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1 **An insight into ancient aeolian processes and post-Noachian aqueous alteration in**
2 **Gale crater, Mars, using ChemCam geochemical data from the Greenheugh capping**
3 **unit.**

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27 **Key Points:**

- 28 • ChemCam geochemical results support that the Greenheugh pediment capping unit was
29 deposited within the Stimson sand sea in the Hesperian.
- 30 • The Greenheugh pediment capping unit was sourced from olivine-rich sands, potentially
31 recycled from units farther up Mt Sharp.
- 32 • Greenheugh experienced less interaction with groundwaters than Stimson at the Emerson
33 and Naukluft plateaus, but shared diagenesis at the unconformity.
- 34

35 **Abstract**

36 Aeolian processes have shaped and contributed to the geological record in Gale crater,
37 Mars, long after the fluvio-lacustrine system existed ~3 Ga ago. Understanding these aeolian
38 deposits, particularly those which have been lithified and show evidence for aqueous alteration,
39 can help to constrain the environment at their time of deposition and the role of liquid water later
40 in Mars' history. The NASA Curiosity rover investigated a prominent outcrop of aeolian
41 sandstone within the Stimson formation at the Greenheugh pediment as part of its investigation
42 of the Glen Torridon area. In this study, we use geochemical data from ChemCam to constrain
43 the effects of aeolian sedimentary processes, sediment provenance, and diagenesis of the
44 sandstone at the Greenheugh pediment, comparing the Greenheugh data to the results from
45 previous Stimson localities situated 2.5 km north and >200 m lower in elevation. Our results,
46 supported by mineralogical data from CheMin, show that the Stimson formation at the
47 Greenheugh pediment was likely sourced from an olivine-rich unit that may be present farther up
48 the slopes of Gale crater's central mound. Our results also suggest that the Greenheugh pediment
49 Stimson formation was cemented by surface water runoff such as that which may have formed
50 Gediz Vallis. The lack of alteration features in the Stimson formation at the Greenheugh
51 pediment relative to those of the Emerson and Naukluft plateaus suggests that groundwater was
52 not as available at this locality compared to the others. However, all sites share diagenesis at the
53 unconformity.

54 **Plain language summary**

55 We use geochemical data from the Curiosity rover's ChemCam instrument to investigate the
56 geological history of the Greenheugh pediment capping unit. The Greenheugh pediment is a
57 geologic feature that has a gently sloping top, forming a broad plain extending from the base of
58 the valley, Gediz Vallis. The Greenheugh pediment is capped by a unit of broadly uniform
59 thickness which represents the remains of the Stimson dune field that existed <2.5 Ga (mid- to
60 late-Hesperian). ChemCam geochemical data shows that the sands deposited at the Greenheugh
61 capping unit were sourced from a nearby olivine-rich unit. Surface waters then cemented the
62 windblown sand deposits, ponding at the unconformity with the underlying mudstone unit,
63 creating concretions towards the base. Episodes of groundwater circulation did not affect the
64 rocks at Greenheugh as much as they did at other Stimson localities with the exception of acid-
65 sulfate alteration that occurred along the unconformity. These results suggest that the ancient
66 Stimson dune field was a dynamic environment, incorporating grains from the surrounding
67 geological units on Mt Sharp. Furthermore, liquid water was stable at the surface in the
68 Hesperian and was available for multiple diagenetic events along bedrock weaknesses.

69 **1 Introduction**

70 Gale crater, where the NASA Mars Science Laboratory (MSL) Curiosity rover has been
71 exploring since August, 2012, contains a diverse geological record which has been shaped by
72 fluvio-lacustrine and later aeolian processes (Banham et al., 2021; Bridges et al., 2017; Edgar et
73 al., 2020; Grotzinger et al., 2012, 2014, 2015; Hobbs et al., 2010; Stack et al., 2019; Thomson et
74 al., 2011). After landing, the Curiosity rover traversed across the Bradbury plains to the foothills
75 of Gale crater's central mound, Aeolis Mons (informally known as Mt Sharp). Along the
76 traverse, the MSL mission has investigated poorly sorted conglomerates, cross-bedded
77 sandstones, and thinly laminated mudstones within the Bradbury group (Fig. 1 and 2), indicative

78 of deposition in a fluviolacustrine sedimentary system (Anderson et al., 2015; Grotzinger et al.,
79 2014; Mangold et al., 2016; Treiman et al., 2016a; Vasavada et al., 2014). Since reaching Mt
80 Sharp, the rover has investigated over 300 m of stratigraphy within the Mt Sharp group,
81 consisting mainly of laminated mudstone deposits of the Murray and Carolyn Shoemaker
82 formations, which are interpreted as evidence for a long-lived, perennial lacustrine environment
83 (Bennett et al., *this issue*: Edgar et al., 2020; Fedo et al., 2018; Grotzinger et al., 2015; Stack et
84 al., 2019). This long-lived river-lake system within Gale crater likely existed soon after the crater
85 formed approximately 3.6–3.8 Ga based on crater counting (Thomson et al., 2011) with aqueous
86 activity in the crater evidenced beyond the analyses conducted by the MSL mission by the
87 detection from orbit of hydrated minerals (Milliken et al., 2010a; Thomson et al., 2011).
88 However, at some point in Gale crater’s geological record, the environment changed to the cold
89 and dry conditions of today, in which geologic processes are dominated by aeolian activity.

90 The geological units preserved in Mt Sharp are hypothesized to have previously filled in
91 the crater, with aeolian erosion largely responsible for the exhumation of Mt Sharp to its current
92 topographic expression (Bennett et al., 2016; Grotzinger et al., 2015; Malin et al., 2000; Steele et
93 al., 2018). Aeolian processes have also resulted in the formation of modern aeolian landforms
94 including wind-ripples, sand shadows (Blake et al., 2013), transverse aeolian ridges (Zimbelman
95 et al., 2020), and dune fields (Bridges et al., 2017, 2018; Ewing et al., 2017; Lapotre et al., 2018)
96 using the available sediment eroded from the surrounding landscape (Day et al., 2016; Meslin et
97 al., 2013), or transported into the crater from elsewhere (Hobbs et al., 2010).

