0°C for a higher portion of each martian year. However, due to precession of Mars on $\sim\text{Myr}$ timescales (Laskar et al., 2004), the modern day latitudinal distribution of olivine-bearing material on the surface of Mars may not correspond to the cumulative relative temperatures

experienced by those materials on Myr timescales. Still, if the modern day atmosphere of Mars could carbonate olivine to sufficiently thick skin depths to be measurable by CRISM, it is unclear why this would manifest in orbital carbonate identifications in Nili Fossae, but not in other olivine-bearing regions of Mars. It is possible that the ROB unit in Nili Fossae is uniquely permeable to the atmosphere compared to other olivine-rich rocks on Mars, which would increase carbonate rind thickness (Cannon et al., 2015; Salvatore et al., 2013). However, many of these olivine-rich terrains are likely clastic (Rogers et al., 2018) and therefore have similar permeabilities to the ROB unit, which is also likely clastic (Kremer et al., 2019). Nonetheless, carbonate rind formation in the ROB unit under modern Mars atmospheric conditions cannot be ruled out.

6.0 Conclusions

In Table 1, we compile observations of the margin fractured unit derived from orbital data and interpret whether they favor groundwater or fluvio-lacustrine origins for carbonates there. The conclusions of this study are summarized below.

1) Carbonate and hydrated silica-bearing units in Jezero crater. There are VNIR spectral absorptions consistent with carbonate in CRISM data covering (1) high TI bedrock in the margin fractured unit (Figures 4-6, S1-S3), (2) large aeolian bedforms proximal to the margin fractured unit (Figures 7, S4-S5), (3) the delta blocky unit (Figure 8), (4) the delta truncated curvilinear unit (Figure 9), (5) undifferentiated smooth material on the western delta (Figures 10, S6), (6) the delta layered rough unit (Figures 11, S7), (7) moderate TI bedrock in the crater floor

fractured 2 (CFF 2) unit (Figures 12-13, S8), and (8) low TI dunes and unconsolidated dark-toned material in the CFF 2 unit (Figure 14). We also find five previously unreported outcrops of hydrated silica in the same dark-toned material that Tarnas et al. (2019) and Dundar et al. (2019) reported hydrated silica in (Figures 15-16, S9).

2) Minerals in assemblage with carbonate in Jezero crater. Carbonate-bearing rocks in the crater floor fractured 2 unit also have absorption features consistent with a mixture of olivine, carbonate, Fe/Mg-phylllosilicate(s), and hydrated silica and/or Al-phylllosilicate (Figures 12, 14, 17, S8). VNIR spectra with carbonate absorptions in the deltaic unit have deeper and narrower 2.3 μm absorptions compared to carbonate spectra elsewhere in the crater (Figures 1, 8-11 17, S6-S7), implying a higher spectral abundance of Fe/Mg-phylllosilicate(s). These spectra also have 2.2 μm absorption features consistent with hydrated silica and/or Al-phylllosilicate and a broad 1.0 μm band consistent with olivine and/or siderite (Figure 17). The margin fractured unit and proximal aeolian bedforms have spectra consistent with a mixture of olivine and/or siderite, carbonate, and Fe/Mg-phylllosilicate(s) (Figures 4-6, 17, S1-S3). As reported by Horgan et al. (2020a), rocks in the margin fractured unit have the lowest 2.3/2.5 μm band depth ratios in Jezero crater, consistent with a higher spectral abundance of carbonate (Figure 19a).

3) Bedrock with strong carbonate absorptions has high thermal inertia. Margin fractured unit outcrops with the deepest 2.5 μm absorptions also have high thermal inertia relative to surrounding rocks, similar to the relationship between the 0.86 μm absorption and thermal inertia in VRR at Gale crater (Figure 2), implying that groundwater alteration/cementation may

generate the topographic features of the margin fractured unit as well as its spectral properties, as has been reported via the Curiosity rover campaign at VRR (Fraeman et al., 2020a). It is possible that this groundwater alteration/cementation would not increase the modal abundance of carbonate in the margin fractured unit, but rather would increase carbonate crystallinity as well as rock cementation, resulting in lesser coverage by unconsolidated material and increasing of the 2.5 μm absorption band depth. These groundwater-induced physical differences are what cause the deep 0.86 μm absorption in VRR (Fraeman et al., 2020b, 2020a; Horgan et al., 2020b; Jacob et al., 2020) without appreciably increasing the modal abundance of hematite in VRR compared to the rest of the Murray Formation (Bristow et al., 2018; Rampe et al., 2020, 2017).

4. Carbonates outside of Jezero crater are spectrally similar to margin fractured unit

carbonates. Carbonates in the margin fractured unit have the lowest 2.3/2.5 μm band depth ratios observed inside Jezero crater. We find widespread outcrops with similar 2.3/2.5 μm band depth ratios in the ROB unit 100-200 km NNE of Jezero crater (Figure 19b-d, S10), as well as immediately outside of Jezero crater (Figure 19a), showing that the spectral signatures of the margin fractured unit are not unique in a regional context. While we do not rule out the possibility of a fluviolacustrine origin for carbonates in the margin fractured unit, we find that it is equally likely that the margin fractured unit is part of the ROB unit, which drapes into Jezero crater and has no clear contacts or major spectral differences with the margin fractured unit. Whatever alteration has generated the low 2.3/2.5 μm absorption depth ratios in these rocks has also occurred elsewhere in the ROB in Nili Fossae.

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926 5. The margin fractured unit is likely 250-300 meters thick. The margin fractured unit is

927 topographically higher than the crater floor fractured unit (Horgan et al., 2020a). We propose

928 three hypotheses to explain this (Figure 19; Section 5.1). In Scenario 1, the margin fractured

929 unit is an exceptionally (~250-300 meters) thick outcrop of the ROB unit because it has been

930 cemented by groundwater and protected from erosion by overlying deltaic and since-eroded

931 material. There may have been enhanced groundwater flow at the contact of the underlying

932 ROB unit and overlying deltaic unit, similar to what has been proposed between the underlying

933 Murray Formation and overlying Stimson Formation in Gale crater to enhance groundwater

934 activity, cementing the underlying material (Bryk et al., 2019; Fraeman et al., 2020b, 2020a;

935 Jacob et al., 2020; Rampe et al., 2020). ROB outcrops in Libya Montes are up to ~100 meters

936 thick (Kremer et al., 2019), therefore it is possible that the ROB was originally deposited as a

937 ~300 meter thick unit that has since been heavily eroded, except in the margin fractured unit

938 where it was cemented by groundwater and protected from erosion by overlying units. In

939 Scenario 2, the margin fractured unit is part of the ROB with a regionally-common thickness of

940 1-25 meters. In this case, topographic variations of an underlying unit—which does not outcrop

941 between the basement and ROB units elsewhere in the Nili Fossae region—controls

942 topography. In Scenario 3, the margin fractured unit is thicker than the ROB unit because it is

943 covered by hundreds of meters of lithified fluviolacustrine sediments. This is feasible given that

944 the Mars Science Laboratory Curiosity rover has traversed ~300 meters of a fluvial, deltaic,

945 lacustrine, and aeolian sediments in Gale crater. Based on the outcropping of strong carbonate

absorptions in cross sections of the margin fractured unit (Figs. 18 Cross Section E, S3), we consider Scenarios 1 and 3 to be the most likely.

6. Margin fractured unit carbonates may have formed in a subsurface environment. Our results indicate that a groundwater origin for carbonates in the Jezero crater margin fractured unit is equally likely as a fluviolacustrine origin for these carbonates. In this scenario, carbonates would form via alteration of primary volcanic minerals by groundwater, which commonly forms alkaline waters and habitable subsurface environments on Earth (Kraus et al., 2021; Rempfert et al., 2017; Schrenk et al., 2013) that preserve biosignatures (Ivarsson et al., 2018; Newman et al., 2020; Onstott et al., 2019). Near-surface, carbonate-forming groundwaters that connected to deep subsurface habitable refugia (Michalski et al., 2013; Tarnas et al., 2018) could have transported life towards the surface, where it may be preserved in near-surface mineralized veins of carbonate and other alteration minerals. After billions of years of erosion, the alteration material formed in these possible subsurface habitable environments, including the carbonates in Jezero crater, would now be exposed at the surface. It is also possible that carbonates in the margin fractured unit formed via both groundwater alteration and fluviolacustrine sediment deposition in multiple distinct episodes, as discussed in Horgan et al., (2020a). In-situ investigation by the Perseverance rover will constrain how these carbonates formed, the habitability of their formation conditions, and the likelihood of this material to preserve biosignatures. Subsurface habitability should be considered in this investigation, as rocks from ancient subsurface habitable environments in Jezero crater may preserve biosignatures.

968

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974 with the National Aeronautics and Space Administration (80NM0018D0004). © 2021. All rights
975 reserved

976

977 **Data availability**

978 Datasets used in this research will be made available on Harvard Dataverse.

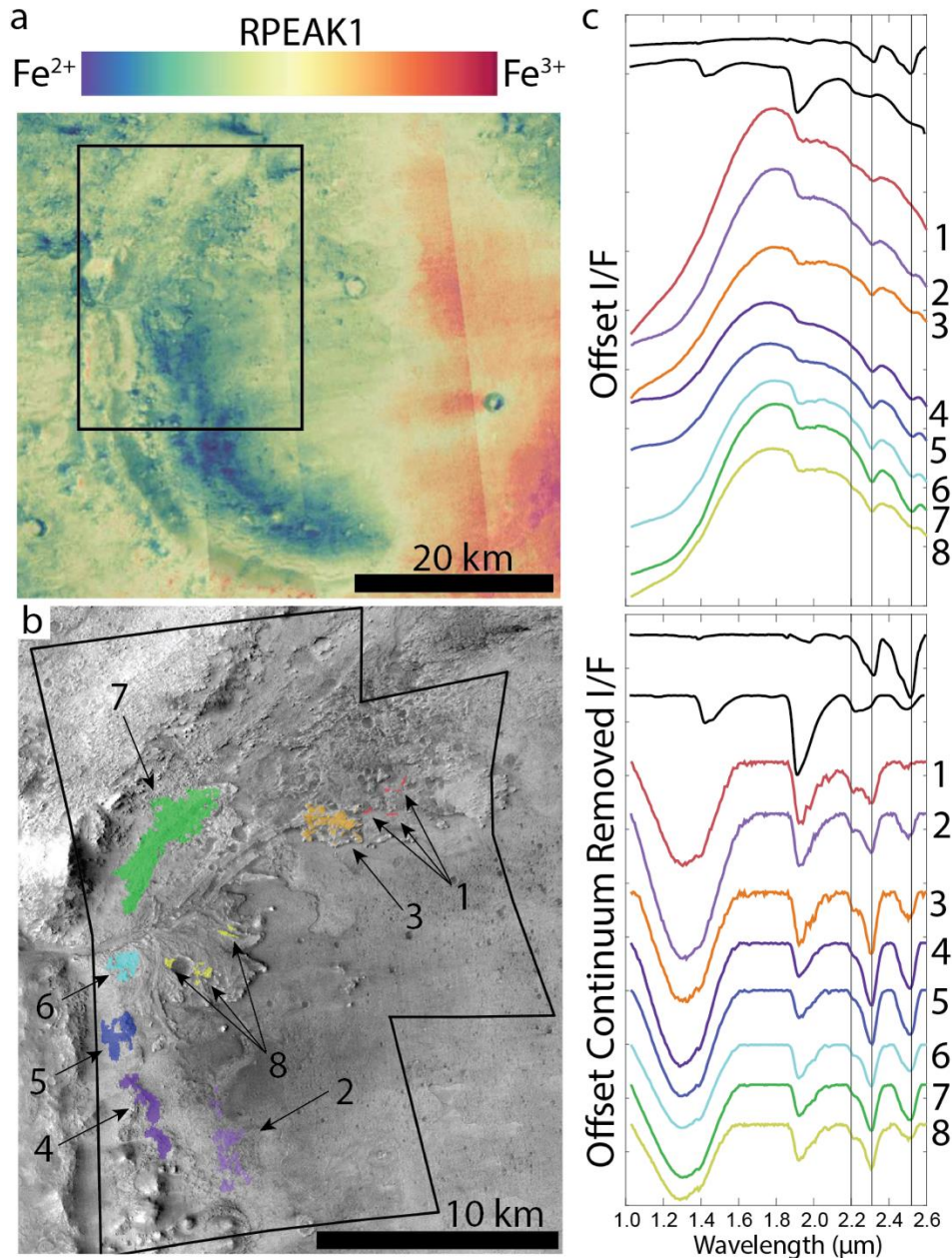


Figure 1 | Overview of Jezero crater carbonate-bearing rocks. (a) RPEAK1 parameter map of CRISM Multispectral TRDR image mosaic, which corresponds to presence of ferric (red color) and ferrous (blue color) Fe. Ferric iron is attributed to presence of martian dust, which occludes spectral signals in the visible-to-near-infrared wavelengths measured by CRISM. The western region of Jezero crater has lower dust coverage, and therefore be less spectrally dominated by dust. (b) Carbonate-bearing rocks are present in the delta truncated curvilinear layered, delta blocky, and undifferentiated smooth units (8), the delta layered rough unit (3), the crater floor fractured 2 unit (2), the margin fractured unit and proximal large aeolian bedforms (4-7) described in Stack et al. (2020). Silica-bearing material is identified in the smooth dark-toned material described in Tarnas et al. (2019). The combined footprints of CRISM images HRL000040FF and FRT000047A3 are shown as black lines. The regions of interest from which

these spectra were extracted are analyzed in greater detail in the Figures covering locations shown in Figure 2. CTX mosaic from Dickson et al. (2018). The colors show the pixels from which the spectra shown in (c) were extracted. Colors and numbers correlate between (b) and (c). (c) Spectra from the University of Massachusetts at Amherst processed CRISM data shown with (bottom) and without (top) continuum removal.