98 The Curiosity rover has encountered lithified aeolian sandstone deposits unconformably
99 overlying the Murray formation mudstones as part of the Stimson formation (Fig. 1 and 2) at the
100 Emerson plateau (Banham et al., 2018), Naukluft plateau (Bedford et al., 2020a), and the Murray
101 Buttes (Banham et al., 2021). Most recently, the Stimson formation has been encountered again
102 at the Greenheugh pediment, this time overlying the Carolyn Shoemaker formation mudstone
103 deposits (Fig. 1 and 2). The Stimson formation is currently the youngest in situ geological unit
104 investigated by the Curiosity rover in Gale crater. In this study, we use geochemical data from
105 the Chemistry and Camera (ChemCam) instrument suite and mineralogical data from the
106 Chemistry and Mineralogy (CheMin) instrument to investigate the geological history of the
107 Stimson formation at the Greenheugh pediment. Thereby providing constraints on the
108 paleoenvironment of Gale crater beyond the time in which the river-lake system existed.

109 1.1 An overview of the Stimson formation and previous localities

110 Initially identified from orbital imagery on the basis of its high thermal inertia with
111 respect to the surrounding units (Fraeman et al., 2016), the Stimson formation is a dark grey,
112 cross-bedded sandstone within the Siccar Point group (Fig. 2) that unconformably overlies the
113 Mt Sharp group (Banham et al., 2018; Watkins et al., 2016). The unconformity between the Mt
114 Sharp group and the Stimson formation has been informally named the basal Siccar Point group
115 unconformity and is a noticeable divide between the dark grey aeolian sandstones above, and the
116 light-toned mudstones of the Mt Sharp group below (Banham et al., 2018; Watkins et al., 2016).
117 Mineral veins in the Mt Sharp group mudstones are observed to terminate at this unconformity
118 (Fig. 3) suggesting that they formed prior to the erosion of the Mt Sharp group and deposition
119 and lithification of the Stimson formation (Kronyak, et al., 2019a).

120 Based on orbital mapping, the Stimson formation is exposed over a region of 20 km²
121 (Kronyak et al., 2019b). Previous ground-based observations of the Stimson formation showed

122 that it contains well-sorted to sub-well sorted, rounded sand grains with an average grain size of
123 medium sand with a coarse-grained skew (Banham et al., 2018). Meter-scale cross-bedding is
124 also common across all Stimson formation outcrops so far investigated by the Curiosity rover
125 (Banham et al., 2018, 2021, *this issue.*; Bedford et al., 2020a). The grain size characteristics are
126 indicative of the physical attrition of grains during transportation by saltation and surface creep
127 within migrating dune bedforms in a dry, aeolian environment (Banham et al., 2018). As such,
128 the Stimson formation is a record of a large aeolian dune field that covered the north-west flank
129 of Mt Sharp (Banham et al., 2018, 2021; Bedford et al., 2020a). Further evidence for this
130 interpretation is: the presence of wind ripple strata and meter-scale cross-bedding associated with
131 sediment transport on dune surfaces; and bounding surfaces associated with migration of
132 superimposed dunes.

133 The Emerson plateau was the first locality in which the Stimson formation was
134 investigated (Fig. 1). The Stimson formation at the Emerson plateau overlies the Murray
135 formation mudstones of the Mt Sharp group surrounding the Pahrump Hills locality (sols 750–
136 940) but was first investigated close-up by the rover at the Marias Pass locality (sols 992–1154).
137 The Stimson formation at the Emerson plateau consists of ~7 m tall outcrops of blocky, dark
138 grey sandstones, predominantly comprising simple cross sets that are usually 0.3–1.0 m thick and
139 are themselves derived of cross-laminations with a uniform thickness of 4 mm (c.f. Banham et
140 al., 2018 for a detailed description of this locality) (Fig. 3C). Bounding surfaces at the Emerson
141 plateau were traced laterally to a distance of ~30 m and are hypothesized to relate to the
142 interdune surfaces (Banham et al., 2018). Three drilled samples were acquired at the Emerson
143 plateau (Fig. 1): the Buckskin drilled sample (sol 1053) of the underlying Murray formation at
144 Marias Pass, the Big Sky drilled sample (sol 1119) representing parent Stimson, and the
145 Greenhorn drilled sample (sol 1137) representing altered Stimson at a fracture-associated halo.

146 The Naukluft plateau is situated west of the Emerson plateau and was the second Stimson
147 locality to be investigated by the Curiosity rover after the first Bagnold dune field campaign (Fig.
148 1). The Naukluft plateau is 250 km wide, with the Stimson formation at this locality varying in
149 thickness between 0.5–10 m (Bedford et al., 2020a). The Stimson formation here is characterized
150 by trough-cross bedded sandstones with a similar sedimentary architecture to the Stimson
151 formation at the Emerson plateau (Bedford et al., 2020a) (Fig. 3D). Three drilled samples were
152 also acquired at the Naukluft plateau locality: the Okoruso sample representing Stimson bedrock
153 (sol 1332, Fig. 3d), the Lubango sample of another light-toned fracture-associated halo in the
154 Stimson bedrock (sol 1320), and the Oudam drilled sample taken in the Murray formation just
155 below the Siccar Point group unconformity (sol 1361). Cross-set dip directions of the Stimson
156 formation at the Emerson plateau and Murray Buttes (Fig. 1) suggest sediment transport toward
157 the northeast (Banham et al., 2018, 2021), which is supported by geochemical trends relating to
158 the physical grain segregation of different basaltic materials that also extend this transport
159 direction to the Stimson formation at the Naukluft plateau (Bedford et al., 2020a).