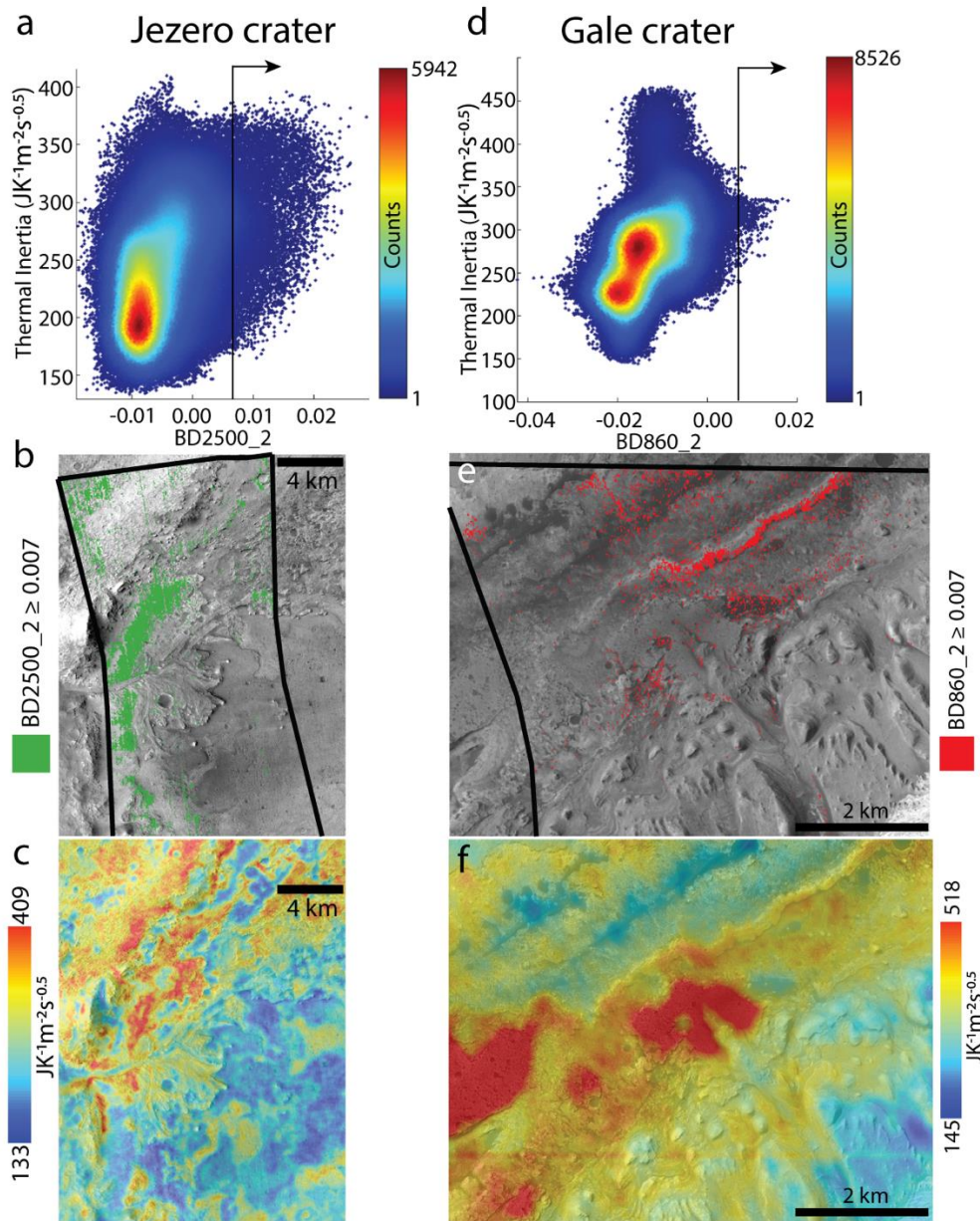


Figure 2 | Similarities between the margin fractured unit in Jezero crater and Vera Rubin ridge in Gale crater. Band parameters are from Viviano-Beck et al., (2014). (a) Relationship between band depth of the 2.5 μm absorption (BD2500_2) in CRISM image HRL000040FF and THEMIS quantitative thermal inertia (TI). The points to the right of the vertical line are the ones that appear in part (b). Pixels with the strongest 2.5 μm absorption band depths also have high TI, except for in regions covered by dunes. (b) CRISM pixels with BD2500_2 values ≥ 0.007 ,

1004 consistent with the presence of carbonate. The black lines show the outline of CRISM image
1005 HRL000040FF. (c) Quantitative TI of the same area shown in (b). (d) Relationship between band
1006 depth of the 0.86 μm absorption (BD860_2) in CRISM image FRT0000B6F1 and THEMIS
1007 quantitative (TI). The points to the right of the vertical line are the ones that appear in part (e).
1008 Pixels with the strongest 0.86 μm absorption band depths also have high-to-moderate TI. (e)
1009 CRISM pixels with BD860_2 values ≥ 0.007 , consistent with the presence of hematite. The
1010 black lines show the outline of CRISM image FRT0000B6F1. (f) Quantitative TI of the same area
1011 shown in (e).
1012

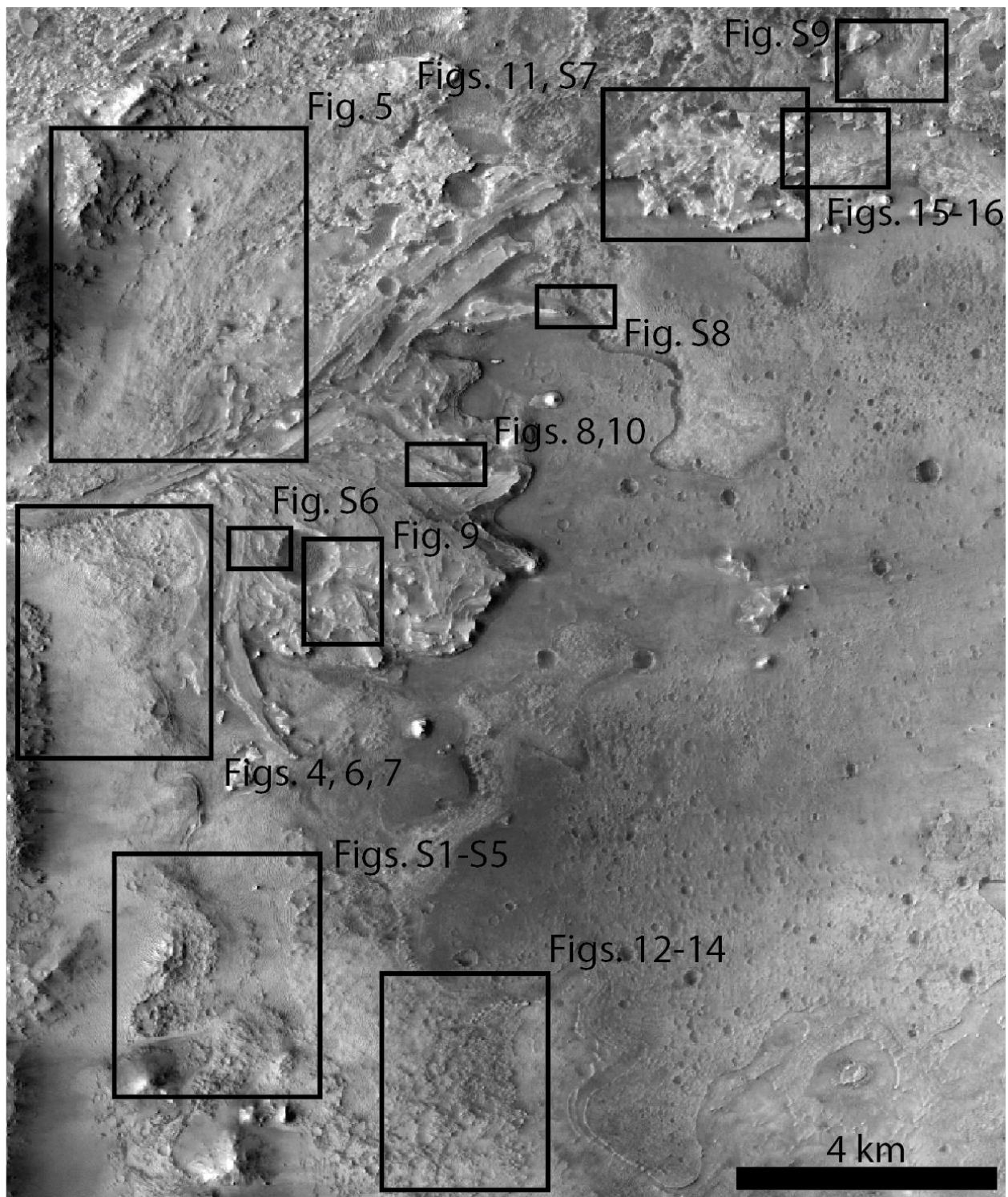


Figure 3 | Locations of sites characterized in this study.

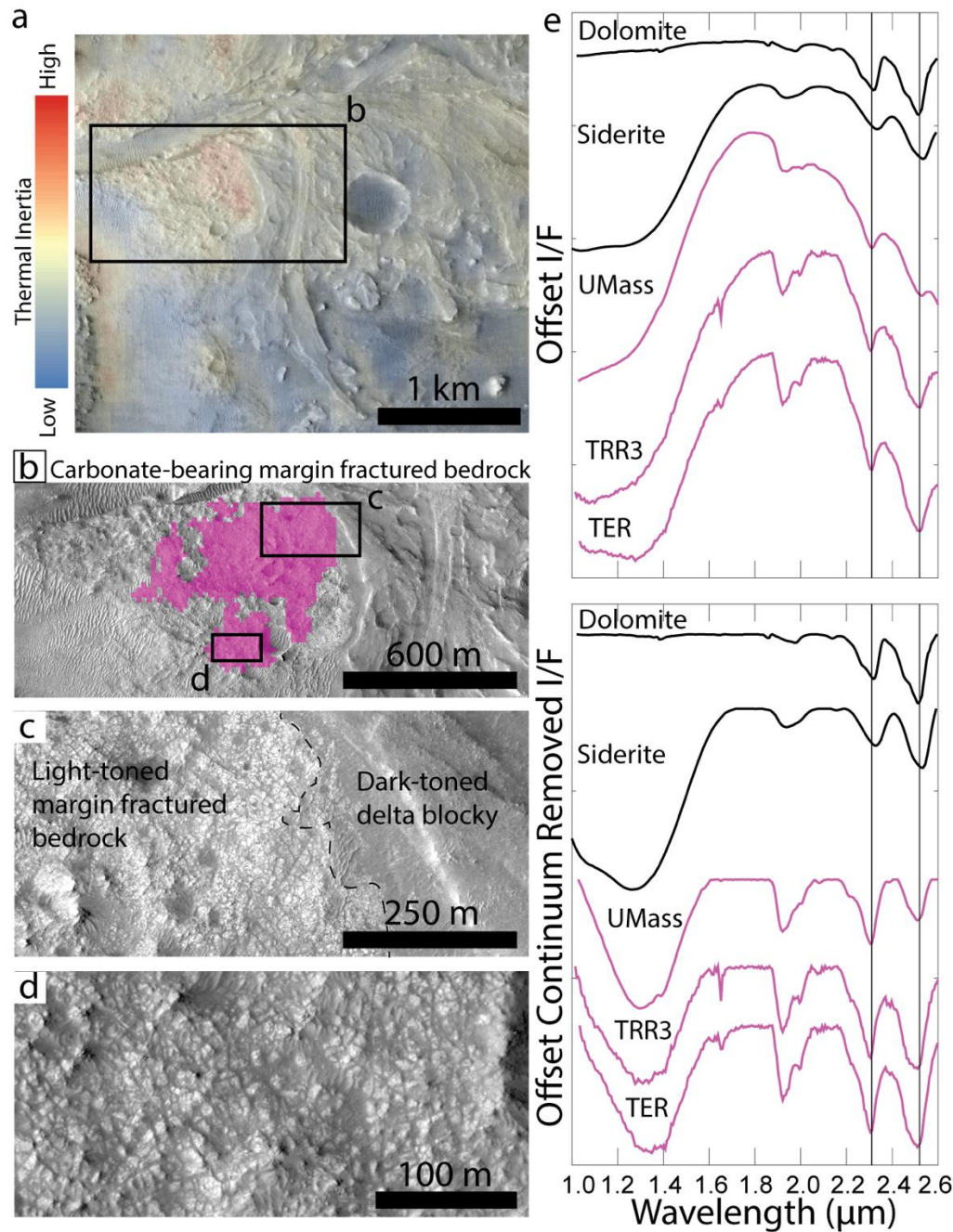


Figure 4 | Carbonate and phyllosilicate-bearing bedrock in the margin fractured unit. (a) THEMIS qualitative thermal inertia map of region of interest. (b) CRISM pixels (pink) for spectra shown in (e). These CRISM pixels cover light-toned fractured bedrock with notably high thermal inertia. (c) Enlarged view of light-toned margin-fractured unit bedrock and contact with dark-toned delta blocky unit material. (d) Enlarged view of light-toned margin fractured unit bedrock. (e) UMass, TRR3, And TER CRISM spectra of pink pixels shown in (b). The 2.3 and 2.5 μm features are consistent with the presence of carbonate-bearing rock. The ratio of the 2.3/2.5 μm absorptions is consistent with presence of some phyllosilicate-bearing material. The absorption centered at 1.0 μm may be due to presence of olivine or presence of siderite.

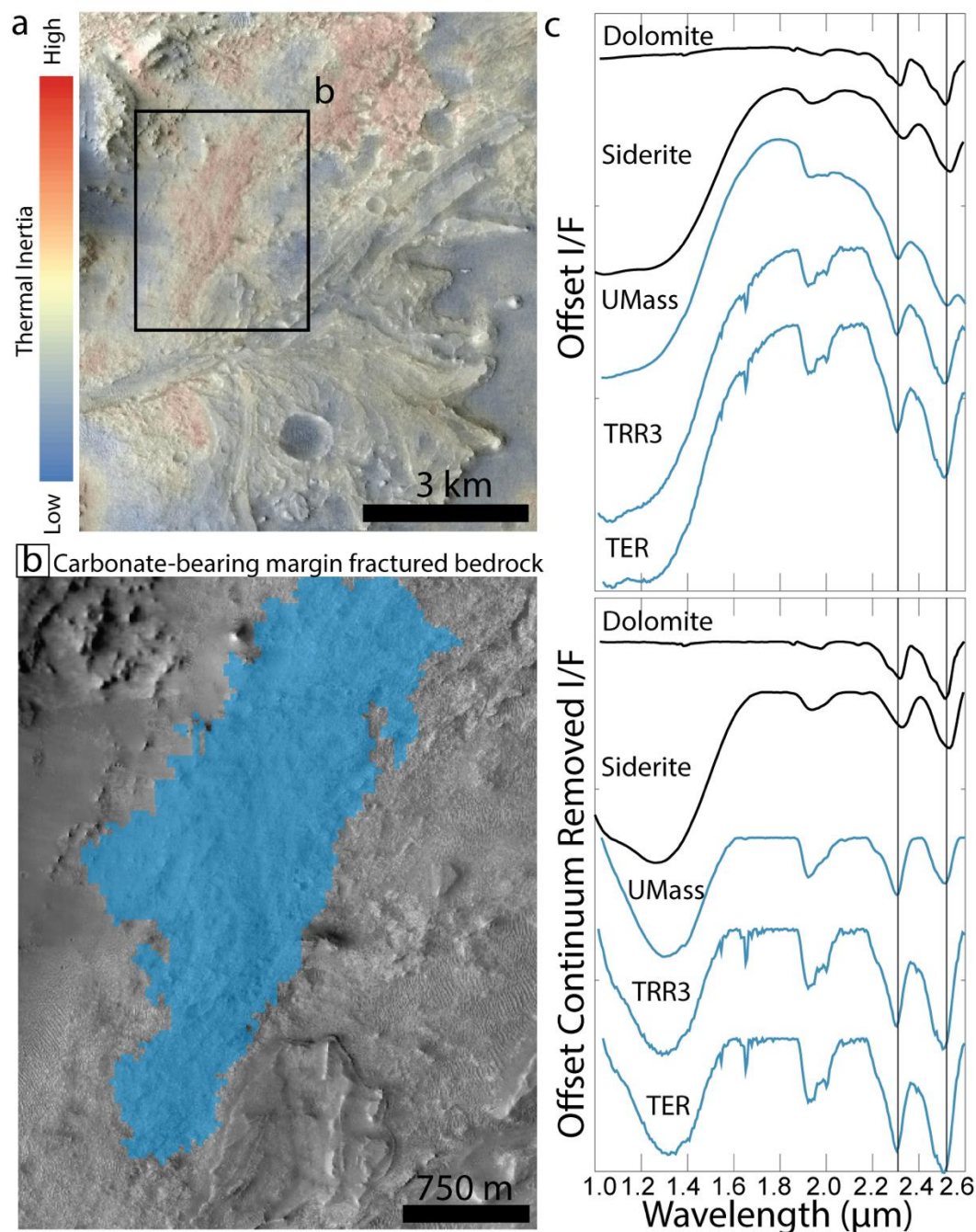


Figure 5 | Carbonate and phyllosilicate-bearing bedrock in the margin fractured unit. (a) THEMIS qualitative thermal inertia map of region of interest. (b) CRISM pixels (blue) for spectra shown in (c). These CRISM pixels cover light-toned fractured bedrock with notably high thermal inertia. (c) UMass, TRR3, And TER CRISM spectra of blue pixels shown in (b). The spectral and geologic interpretation is the same as is described in Figure 4.

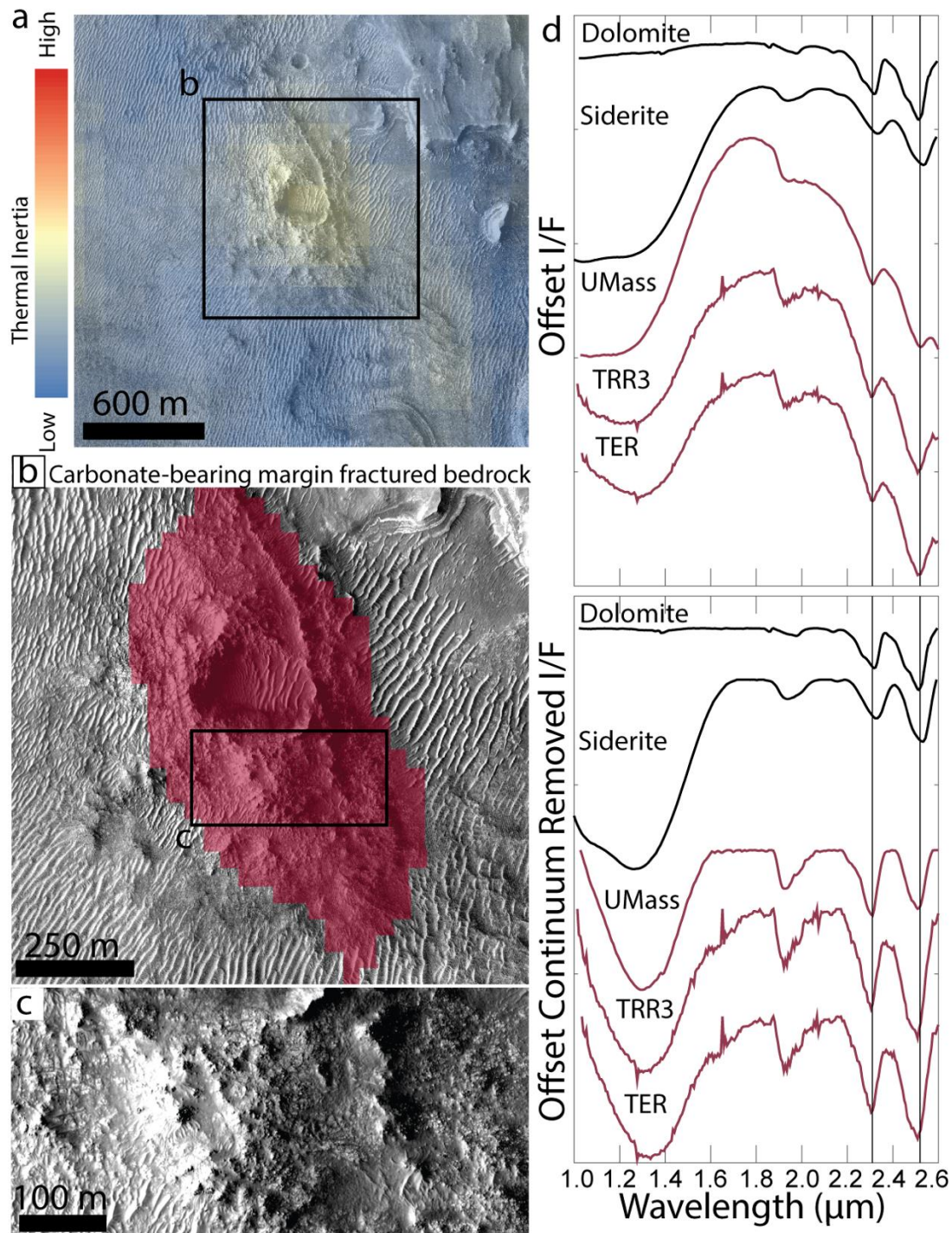
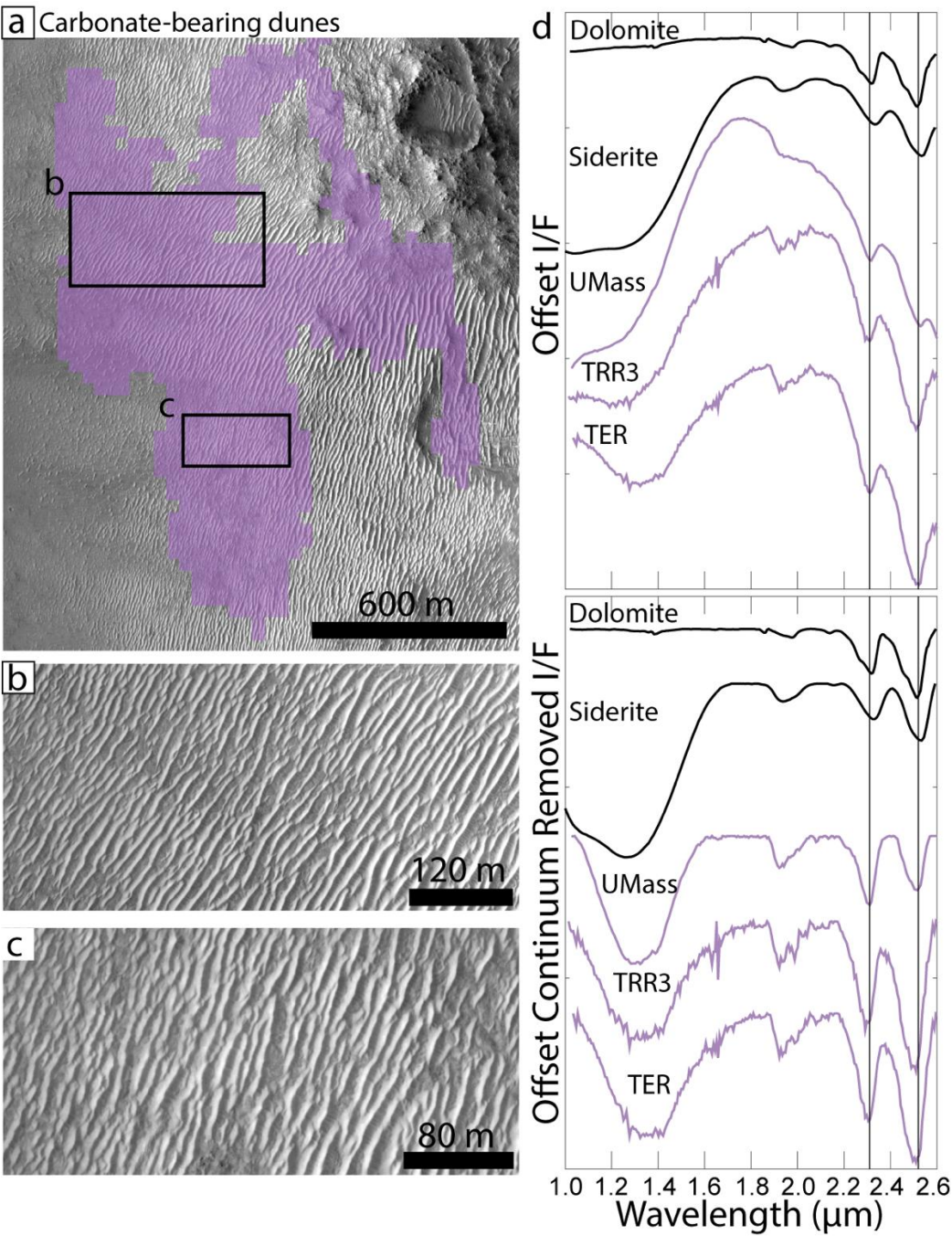