160 The Murray Buttes locality was the third locality in which the Stimson formation was
161 investigated (Fig. 1). The Murray Buttes were encountered to the southwest of the Naukluft
162 plateau between sols 1412 and 1482 and form a set of isolated buttes and mesas between 5 and
163 16 m high (Banham et al., 2021). The talus and steep slopes present at the buttes prevented
164 contact science opportunities. As such, the Stimson formation here was only analyzed using
165 Mastcam images, with no opportunities for in situ geochemical or mineralogical analysis
166 (Banham et al., 2021). The Stimson formation at this locality is present as erosion resistant,

167 cross-bedded cap rock on the buttes and provides the thickest, three-dimensional exposure of the
 168 aeolian sandstone deposit of all locations to date (Banham et al., 2021). Once again, the
 169 sedimentary architecture at the Murray Buttes is similar to that of the Emerson and Naukluft
 170 plateaus, containing meter-scale planar and trough cross-bedding, with the cross-beds consisting
 171 of millimeter-thick pinstripe laminations that are laterally continuous for tens of centimeters
 172 (Banham et al., 2021). The results from the image analysis supports that the Stimson formation
 173 represents a large aeolian sand sea, composed of aggrading compound dunes that, at this
 174 location, migrated approximately to the north, downslope of Mt Sharp, which is a slight
 175 deviation from the northeast-southwest direction of the Emerson and Naukluft plateaus (Banham
 176 et al., 2021). Alteration halos were not observed at the Murray Buttes locality, but calcium
 177 sulfate cement and veins and nodules were present in the Murray formation mudstone and
 178 Stimson formation sandstone.

179 1.1.1. The Greenheugh pediment capping unit mini-campaign

180 The Greenheugh pediment was investigated by the Curiosity rover during the Glen
 181 Torridon campaign (Bennett et al., *this issue*; Bryk et al., 2019). The Glen Torridon locality
 182 comprises lacustrine mudstone of the Carolyn Shoemaker formation within the Mt Sharp group
 183 that has a strong phyllosilicate-signature from orbit (Bennett et al., *this issue*), and a diverse
 184 secondary mineralogy containing phyllosilicates, carbonates, and iron oxides that may be
 185 indicative of lacustrine groundwater mixing (Thorpe et al., *this issue*). The Stimson formation of
 186 the Greenheugh capping unit overlies the Mt Sharp group in the Glen Torridon area (Banham et
 187 al., *this issue*; Bryk et al., 2019). On the approach to the Greenheugh pediment capping unit in
 188 Glen Torridon, the underlying Carolyn Shoemaker formation mudstone changed in
 189 geochemistry, became visually lighter-toned in color (Dehouck et al., *this issue*), and increased
 190 in the abundance of F/MgO-rich and FeO_T/MnO-rich alteration features (Gasda et al., *this issue*;
 191 Forni et al., 2021), indicative of alteration of the mudstone just below the Basal Siccar Point
 192 group unconformity. The mudstone situated close to the Basal Siccar Point group unconformity
 193 at the Hutton drill hole (Fig. 2) shows a change in mineralogy with a lower abundance of clay
 194 minerals relative to the other drilled samples in Glen Torridon, in addition to minor cristobalite
 195 and opal-CT (Thorpe et al., *this issue*; Rampe et al., 2020).

196 On sol 2695, the Curiosity rover ascended the Greenheugh pediment and reached the
 197 pediment capping unit. The Stimson formation of the Greenheugh pediment capping unit can be
 198 divided into three stratigraphic intervals. The Gleann Beag interval is the lowermost interval with
 199 a thickness of ~3.6 m (Fig. 3B). The Gleann Beag interval is composed of grey, compound cross-
 200 bedded sandstones with concretions. Here, cross-bedding dip azimuths are oriented to the
 201 northeast (Banham et al., *this issue*). In total, 13 ChemCam targets (113 observation points) were
 202 measured at the Gleann Beag interval analyzing concretionary and non-concretionary bedrock.
 203 The concretions of this interval range in size between 5–20 mm diameter and preserve the
 204 sedimentary features of the sandstone (Banham et al., *this issue*). Two additional targets were
 205 measured at the base of the Gleann Beag interval that also sampled light-toned Stimson bedrock
 206 immediately above the unconformity with irregularly shaped, white clasts ~1 mm long (Banham
 207 et al., *this issue*). These clasts were interpreted as rip-up clasts derived from the Carolyn
 208 Shoemaker formation due to their similar visual appearance to the underlying mudstone and
 209 similar shape to rip-up clasts identified at the Marias Pass locality (Banham et al., *this issue*;
 210 Newsom et al., 2016).

211 Above the Gleann Beag interval is the Ladder interval (Fig. 3A and 3B). This interval
 212 consists of simple and concordant cross-sets with cross-laminations that dip to the south.
 213 Concretions are rare in this interval (Banham et al., *this issue*). Five ChemCam targets sampled
 214 the Ladder interval, although only three analyzing the uppermost Machir Bay outcrop (25
 215 observation points) are of a sufficient data quality to be included in the study.

216 Finally, the Edinburgh interval is the uppermost stratigraphic interval that was
 217 investigated by the Curiosity rover (Fig. 3A). This interval contains trough cross-bedded
 218 sandstone with cross-set dip directions to the west (Banham et al., *this issue*). In this interval,
 219 concretions were very rare, and one drilled sample (Edinburgh, sol 2715, Fig. 3a) was delivered
 220 to the CheMin and SAM instruments (Sutter et al., 2022; Rampe et al., 2020). The Edinburgh
 221 drilled sample contained a high abundance of plagioclase feldspar (28.9 wt%), pyroxene (20.2
 222 wt%), olivine (8.4 wt%) and magnetite (10.2 wt%), in addition to K-feldspar (3.4 wt%),
 223 phyllosilicates (7 wt%), trace fluorapatite and an X-ray amorphous component (20 wt%) (Thorpe
 224 et al., *this issue*; Rampe et al., 2020). ChemCam analyzed 30 targets in this interval (259
 225 observation points) in addition to 1 target of a sedimentary float rock (Cheviot Hills, sol 2696)
 226 and 1 soil (Digg, sol 2710).

227 *1.2 A geochemical and mineralogical analysis of Gale crater's ancient dunes*

228 *1.2.1. Constraining aeolian net sediment transport direction and provenance*

229 Previous studies of aeolian dunes derived from basaltic material on Earth and Mars have
 230 shown that major element chemistry can provide information on mineral sorting processes that
 231 can in turn constrain net sediment transport direction and changes in the sediment source
 232 (Baratoux et al., 2011; Bedford, et al., 2020a; Cornwall et al., 2015; Sara, 2017; Mangold et al.,
 233 2011; Mountney et al., 2004; O'Connell-Cooper et al., 2017; Rampe et al., 2018). Basaltic sand
 234 grains can be monomineralic, composed of olivine, pyroxene, and feldspar grains, derived from
 235 volcanic or impact glass, or polymineralic, composed of a mixture of basaltic minerals and glass
 236 (Cornwall et al., 2015; Fedo et al., 2015). The minerals within each grain determine the density,
 237 shape, and resistance to physical and chemical weathering that in turn determine how they will
 238 be sorted hydrodynamically with transportation in an aeolian environment (Cornwall et al., 2015;
 239 Fedo et al., 2015; Jerolmack et al., 2006; Morrone et al., 2017; Willetts et al., 1982).