Figure 6 | Carbonate and phyllosilicate-bearing bedrock in the margin fractured unit. (a) THEMIS qualitative thermal inertia map of region of interest. (b) CRISM pixels (dark red) for spectra shown in (e). These CRISM pixels cover light-toned fractured bedrock with notably high thermal inertia. (c) Enlarged view of light-toned margin-fractured unit bedrock and contact with dark-toned delta blocky unit material. (d) UMass, TRR3, And TER CRISM spectra of dark red pixels shown in (b). The spectral and geologic interpretation is the same as is described in Figure 4.

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Figure 7 | Carbonate and phyllosilicate-bearing aeolian bedforms proximal to margin fractured unit. (a) CRISM pixels (purple) for the spectra shown in (d). These pixels cover aeolian bedforms immediately west of the margin fractured bedrock shown in Figure 6. (b-c) Enlarged views of carbonate and phyllosilicate-bearing aeolian bedforms. (d) UMass, TRR3, And TER CRISM spectra from purple pixels shown in (a). The spectral interpretation is the same as described in Figure 4. This aeolian material is therefore interpreted to derive from the proximal margin fractured unit bedrock.

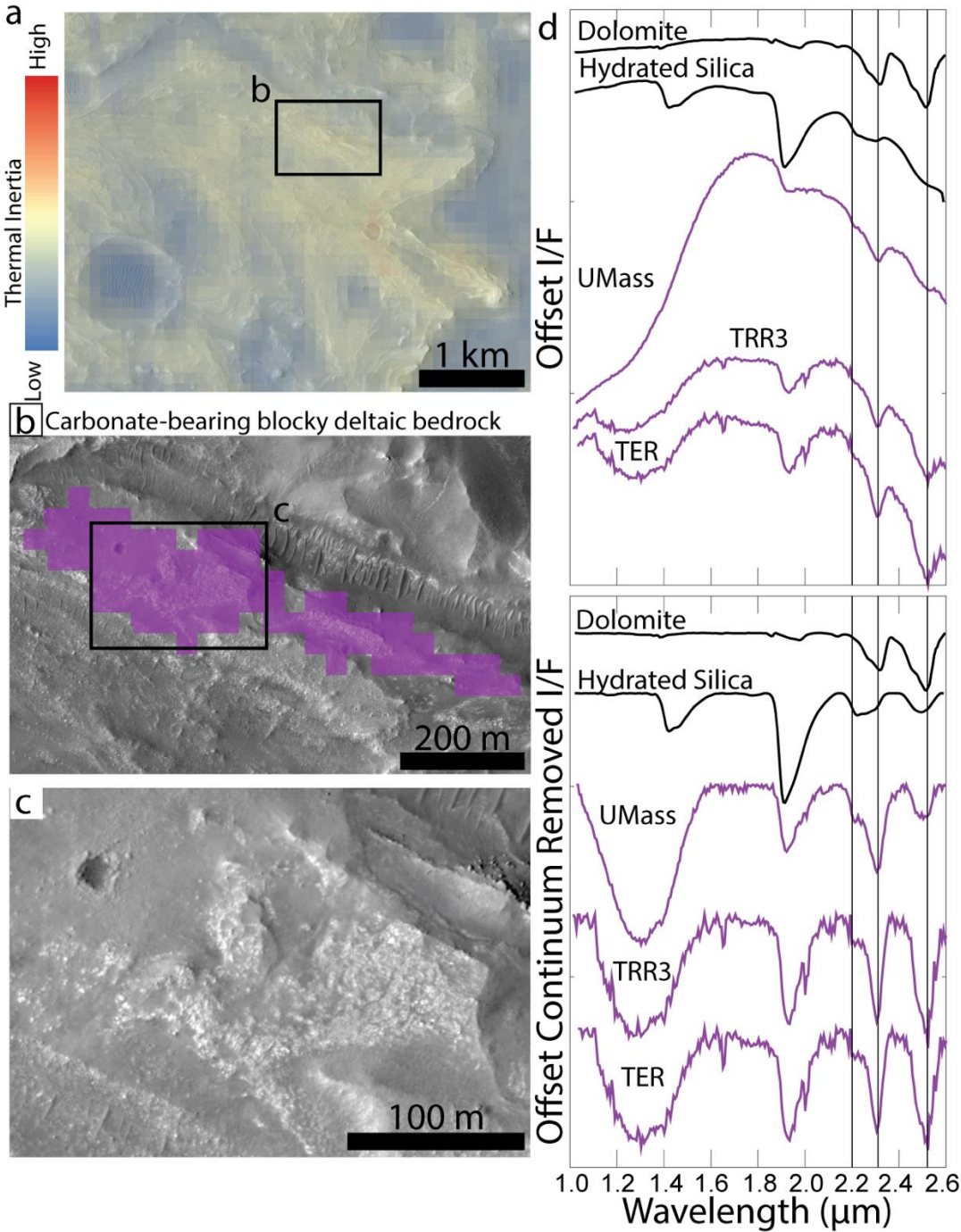


Figure 8 | Carbonate and phyllosilicate-bearing rock in the western Jezero delta blocky unit.

(a) THEMIS thermal inertia map of region of interest. (b) CRISM pixels (purple) for spectra shown in (d), which cover light and dark-toned bedrock in the delta blocky unit. (c) Enlarged view of light and dark-toned bedrock. (d) UMass, TRR3, And TER CRISM spectra from purple pixels shown in (b). The 2.3 and 2.5 μm absorptions are consistent with the presence of carbonate-bearing rock. The 2.3/2.5 μm absorptions are consistent with the presence of phyllosilicate-bearing material. The small absorption at 2.2 μm is consistent with the presence

of either hydrated silica and/or Al-phylosilicate. Overall, this composition resembles the crater floor fractured 2 unit, but with a stronger 2.3 μm feature, consistency with stronger spectral presence of phyllosilicates.

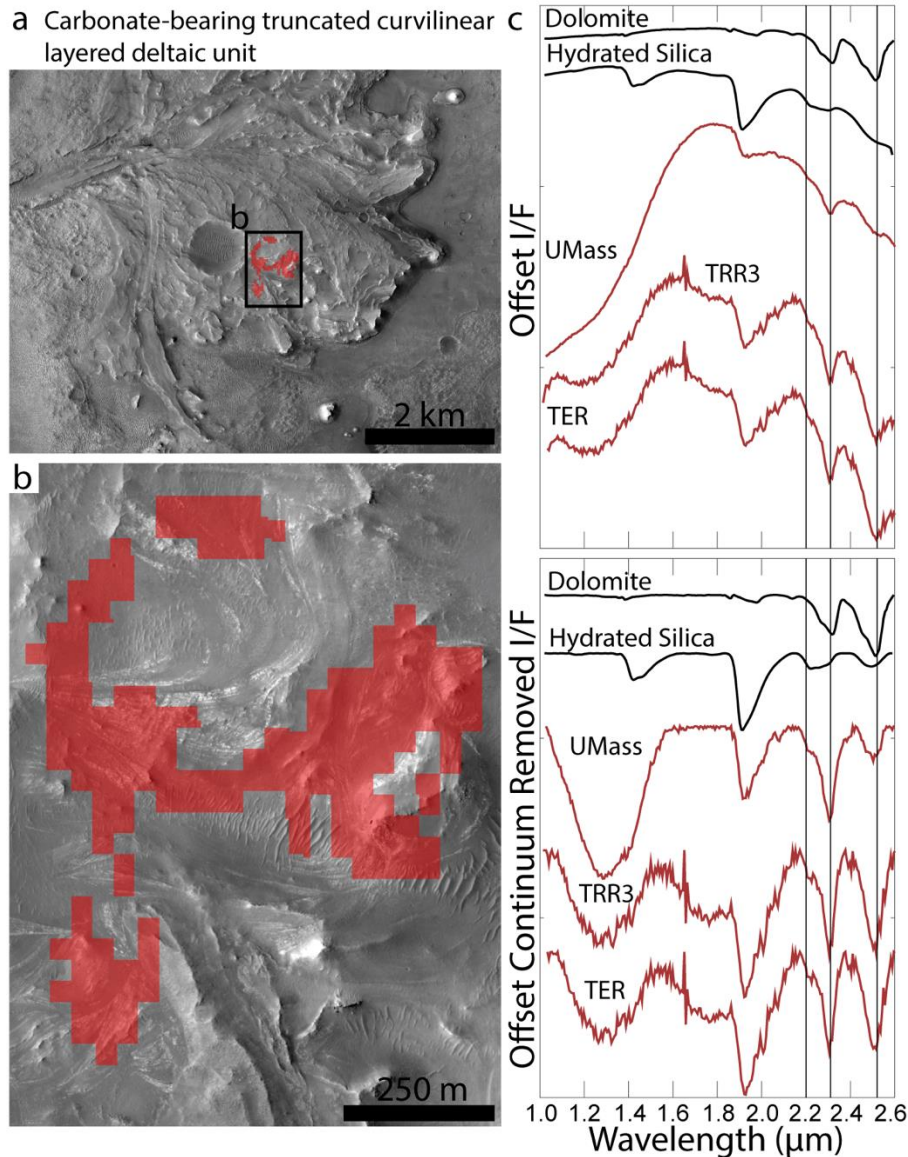


Figure 9 | Carbonate and phyllosilicate-bearing rock in the western Jezero delta truncated curvilinear layered unit. (a) Regional view of reported compositional detections from CRISM pixels shown in red. (b) CRISM pixels (red) for spectra shown in (c), which cover possible point bar strata in the delta truncated curvilinear layered unit (Goudge et al. 2018). (c) UMass, TRR3, And TER CRISM spectra from red pixels shown in (a & b). The spectral interpretation is the same as described in Figure 8, but is present on a different deltaic unit.

a Carbonate-bearing undifferentiated smooth deltaic unit cover

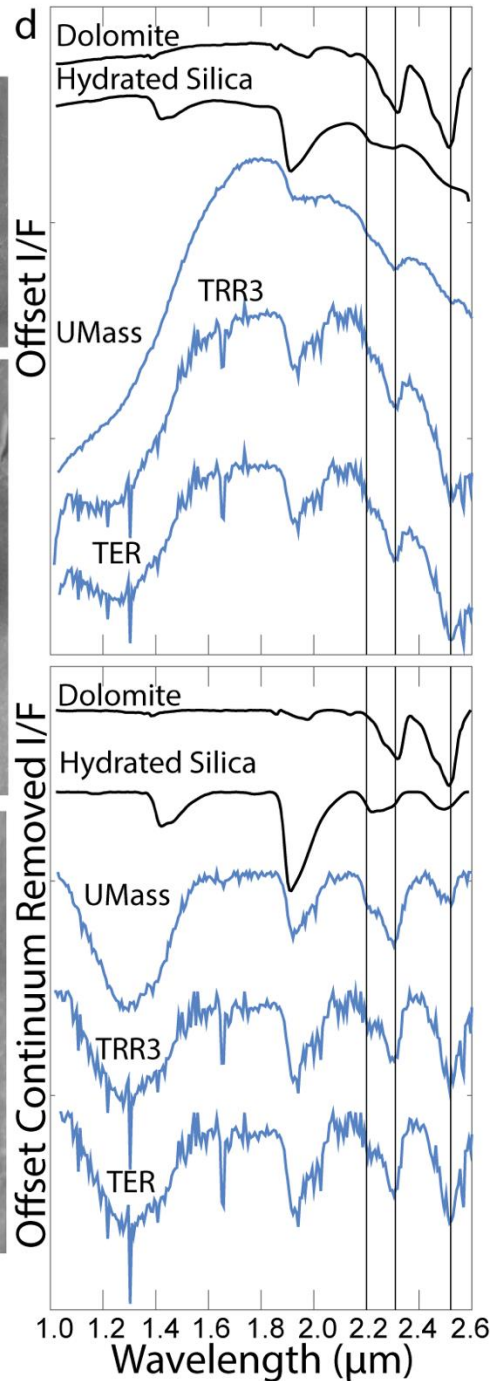
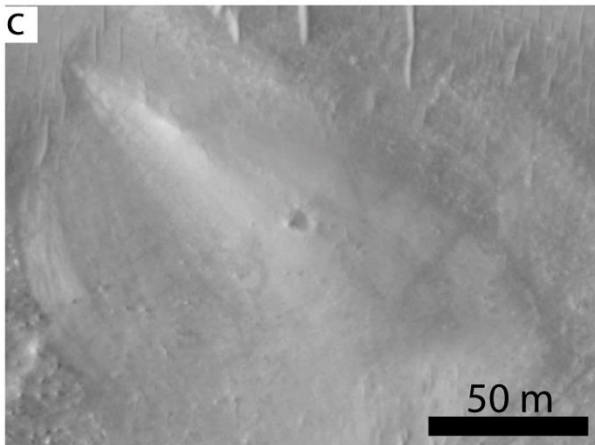
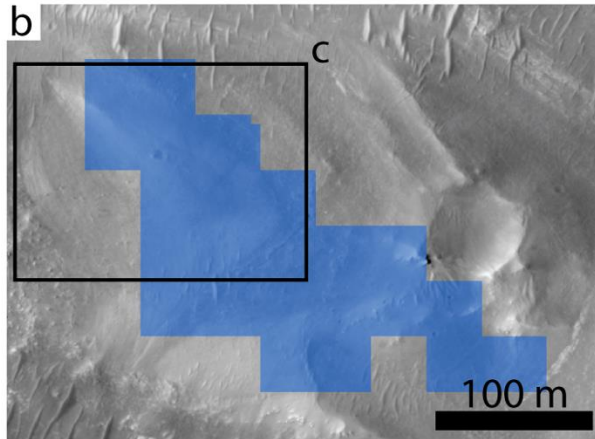
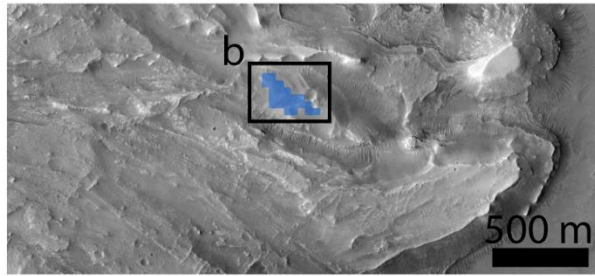


Figure 10 | Carbonate and phyllosilicate-bearing rock in the western Jezero delta undifferentiated smooth unit. (a) Regional view of reported compositional detections from CRISM pixels shown in blue. (b) CRISM pixels (blue) for spectra shown in (c), which cover primarily dark-toned bedrock in the delta undifferentiated smooth unit. (c) UMass, TRR3, And TER CRISM spectra from blue pixels shown in (a & b). The spectral interpretation is the same as described in Figure 8, but is present on a different deltaic unit.

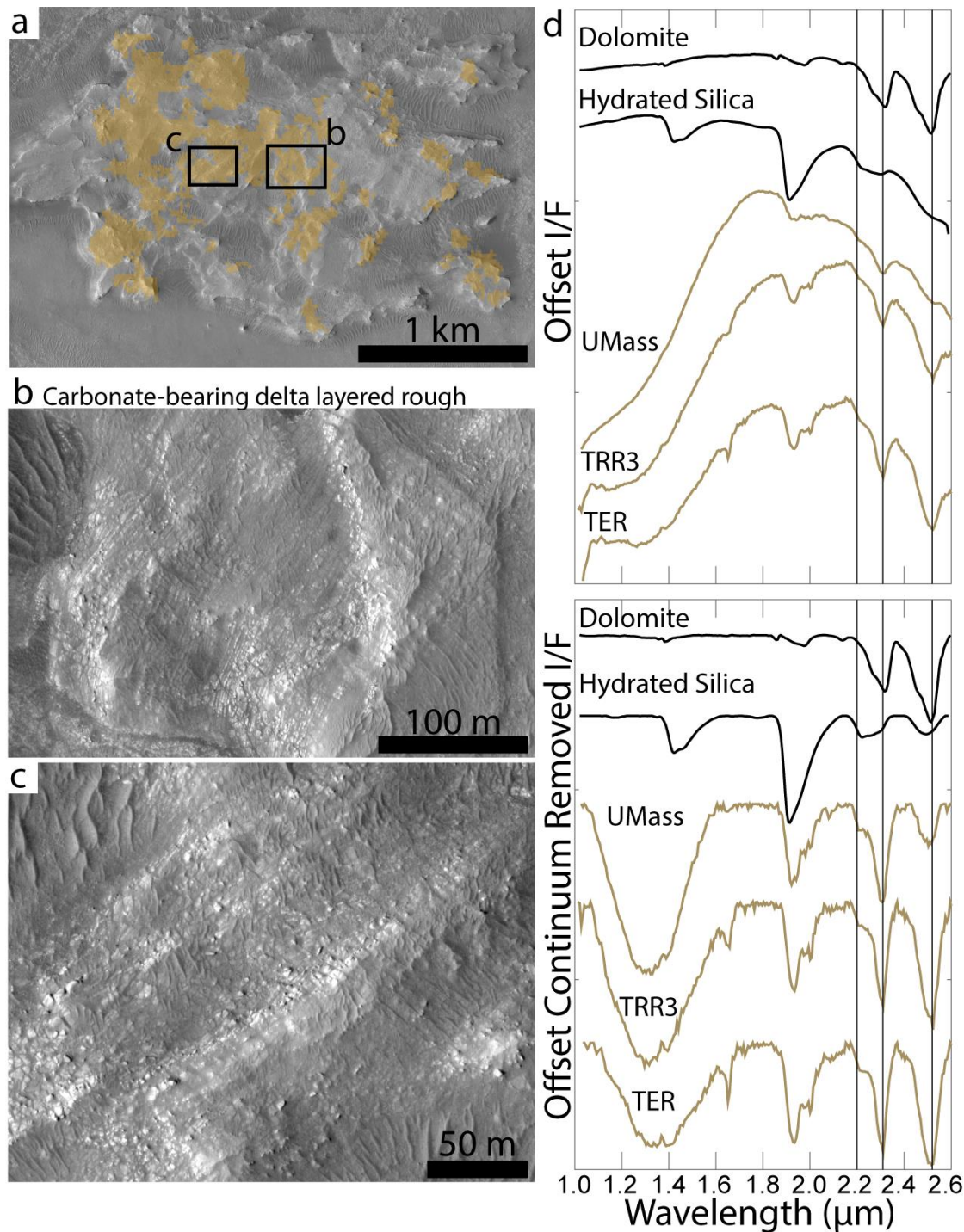


Figure 11 | Carbonate and phyllosilicate-bearing rock in the northern Jezero delta layered rough unit. (a) Regional view of reported compositional detections from CRISM pixels shown in orange, which cover the northern delta. (b & c) Enlarged views of layered light-toned bedrock in northern delta. (d) UMass, TRR3, And TER CRISM spectra from orange pixels shown in (a & b). The spectral interpretation is the same as described in Figure 8, but is present on a different deltaic unit.

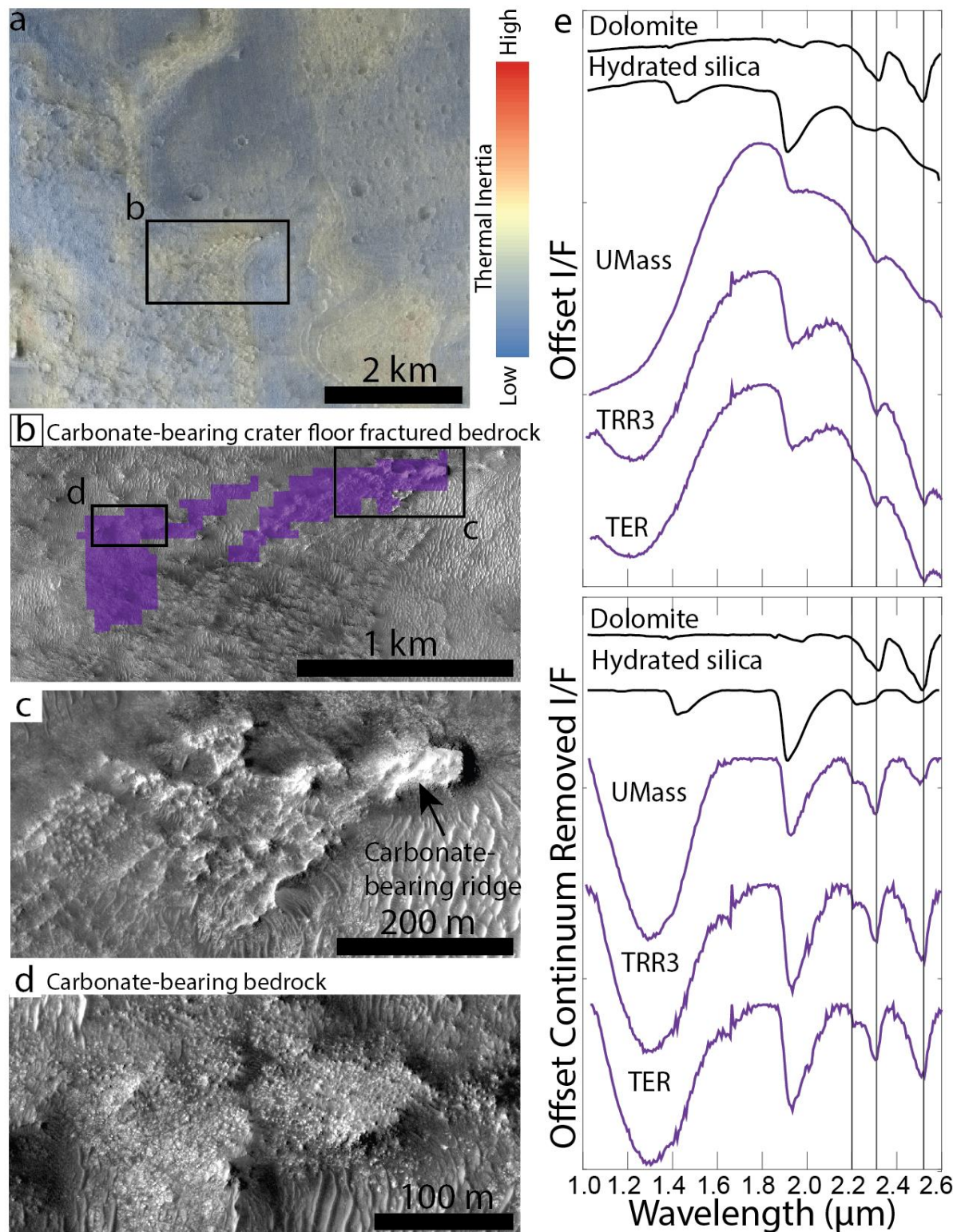


Figure 12 | Carbonate, silica, and olivine-bearing bedrock. (a) THEMIS qualitative thermal inertia map from Fergason et al. (2006), coregistered to CTX and HiRISE mosaics by Dickson et al. (2018). Warmer colors correspond to more consolidated (warmer colors) and less