240 On Earth, most mature aeolian sands are dominated by quartz as this mineral is the most
 241 resistant to physical and chemical weathering (Muhs, 2004; Pettijohn, 1954). Mafic minerals in
 242 sedimentary environments on the Earth are uncommon because Earth's continental crust largely
 243 consists of felsic materials and reactive basaltic materials are easily chemically weathered under
 244 Earth's active hydrological cycle (Nesbitt et al., 1992). However, on Mars, the crust is
 245 predominantly basaltic, and the cold and arid climate of the Amazonian geological period has
 246 resulted in physical weathering being the main driver in the diminution of aeolian sediments.
 247 Physical abrasion studies by Cornwall et al (2015) show that olivine grains are rapidly abraded to
 248 a spherical shape that aides its transportation in aeolian environments. Meanwhile the cleavage
 249 planes of feldspar and pyroxene act as zones of weakness in monomineralic sand grains and
 250 hinder the sphericity of these grains. Glass and polymineralic basaltic grains show the most
 251 resilience to physical abrasion. These properties result in olivine minerals, glass, and basaltic

252 fragments being transported farther and concentrating in mature aeolian sands, relative to
253 feldspar grains. Mineral sorting studies of basaltic aeolian sands in Iceland show that an increase
254 in $\text{MgO} + \text{FeO}_T$ with distance from the source corresponds to an increase in olivine relative to
255 feldspar regardless of the presence of basaltic grains and glass (Baratoux et al., 2011; Sara, 2017;
256 Mangold et al., 2011). As the mineral sorting acts on feldspar minerals and olivine, it is possible
257 to use the variation in $\text{MgO} + \text{FeO}_T$ relative to $\text{Al}_2\text{O}_3 + \text{Na}_2\text{O} + \text{K}_2\text{O}$ to measure the variation in
258 the mafic and felsic basaltic materials, respectively (Baratoux et al., 2011; Mangold et al., 2011).

259 In Gale crater, several studies have used the geochemical data from the ChemCam
260 instrument to probe the geochemical variation in aeolian sediments in relation to mineralogical
261 variations (Bedford et al., 2020a; Cousin et al., 2017; Meslin et al., 2013). ChemCam
262 geochemical data of modern aeolian sediments from Gale crater show that locally derived sands
263 consist of coarse feldspar grains in addition to finer mafic material (Cousin et al., 2015; Meslin et
264 al., 2013), whereas, the mature aeolian sands of the Bagnold dune field only contain feldspar
265 within polymineralic grains, or as fine monomineralic grains (Cousin et al., 2017; Rampe et al.,
266 2018). A statistical geochemical analysis of ChemCam data from the Emerson and Naukluft
267 plateaus was applied to determine trends associated with mineral sorting during transportation
268 prior to deposition and lithification (Bedford et al., 2020a). The results showed that the Emerson
269 plateau had a higher abundance of MgO and FeO_T relative to the Naukluft plateau, which was
270 more felsic in chemistry and had a greater proportion of coarser feldspar grains identified in
271 ChemCam Remote Micro-Imager (RMI) images (Bedford et al., 2020a). The geochemical
272 variation of the Stimson formation at the Emerson and Naukluft plateaus indicated that net
273 sediment transport direction was from southwest-to-northeast (Bedford et al., 2020a), supporting
274 the results of Banham et al. (2018). This net sediment transport direction is opposite to that of the
275 active Bagnold dunes, and when the geochemistry of the ancient dunes and modern dunes are
276 directly compared to one another, they show geochemical differences indicative of different
277 basaltic provenances (Bedford et al., 2020a). The geochemistry of the Stimson formation
278 sandstone was more similar to that of the subalkaline basaltic sediments that are a source for the
279 Bradbury group fluvial sandstones, suggesting that the two geological units either shared a
280 similar sediment source, or that the Stimson sandstones may consist of recycled Bradbury group
281 material (Bedford et al., 2020a). Due to the variation in cross-set dip directions between intervals
282 in the Stimson formation at the Greenheugh capping unit (cf Banham et al., *this issue*), we will
283 compare the geochemistry of each interval at the Greenheugh pediment to that of the Stimson
284 formation at the Emerson and Naukluft plateaus using the methods of Bedford et al. (2020a) to
285 determine if the geochemical variation supports changes in sediment transport direction and
286 potentially sediment provenance.

287 1.2.2. Constraining aqueous alteration of the Stimson sandstone

288 The alteration features of the Stimson sandstone have been of particular interest in
289 several studies as they can provide information on the nature of the groundwater that existed
290 after the habitable river-lake system ended, and after the erosional moat formed within Gale
291 crater. Although the timing of Stimson deposition and lithification on the slopes of Mt Sharp is
292 not well constrained, it is likely that the Stimson sandstones formed in the mid- to late-Hesperian
293 (Banham et al., 2021), making the alteration features within the Stimson formation representative

294 of some of the youngest aqueous alteration events observed in situ, at the time of this
295 publication. Previous research suggests that the Stimson formation at the Emerson and Naukluft
296 plateaus was potentially lithified by a rising capillary fringe that did not interact with the
297 depositional surface (Banham et al., 2018). This interaction between groundwater and mafic
298 minerals in the aeolian sediments (particularly olivine) caused the formation of an iron oxide and
299 X-ray amorphous materials as cement, isochemical to the previous sandstone deposit, and
300 preferentially cemented as concretions closer to the unconformity (Bedford et al., 2020a;
301 Hausrath et al., 2018).

302 After lithification of the Stimson sandstone, repeated episodes of groundwater
303 emplacement and desiccation formed the polygonal fractures and calcium sulfate hydrofracture
304 veins in the Stimson sandstone (Frydenvang et al., 2017; Hausrath et al., 2018; Kronyak et al.,
305 2019a, 2019b; Yen et al., 2017, 2021). Multiple alteration features occur within the Stimson
306 formation of the Emerson and Naukluft plateaus such as concretions, mineral veins and the
307 alteration halos which were sampled at Greenhorn and Lubango (Fig. 4). Concretions are more
308 common towards the unconformity, similar to what is observed at the Greenheugh pediment
309 (Banham et al., 2018, *this issue*; Bedford et al., 2020a) (Fig. 4). An analysis of concretionary
310 versus non-concretionary Stimson bedrock at the Emerson and Naukluft plateaus by the
311 ChemCam instrument showed that there is no statistically significant geochemical variation of
312 these features to the surrounding bedrock (Bedford et al., 2020a). This supports the hypothesis
313 that the concretions at these localities likely formed through in situ isochemical alteration and
314 represent parts of the bedrock that were preferentially cemented during the lithification of the
315 sandstone (Banham et al., 2018; Bedford et al., 2020a).

316 Light-toned, silica-rich Stimson bedrock surrounding large fractures, known as silica-rich
317 alteration halos, are present in the Stimson formation at the Emerson and Naukluft plateaus
318 (Bedford, et al., 2020a; Frydenvang et al., 2017; Yen et al., 2017). Silica-rich halos are
319 geochemically characterized by an increase in SiO₂ abundances up to 80 wt% and a decrease in
320 all other major element oxides except TiO₂ and K₂O whose compositions are scattered across
321 observation points (Bedford et al., 2020a; Frydenvang et al., 2017; Yen et al., 2017). Two
322 hypotheses exist for the formation of the halos. Either, the halo alteration occurred through the
323 remobilization of silica from the underlying mudstones by neutral to alkaline pH fluids into the
324 fractures in the Stimson bedrock where they precipitated to form the halos (Frydenvang et al.,
325 2017). Alternatively, it has been hypothesized that they formed as a result of acidic fluids
326 circulating through the fractures, interacting with the mafic minerals in the surrounding bedrock,
327 remobilizing their cations and forming a silica-rich residue (Hausrath et al., 2018; Yen et al.,
328 2017). The timing of the silica-enrichment that formed the light-toned halos relative to the
329 formation of the calcium sulfate veins and cement is currently unknown, however, previous
330 studies agree that these represent some of the latest stages of diagenesis in the Stimson formation
331 and underlying Murray formation at these localities (Frydenvang et al., 2017; Yen et al., 2017).

332 Polygonal fractures with average widths of 7.5 m can also be identified by orbital images
333 and from the surface in the Stimson formation at the Emerson plateau, Naukluft plateau, and to a
334 lesser extent at the Greenheugh pediment (Kronyak et al., 2019b). These polygonal fractures are
335 similar in appearance to the surrounding bedrock, only evident as they are slightly raised due to
336 their greater erosion resistance (Kronyak et al., 2019b). Based on their geomorphology, the
337 polygons likely formed as a result of multiple cycles of expansion and contraction of the bedrock
338 caused by the recharge of near-surface groundwater preferentially cementing these zones

339 (Kronyak et al., 2019b). The polygons are not likely related to the diagenesis that formed the
 340 silica-rich halos or the calcium sulfate mineral veins as these features do not consistently
 341 correlate spatially across the Siccar Point group (Kronyak et al., 2019b).

342 The small sampling size of the ChemCam laser in addition to the thorough
 343 documentation of each target by RMI images before and after sample acquisition makes it
 344 possible to isolate observation points with alteration features and compare them to the bedrock,
 345 constraining alteration processes. Here, we compare alteration features and examine variations in
 346 potential sandstone cementing agents between the different Stimson formation locations in order
 347 to further constrain the spatial scale of these late-stage aqueous alteration events in Gale crater.

348

349 **2 Materials and Methods**

350 *2.1 The ChemCam instrument*

351 The ChemCam instrument suite utilizes Laser-Induced Breakdown Spectroscopy (LIBS)
 352 to obtain elemental analyses of a geological target situated over ~2 m from the rover mast, with
 353 most of the targets selected at a distance less than 5 m from the rover mast (Maurice et al., 2012,
 354 2016; Wiens et al., 2012). LIBS spectra are pre-processed to remove ambient light background,
 355 continuum background, and noise (Wiens et al., 2013). The quantification of elemental
 356 abundances is obtained for major elements (Si, Ti, Al, Fe, Mg, Ca, Na, K) through a combination
 357 of partial least squares regression sub-models and independent component analysis using a large
 358 spectral training set (Anderson et al., 2017; Clegg et al., 2017; Forni et al., 2013). Minor and
 359 trace elements are quantified individually, often using the same spectral library. Relevant to this
 360 work, MnO quantification is described in Gasda et al. (2021). Hydrogen quantification has been
 361 shown to require special attention to the matrix (Rapin et al., 2017a; Rapin et al., 2017b;
 362 Schröder et al., 2015; Thomas et al., 2018); nevertheless it has proven useful in some specific
 363 cases (e.g., Rapin et al., 2016), and rough quantification can be used for large-scale studies
 364 (Thomas et al., 2020). The ChemCam team discovered that while F atomic emission lines are not
 365 generally detectable, F enrichments can be observed and quantified via the CaF molecular
 366 emission band, which has indicated the presence of apatite, fluorite, and mica grains (Forni et al.,
 367 2015).

368 The uncertainty of major elemental abundances based on ChemCam data has been
 369 presented as accuracy and precision to 1σ (Anderson et al., 2017; Clegg et al., 2017; Wiens et al.,
 370 2012) and is provided in the Supplementary information (Table S.1). Accuracy relates to the root
 371 mean square error of prediction (RMSEP) for representative geological samples that share
 372 abundance ranges similar to those in the calibration regression models (Clegg et al., 2017). The
 373 precision of the predictions is calculated as the variation observed across the 25 shots that derive
 374 the average spectrum for each observation point (Blaney et al., 2014), or across targets from a
 375 uniform unit (Mangold et al., 2016).