consolidated (cooler colors) materials. (b) CRISM pixels (purple) on HiRISE image from which the spectra shown in (e) are derived. The CRISM pixels were selected based on THEMIS thermal inertia and HiRISE data to cover bedrock in the crater floor fractured 2 unit. (c) Enlarged view of a ridge in the crater floor fractured 2 unit (Stack et al. 2020). (d) Enlarged view of rubbly bedrock in the crater floor fractured 2 unit. (e) Spectra from UMass, TRR3, And TER processed CRISM data (purple) extracted from the pixels shown in (b) and compared to laboratory spectra of dolomite (RELAB ID C1CY07) and hydrated silica. The spectra are consistent with the presence of carbonate-bearing rock based on the presence of 2.3 and 2.5 μm features. The ratio of 2.3/2.5 μm band depth and narrow shape of the 2.3 μm feature are also consistent with the presence of phyllosilicate-bearing rock. The absorption at 2.2 μm , which combines with the 2.3 μm absorption to create a broad band, is consistent with the presence of hydrated silica and/or Al-phyllosilicate. The absorption centered at 1.0 μm is consistent with the presence of olivine-bearing rock, which is widespread throughout this unit. Hydrated silica is a product of carbonation of olivine (Falk & Kelemen, 2015). The thermal inertia, visible texture and topography, geologic context, and VNIR spectra of this outcrop are consistent with carbonate, hydrated silica, and olivine-bearing bedrock.

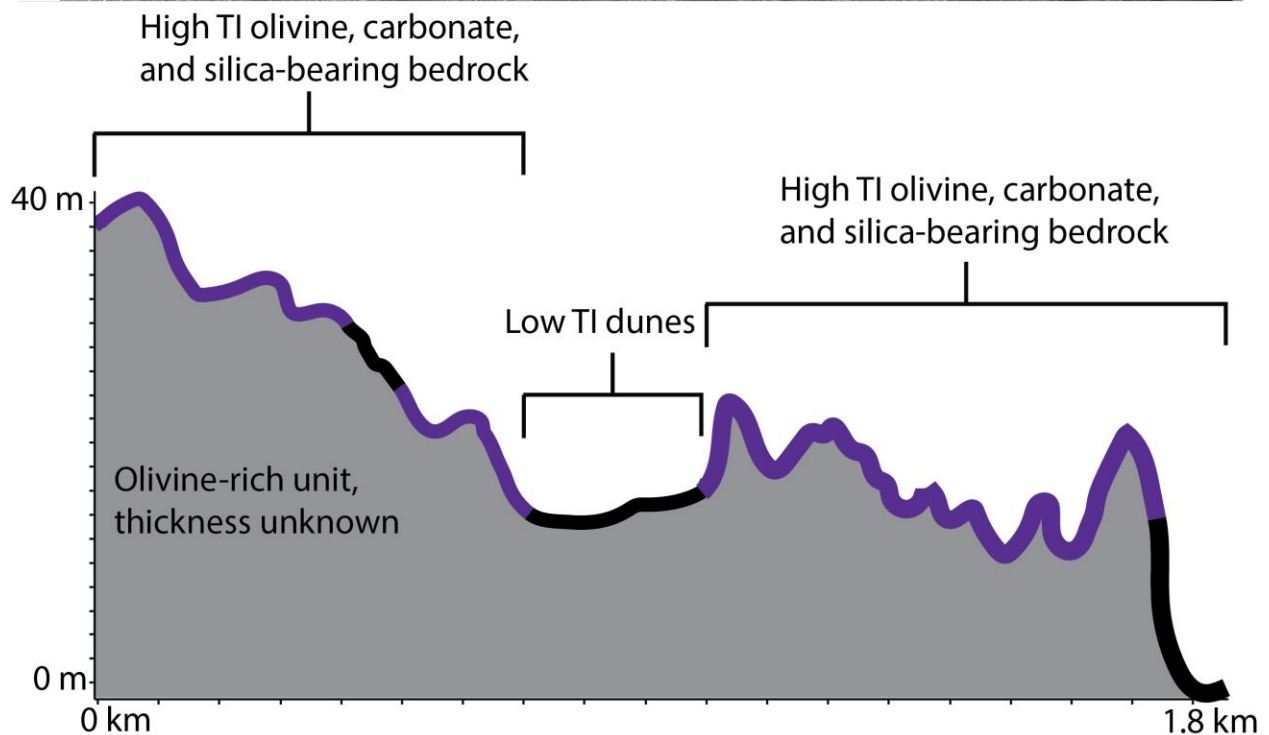
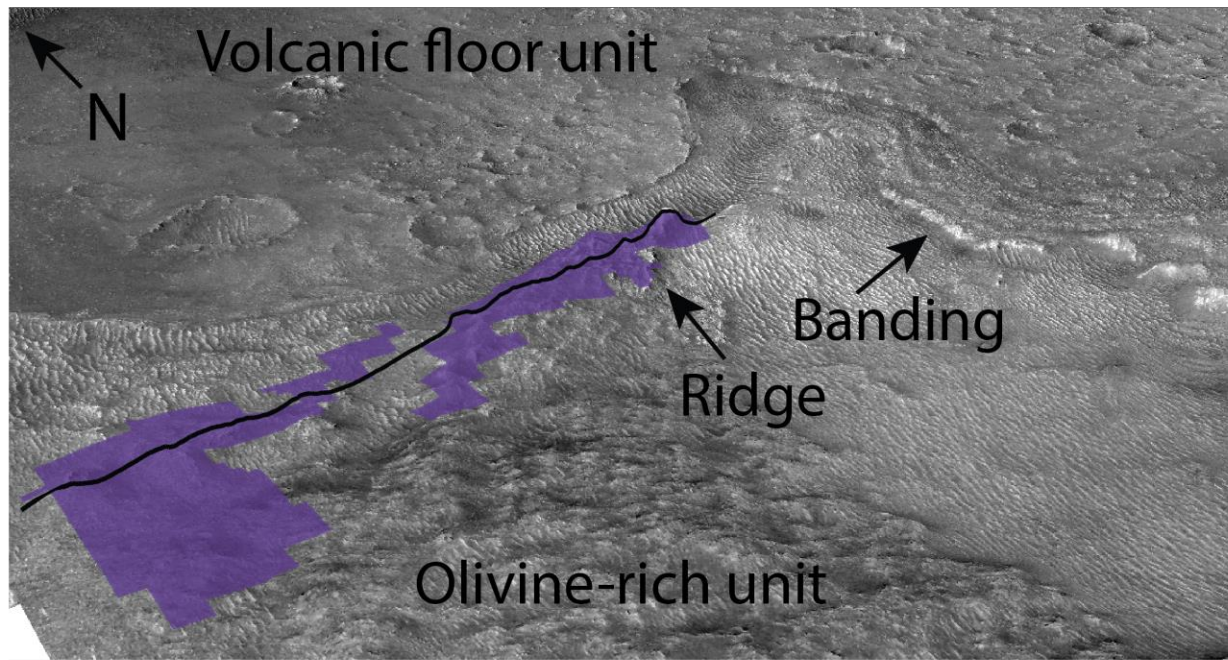


Figure 13 | Olivine, carbonate, and silica-bearing ridge in crater floor fractured 2 unit. (top) HiRISE mosaic overlay on HiRISE digital elevation model (DEM). The ridge occurs near the contact between the crater floor fractured 2 and undifferentiated smooth floor units as described by Stack et al. (2020). Banding can be seen at this contact towards the east. (bottom) The spectra shown in Figure 12 were extracted from topographic highs with moderate thermal inertia, which is consistent with the hypothesis that this material is olivine, carbonate, phyllosilicate, and silica-bearing bedrock.

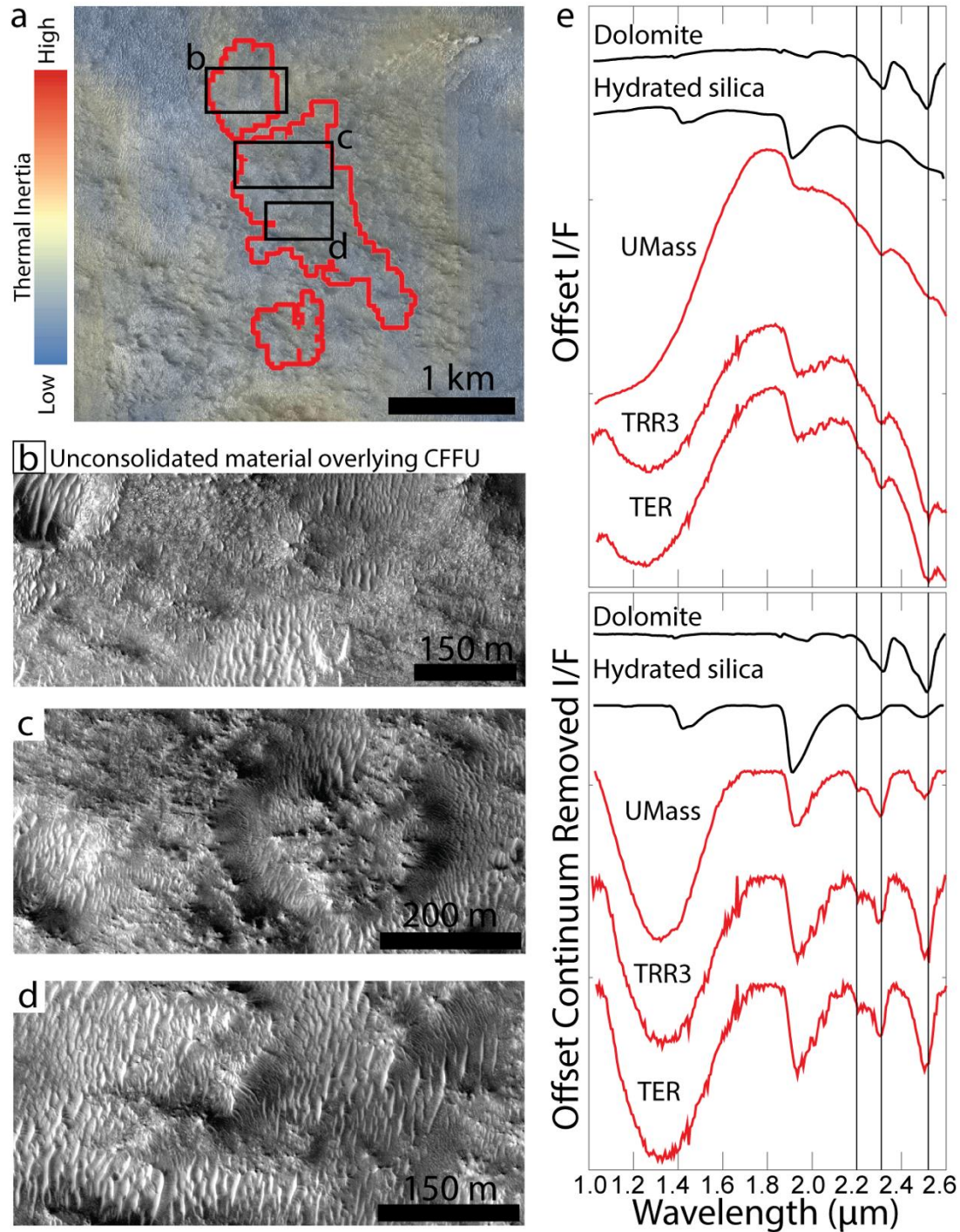


Figure 14 | Olivine, carbonate, phyllosilicate, and silica-bearing unconsolidated material in crater floor fractured 2 unit. (a) THEMIS qualitative thermal inertia map. The CRISM pixels for the spectra in (e), which are within the red outlines, cover dunes and unconsolidated dark-toned material that has low thermal inertia. (b) Enlarged view of dunes and dark-toned unconsolidated dark-toned material overlying bedrock, which in some locations protrudes through this material. (c) Enlarged view of dunes and knobby bedrock that is covered by dark-toned unconsolidated material. (d) Enlarged view of dunes and unconsolidated dark-toned

material overlying bedrock. (e) UMass, TRR3, and TER processed CRISM spectra from pixels shown in (b) compared to library spectra of dolomite and hydrated silica. The spectral features are the same as those described for bedrock in Figure 12, implying that the dunes and unconsolidated dark-toned material are locally sourced and have the same composition as the proximal and underlying crater floor fractured 2 unit bedrock.

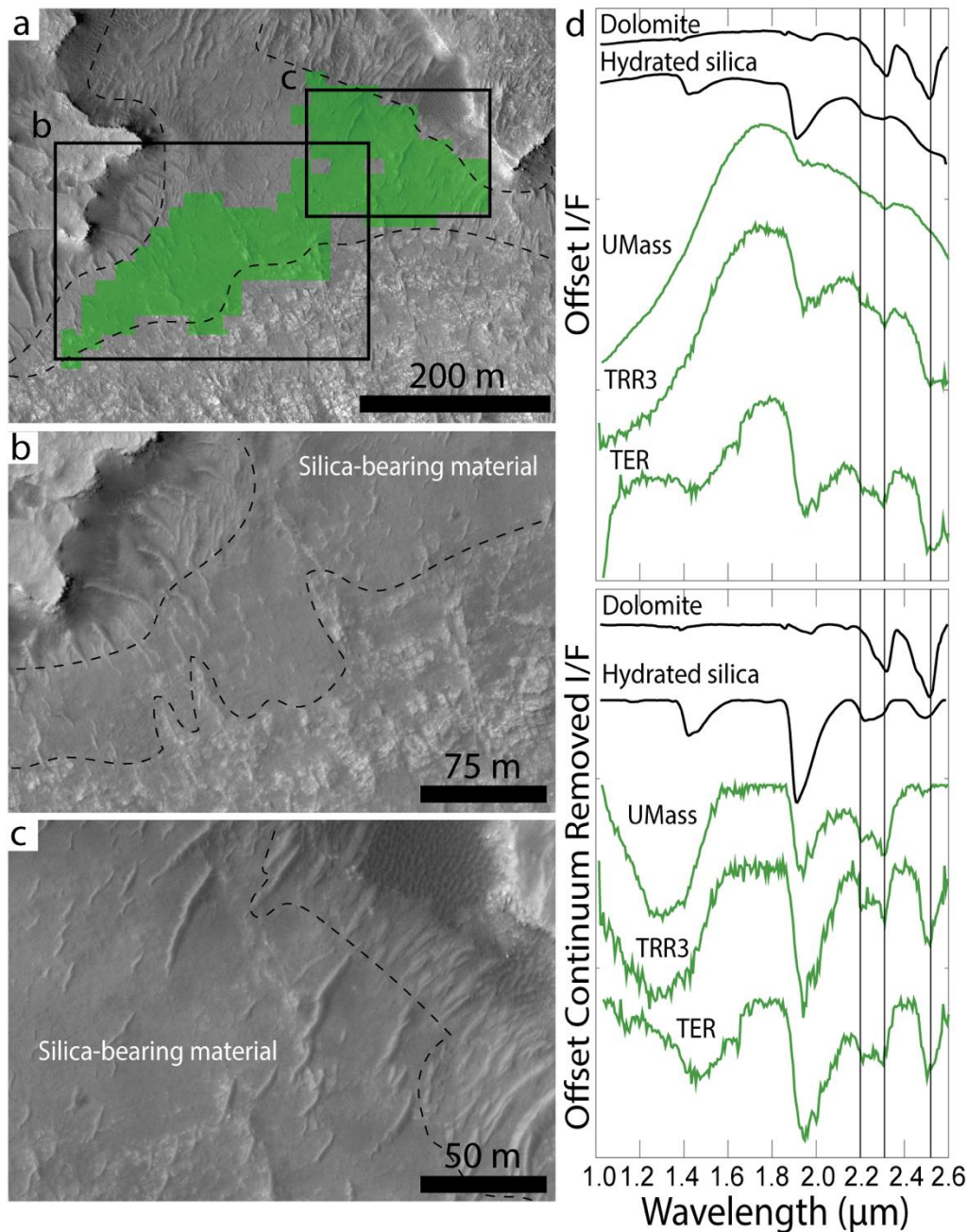


Figure 15 | Smooth dark-toned silica-bearing material overlying crater floor fractured unit. (a) CRISM pixels (green) for the spectra shown in (d). These pixels cover the smooth dark-toned

material reported to be silica-bearing by Tarnas et al. (2019). (b) Enlarged view of smooth dark-toned silica-bearing material overlying the crater floor fractured unit. (c) Another enlarged view of smooth dark-toned silica-bearing material overlying the crater floor fractured unit. (d) UMass, TRR3, And TER CRISM spectra from green pixels shown in (a). These spectra are consistent with hydrated silica and the Jezero crater hydrated silica detections reported in this material in different locations by Tarnas et al. (2019).

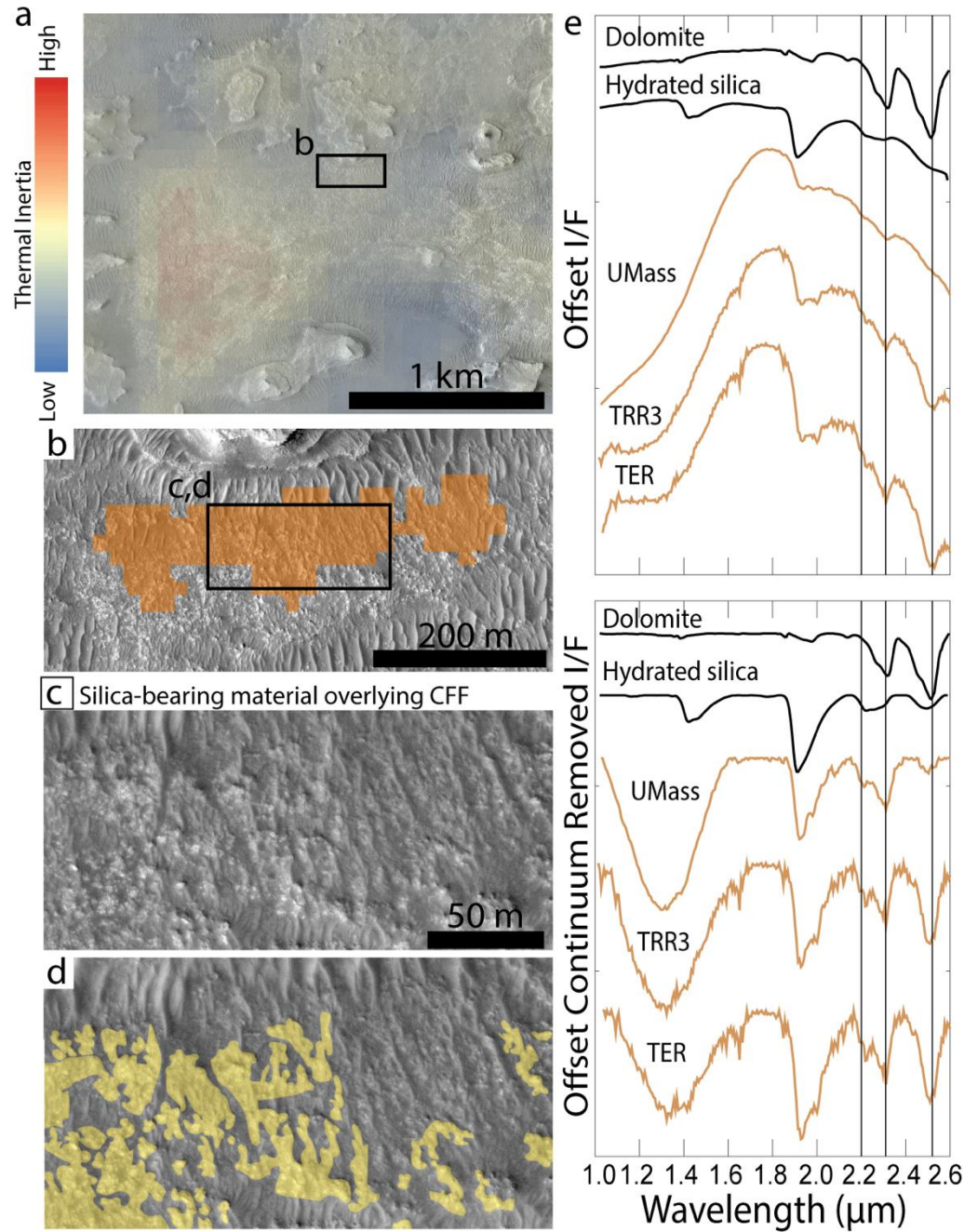


Figure 16 | Silica-bearing material overlying crater floor fractured unit. (a) THEMIS thermal inertia of target area. The area of interest (within the black outline) has moderate thermal inertia. (b) CRISM pixels for the spectra in (e) shown in orange. (c) Enlarged view of dark-toned

unconsolidated material overlying crater floor fractured unit bedrock. (d) Same image as (c), but with crater floor fractured unit bedrock mapped in yellow. Most of the area covered by the CRISM pixels shown in (a) is comprised of dark-toned unconsolidated material. (e) UMass, TRR3, And TER CRISM spectra of pixels shown in (a). The broad 2.2 μm feature is consistent with the presence of hydrated silica, which was also reported in smooth dark-toned material in Jezero crater by Tarnas et al. (2019). The additional 2.3 μm absorption that merges with the 2.2 μm features is consistent with the presence of phyllosilicate-bearing material. The geologic context, visible properties, and spectra of this outcrop are consistent with hydrated silica-bearing dark-toned unconsolidated material overlying phyllosilicate-bearing crater floor fractured unit bedrock.

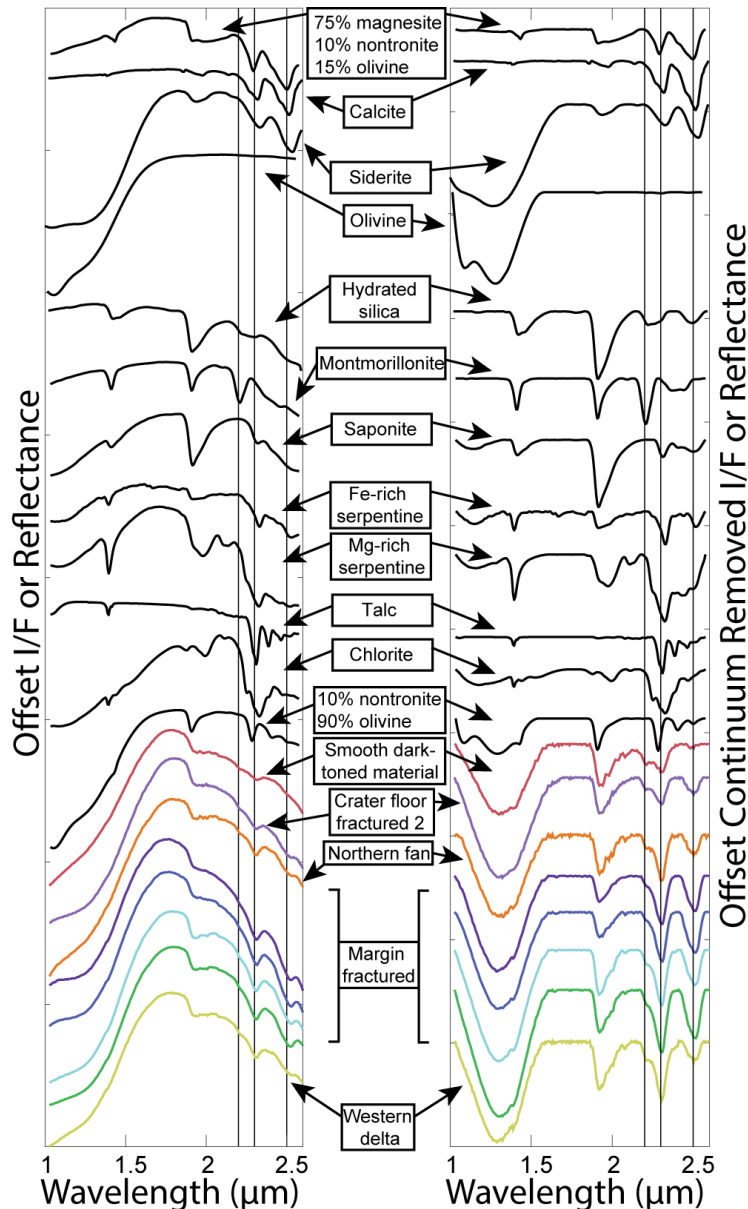


Figure 17 | Spectra of carbonate-bearing units in Jezero crater compared to library spectra. The colors of CRISM spectra correspond to those in Figure 1, while the library spectra are

shown in black. The CRISM spectra are likely mixtures of one or more of the minerals with displayed library spectra. Interpretations for mineral assemblages consistent with each CRISM spectrum are provided in the main text. The library spectra are RELAB IDs CABE01, BNR1BE052, BIR1BE030, C1CY07, CASA59, BIR1SR052A, C1RK67, 397S013, BKR1SR068, and 397S214.