376 ChemCam targets are taken in a linear or grid raster configuration i.e., 3×3 or 1×10
 377 (Maurice et al., 2016). Each point of the raster is known as an observation point, where each
 378 observation point consists of between 30–50 laser shots. Spectra from the first 5 laser shots are
 379 not included in the average spectrum and serve to minimize surface dust contamination (Clegg et
 380 al., 2017). The ChemCam instrument suite also contains a Remote Micro-Imager (RMI) which

381 takes high-resolution (60–100 μ rad) images of targets pre- and post-acquisition of the LIBS
 382 spectral data in order to provide geological context for the targets in question (Le Mouélic et al.,
 383 2015).

384 *2.2 The CheMin instrument suite*

385 CheMin housed within the rover body and generates X-ray diffraction (XRD) patterns of drilled
 386 rock or scooped sediment samples (Blake et al., 2012; Morrison et al., 2018a). Rietveld
 387 refinement and full-pattern fitting techniques are used to quantify abundances of minerals and X-
 388 ray amorphous material and to determine unit-cell parameters of the major crystalline phases
 389 (Blake et al., 2012, Morrison et al., 2018a). Mass-balance calculations using mineral abundances
 390 and crystal chemistry from CheMin and geochemical data from APXS provide estimates of the
 391 composition of the amorphous component (e.g., Smith et al., 2018). Evolved gas analyses from
 392 the Sample Analysis at Mars (SAM) instrument suite help identify clay minerals and amorphous
 393 materials (e.g., Sutter et al., 2017). In this study, we use the CheMin-derived mineral abundances
 394 of the Stimson formation sandstone at the Emerson plateau (Big Sky, sol 1119; Morrison et al.,
 395 2018; Yen et al., 2017), Naukluft plateau (Okoruso, sol 1332; Morrison et al., 2018b; Yen et al.,
 396 2017), and Greenheugh pediment capping unit (Edinburgh, sol 2711; Rampe et al., 2020) to
 397 place the geochemical compositions from the ChemCam instrument into mineralogical context.

398 *2.3 The on-board science cameras and target classifications*

399 In this study, we have individually classified each ChemCam observation point using the
 400 RMI images according to whether the observation point has analyzed bedrock, secondary
 401 diagenetic features, unconsolidated sediments, or float. Where available, images from the
 402 Curiosity rover's other on-board science cameras, Mastcam (Mast camera) and MAHLI (Mars
 403 Hand Lens Imager), were used to support the classification at the scale of the outcrop and target,
 404 respectively. Mastcam includes two, fixed-focal length, multispectral, color CCD imagers
 405 situated on Curiosity's mast (Malin et al., 2017). The two Mastcam imagers are separated by
 406 24.2 cm and positioned approximately 2 m from the surface (Malin et al., 2017). The left
 407 Mastcam (M-34) has a focal length of 34 mm, a field of view (FOV) of $20^\circ \times 15^\circ$, and a pixel
 408 scale of 0.22 mrad/pixel (Malin et al., 2017). Meanwhile, the right Mastcam (M-100) has a 100
 409 mm focal length, a FOV of $6.8^\circ \times 5.1^\circ$, and a 0.074 mrad/pixel scale of sampling (Malin et al.,
 410 2017). Data from this instrument were used to identify large-scale features, such as cross-
 411 bedding or alteration halos, and to locate the ChemCam targets with respect to the stratigraphic
 412 units. MAHLI is situated on the turret at the end of the Curiosity rover's arm and uses a 2-mega-
 413 pixel color CCD camera to analyze the textures, structures, and morphologies present at the
 414 finest grain size possible for Curiosity's cameras, 0.0042 mm (silt) (Edgett et al., 2012; Mangold
 415 et al., 2017). Data from this instrument were used to provide additional, high resolution, color
 416 data to support the classifications of the black and white images from the ChemCam RMI.

417 Bedrock is defined here as an indurated, lithified unit, possibly containing sedimentary
 418 structures such as bedding or laminations, and secondary structural features such as bedrock
 419 fractures. As the sedimentary units of the Bradbury group, Mt Sharp group, and Siccar Point
 420 group (Stimson formation) have undergone compaction and cementation during diagenesis, it is
 421 important to note that these analyses are not completely free of alteration as they include
 422 diagenetic cement. However, provided that the diagenesis which formed the sandstone cement
 423 was relatively isochemical, the geochemistry can be used to identify mineral sorting trends

424 before the units became lithified, in addition to sediment source rock characteristics. Concretions
425 are abundant in the Stimson formation which likely formed through the preferential cementation
426 of the Stimson sands (Banham et al., 2018; Bedford et al., 2020a; Potter-McIntyre et al., 2014;
427 Potter et al., 2011). A previous analysis of concretions in the Stimson formation sandstone by
428 both the Alpha Particle X-ray Spectrometer (APXS; e.g., Gellert & Clark, 2015) and ChemCam
429 instruments showed that they have the same geochemical composition to non-concretionary
430 Stimson (Bedford et al., 2020a; Siebach et al., 2017) and are hypothesized to relate to closed-
431 system diagenesis that preserves the bulk rock geochemistry (Banham et al., 2018; Bedford et al.,
432 2020a; Siebach et al., 2017). This is also true for the Greenheugh pediment capping unit for
433 concretions smaller than 5 mm (Appendix A); as such they are included in the bedrock dataset.
434 However, larger concretions that are greater than 5 mm (Fig. 4), with multiple ChemCam
435 observation points directly on the concretions, and which occur closer to the unconformity have
436 been isolated from the bedrock dataset of all Stimson localities for a more in-depth analysis on
437 the diagenesis that formed these features. As the Stimson formation across all localities has a
438 relatively uniform grain size (Banham et al., 2018; 2021; *this issue*), we have not classified
439 bedrock targets according to grain size. Bedrock targets do not include analyses of drill tailings
440 and dump piles as these samples have been modified by the rover. Furthermore, bedrock
441 analyses have been restricted to only include observation point data with oxides that sum to
442 between 90–105 wt% to minimize the influence of volatile elements (S, Cl, and H), or elements
443 such as Ni which are not nominally quantified in ChemCam data. Removing the potential impact
444 of volatile elements is important for our bedrock dataset as these elements may have been
445 emplaced due to open-system alteration. Restricting the total sum of oxides can further remove
446 any targets that have been contaminated with Martian soil, as these deposits also have higher
447 concentrations of volatiles (Bish et al., 2013; Ehlmann et al., 2017; Lasue et al., 2018; Meslin et
448 al., 2013; O’Connell-Cooper et al., 2017).