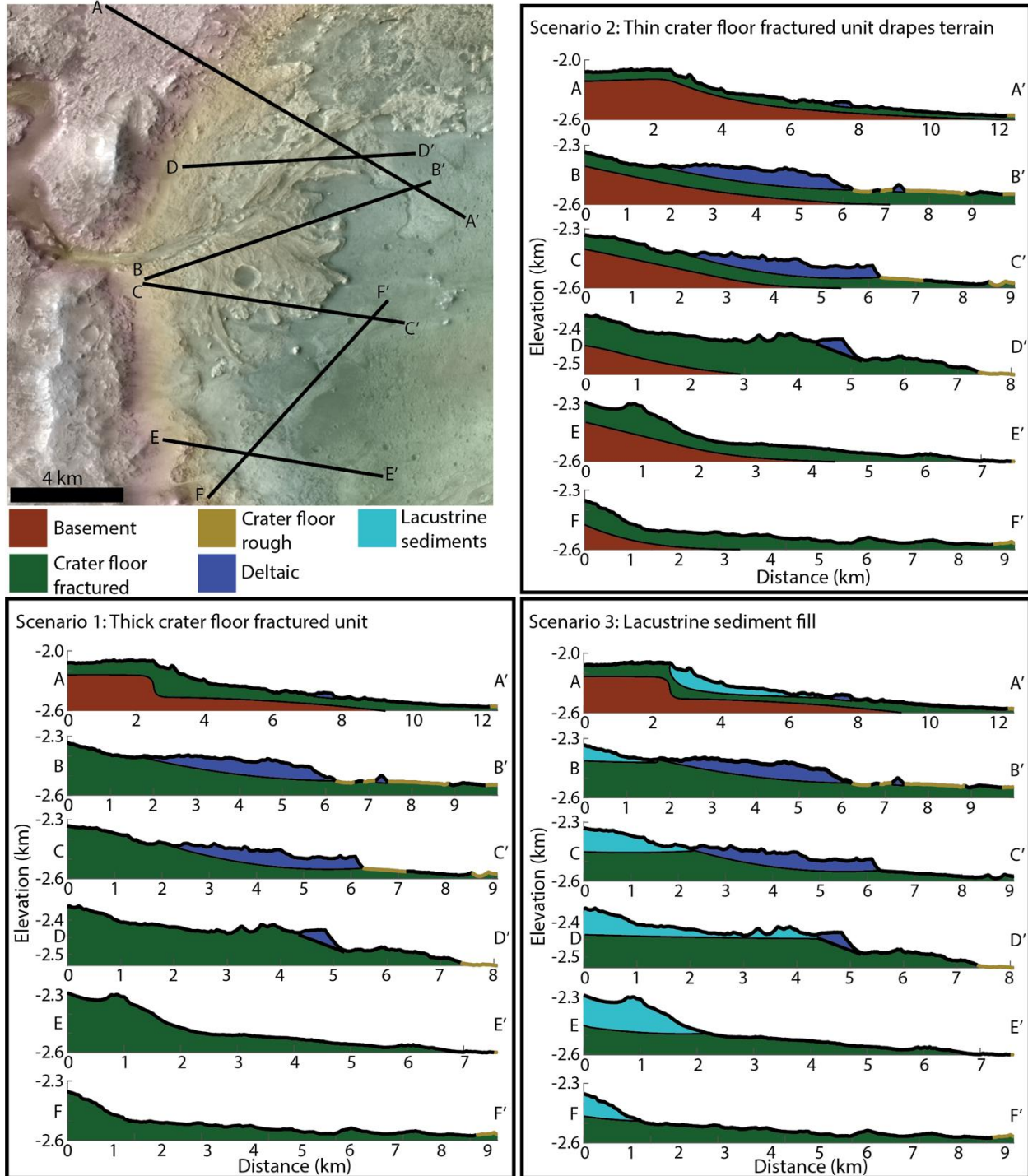


Figure 18 | Cross sections of topography in western Jezero crater, with different interpretations annotated. All illustrated hypothetical contacts are unconformable. Scenarios 1-3 are detailed in Section 5.2.

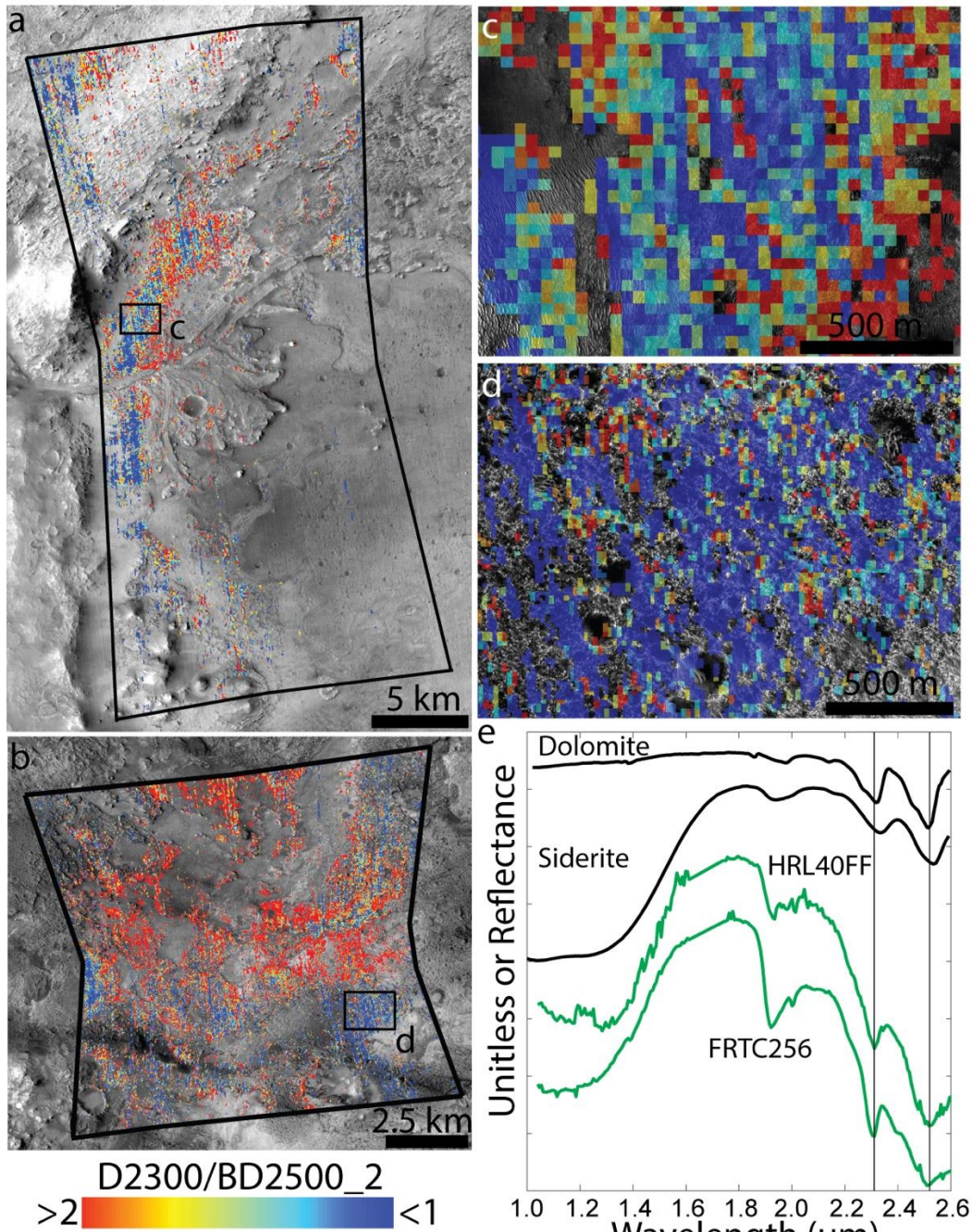


Figure 19 | Comparison of carbonate spectral properties in Jezero crater and elsewhere in the Nili Fossae region. (a) Ratio of the 2.3 and 2.5 μm absorptions (D2300/BD2500_2) in CRISM pixels of image HRL000040FF with BD2500_2 ≥ 0.005. As noted by Horgan et al., (2020a), the most widespread collection of pixels with the lowest 2.3/2.5 μm absorption band depth ratio is associated with the margin fractured unit. However, the 2.3/2.5 μm band depth ratio is also low

1177 in the ROB unit immediately outside of Jezero crater. The black outline shows the extent of
1178 CRISM image HRL000040FF. (b) Ratio of the 2.3 and 2.5 μm absorptions (D2300/BD2500_2) in
1179 CRISM pixels of image FRT0000C256 with BD2500_2 $\geq$ 0.005. There are large outcrops of
1180 carbonate with similar 2.3/2.5 μm band depth ratios as the margin fractured unit. All pixels
1181 shown here are associated with the ROB unit. Figure S10 shows an expanded view of 2.3/2.5
1182 μm band depth ratios in this region, mosaicking multiple CRISM images. (c) Enlarged view of
1183 box shown in (a). (d) Enlarged view of box shown in (b). (e) Spectral endmembers of pixels
1184 shown in (a) and (b) calculated via HySime (green, Section 3.1) compared to library spectra of
1185 carbonates (black). Both endmembers are consistent with a mixture of carbonate,
1186 phyllosilicate(s), and olivine.
1187

Observations	Fluvio-acustrine	Not fluvio-lacustrine
Strong VNIR carbonate absorptions mostly surrounded by maximum lake level contour in Jezero	✓	✗
Gradational relationship, no contact, with regional olivine-bearing unit that drapes into Jezero crater	✗	✓
Strongest margin fractured VNIR carbonate absorptions are in the highest thermal inertia bedrock	~	✓
Equivalent 2.3/2.5 µm band depth ratios inside Jezero crater and elsewhere in Nili Fossae	~	~
Thickness of margin fractured unit relative to regional olivine-bearing unit	✓	~
Presence of primary minerals in margin fractured unit	✓	✓
Gradational relationship, no visible contact, with carbonate-bearing crater floor fractured 2 unit	~	~
Distribution of spectral features with respect to inlet channel	~	~

✓	Consistent
✗	Inconsistent
~	Indeterminate

Table 1 | Orbital observations of Jezero margin fractured unit and interpreted favorability for either a fluvio-lacustrine or not fluvio-lacustrine (groundwater) origin for carbonates there. The evidence from orbit is inconclusive regarding the processes that formed carbonates in Jezero crater. Either hypothesis may be correct, or an intermediate hypothesis—where both carbonate formation mechanisms occurred during distinct time periods—may be correct. Observations by the Perseverance Mars 2020 rover will constrain carbonate formation processes in the Jezero crater margin fractured unit.

1197

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