449 ChemCam analyses that have targeted secondary diagenetic features in the Stimson
450 sandstone have been placed into an alteration feature dataset and subclassified according to
451 “vein”, and “halo” categories. Vein alteration targets relate to light-toned mineral veins that in-
452 fill hydrofractures in the Stimson formation (Bedford et al., 2020a; Kronyak et al., 2019a). Halos
453 relate to the light-toned halos that extend into the Stimson bedrock ~20 cm either side of large
454 fractures (Fig. 4) (Bedford et al., 2020a; Frydenvang et al., 2017; Yen et al., 2017). These
455 secondary diagenetic features have been separated from the bulk rock dataset to isolate the
456 effects of later diagenetic alteration from geochemical trends that may relate to the sediment
457 source characteristics of the Stimson formation across all localities, in addition to pre-
458 lithification mineral sorting processes, and the formation of the sandstone cement.

459 Other classifications include float and soils. Float targets in Gale crater include pebble to
460 boulder grain size fragments of rock that can be igneous, sedimentary, or meteoritic in nature.
461 Soil targets are classified as unlithified deposits, with a lack of obvious cement that derive fine-
462 grained accumulations of sand- to silt-grained particles (Bish et al., 2013; Cousin et al., 2015;
463 Meslin et al., 2013). The Curiosity rover also encountered two large, active, aeolian dune fields,
464 and investigated them in two phases of the Bagnold dune campaigns (Bridges et al., 2018;
465 Lapotre et al., 2018). The aeolian dunes occur as large bedforms, ~15 m across, which are dark
466 in color and can be considered a modern analog of the ancient Stimson formation (Achilles et al.,
467 2017; Bedford et al., 2020a; Hausrath et al., 2018). Due to the similarities in characteristics of

468 the dune and soil deposits, ChemCam analyses of these deposits have been placed into the soil
469 category.

470 *2.4 Statistical methods and data interpretation*

471 The ChemCam LIBS laser sampling footprint is 350–750 μm for distances 3–7 m from
472 the rover mast (Maurice et al., 2012), which is a similar size to the average grain size of the
473 Stimson formation (336–634 μm at the Greenheugh pediment, Banham et al., *this issue*). As
474 such, target analyses are not likely to be representative of whole rock compositions as individual
475 grains are more likely to be sampled (Cousin et al., 2017). However, the bedrock Stimson
476 formation ChemCam dataset to date (August, 2021) consists of 741 data points which permits
477 the use of a statistical approach to both estimate a bulk rock composition and detect geochemical
478 trends that may relate to mineralogical variations in the relatively alteration-free bedrock dataset.
479 For each locality and stratigraphic interval of the Stimson formation, we give the basic statistics
480 (mean, median, range, and standard deviations), followed by density contour plots which will be
481 used to estimate the bulk rock composition. Density contours display the data across two
482 variables and represent the density of smoothed data within each pixel on the bivariate plot
483 defined according to the bin size (Bedford et al., 2019a; Eilers et al., 2004). Data densities are
484 smoothed according to Eilers and Goeman (2004), and contours are generated using MATLAB
485 (2019a). This method is used to estimate the bulk rock composition which is in turn used as a
486 comparison to the geochemical variation associated with the alteration feature datasets. By
487 comparing the estimated whole rock composition to alteration features, we can constrain how
488 these features formed.

489 After the density contour analysis, sandstone components (the minerals and amorphous
490 materials that comprise the sand and cement) are delineated using a hierarchical, agglomerative
491 cluster algorithm on the bedrock dataset with Minitab 19 statistical software (2020), following
492 the methods of Bedford et al. (2020a) and Bedford (2019a). In this study, we use the complete
493 linkage measure of association for Euclidean distances with all major oxides, now including
494 MnO, and the total sum of oxides, as the variables. Complete linkages and Euclidean distances
495 were used as these methods provided the best fit for the dataset showing the most defined
496 clusters and was successfully used in a previous investigation of the Stimson formation (Bedford
497 et al., 2020a). Clustering methods have also been successfully employed in other studies of Gale
498 crater sediments (Rammelkamp et al., 2021; Gasnault et al., 2019, 2013). Variables were
499 standardized in order to minimize the effect of scale differences.

500 **3 Results**

501 *3.1. Distribution of the classified data across the Stimson formation*

502 In total, 430 ChemCam observation points were acquired during the Greenheugh
503 pediment mini campaign that were analyzed <4 m from the rover and were of good data quality.
504 This included 324 bedrock analyses, two analyses of calcium sulfate veins, 10 soil analyses, 10
505 float analyses, 20 analyses that targeted large (>5 mm) nodules, and 15 analyses of the
506 Edinburgh drill hole wall and tailings (Fig. 5). This is in contrast to the distribution of
507 observation point classifications for the Emerson and Naukluft plateaus which has a much larger
508 proportion of observation points (50% at Emerson plateau, 65% at Naukluft plateaus) that
509 targeted alteration features such as halos, concretions, and calcium sulfate veins (Fig. 5). These

510 proportions are not a result of the true distribution of these features relative to bedrock as
511 targeting bias to investigate certain features has played a role. Regardless, the diversity of
512 alteration at the Greenheugh capping unit is significantly less than the diversity of the features
513 seen previously, with no alteration halos were identified at the Greenheugh pediment capping
514 unit, unlike at the Emerson and Naukluft plateaus (Fig. 5) or any obvious raised ridge features in
515 comparison to the 15 raised ridge analyses at the Emerson plateau spanning 2 targets (Tinder
516 Box and Duperow), and the 7 raised ridge analyses of the Uubvley and Bloedkoppie targets at
517 the Naukluft plateau.

518 Calcium sulfate mineral veins are rare at the Greenheugh pediment capping unit with
519 only two observation points targeting a very thin mineral vein (Fig. 4) and rare occurrences of
520 veins identified in the Mastcam images. Calcium sulfate veins were also rare at the Emerson and
521 Naukluft plateaus with 15 and 27 ChemCam targets of veins respectively, but where they did
522 occur the veins were larger in size and extent than the vein investigated at Greenheugh (Fig. 4).
523 Concretions are also present across all sites. All the large and small concretions at the
524 Greenheugh locality were observed in the lowermost Gleann Beag interval in the ChemCam data
525 and Mastcam images (Fig. 3B and 4). Consequently, there is a similar number of large
526 concretion analyses in the Stimson formation at the Greenheugh pediment (20 points over 4
527 targets) and the Naukluft plateau (29 points over 6 targets). Only three observation points in the
528 targets Dakota and Heath sampled large concretions in the Stimson formation at the Emerson
529 plateau.

530 Of the 324 bedrock analyses, 220 constituted data within the 90–105 wt% total sum of
531 oxide range. This suggests that 104 observation points on the Greenheugh capping unit contains
532 a higher abundance of volatile elements (S, H, or Cl) or other elements which are not routinely
533 quantified, 82% of which are within the Edinburgh interval. Due to a distinct geochemistry in
534 two ChemCam targets situated just above the Siccar Point group unconformity, 20 observation
535 points were placed into their own dataset for an analysis of the unconformity relative to the bulk
536 Greenheugh pediment capping unit.

537 *3.2. Geochemistry of the Greenheugh pediment capping unit compared to the Stimson* 538 *formation at the Emerson and Naukluft plateaus*

539 The geochemistry of the Greenheugh pediment capping unit is similar to that of the
540 Stimson formation analyzed previously, with geochemical distributions, mean compositions, and
541 standard deviations within family to both previous Stimson formation localities for TiO_2 , FeO_T ,
542 and K_2O (Fig. 6 and Table 1). For all localities, TiO_2 , FeO_T , and K_2O have unimodal
543 distributions, with mean compositions and standard deviations of 0.9 ± 0.2 wt%, 19.5 ± 2.8 wt%,
544 and 0.6 ± 0.4 wt%, respectively (Fig. 6). Both distributions for TiO_2 and K_2O have a heavy right
545 tail, which is the most prominent for the Emerson plateau, though these heavy right tails do not
546 significantly skew the median compositions from the mean. Two-way equivalence tests also
547 support that these major element oxides are equivalent across the three Stimson localities within
548 the upper and lower limits defined by ChemCam precision (Supplementary material) and they
549 suggest that Na_2O can be claimed as equivalent across the three sites, despite the slight variation
550 in distribution (Table S.2). For all three sites, Na_2O has a mean composition and standard
551 deviation of 3.0 ± 0.8 wt%, with multimodal distributions presenting peaks at 1.8 wt%, 2.7 wt%,
552 3.1 wt%, and a heavy tail to the right. However, the distribution in data between these peaks
553 differ across all sites (Fig. 6). The dominant peak for the Emerson plateau and Greenheugh

554 capping unit is at 2.7 wt%, with both of these localities showing fewer data extending to higher
 555 Na₂O concentrations. In comparison, the Naukluft plateau's dominant peak is at 3.1 wt%, with a
 556 greater proportion of data points situated over 4 wt% Na₂O.

557

558

559 The mean compositions of SiO₂, Al₂O₃, MgO, CaO, and MnO across the Stimson
 560 formation localities are all situated within 1 standard deviation of each other (Table 1), however,
 561 variations in the distribution of the data suggests that there may be statistically significant
 562 differences, supported by equivalence test results (Table S.2). The Emerson plateau and
 563 Greenheugh pediment capping unit both share unimodal SiO₂ distributions with peaks at 45 wt%
 564 (Fig. 6). However, the distribution of SiO₂ at the Emerson plateau shows a right skew to higher
 565 SiO₂ concentrations up to 57.1 wt%, whereas the Greenheugh pediment capping unit shows a
 566 skew to the left with a greater proportion of data points <44 wt% SiO₂ causing the non-
 567 equivalence between the Emerson plateau and Greenheugh pediment capping unit. The Naukluft
 568 plateau has a distinct bimodal distribution for SiO₂ sharing the peak at 45 wt% with an
 569 additional, larger peak at 48 wt%. This second larger peak for the Naukluft plateau, in addition to
 570 values up to 61.7 wt% SiO₂, results in a lack of equivalence for SiO₂ between the Naukluft
 571 plateau and other Stimson localities. The high SiO₂ observation points of the Emerson and
 572 Naukluft plateaus were cross-checked with the locations of the silica-rich alteration halos and the
 573 RMI images, and either correlate with large, light-toned mineral grains (Fig. 3d), or they were
 574 randomly interspersed within the target raster which is not consistent with the 10s of cms-scale
 575 alteration halo features (Bedford et al., 2020a; Frydenvang et al., 2017). Instead, the correlation
 576 of these high SiO₂ values with high Na₂O and/or K₂O, and low MgO and FeO_T suggest that they
 577 may relate to ChemCam analyses that have targeted a greater proportion of felsic grains (Bedford
 578 et al., 2020a). Thus, the statistically significant differences may be indicative of different
 579 proportions of minerals comprising the sand (e.g., olivine, basaltic glass, and feldspar) or
 580 differences in cement composition at each locality.

581 The Greenheugh capping unit shows statistically significant geochemical variation from
 582 the Emerson and Naukluft plateaus for Al₂O₃, and CaO, which is on average lower at the
 583 Greenheugh capping unit, and MnO and MgO which is higher in mean abundance (Fig. 6). Al₂O₃
 584 compositions show a bimodal distribution for the Emerson and Naukluft plateaus, with the
 585 histograms showing peak abundances at 10 wt% and 17 wt% for the Emerson plateau, and peaks
 586 at 12 wt% and 17 wt% at the Naukluft plateau. Meanwhile, the Greenheugh pediment capping
 587 unit has a multimodal distribution with peaks at 8 wt%, 10 wt%, and 16 wt%. The additional
 588 peak of the Greenheugh capping unit at 8 wt% has resulted in lower mean (10.3 ± 0.2 wt%) and
 589 median (9.6 wt%) compositions compared to the rest of the Stimson formation localities with
 590 average compositions of 11.3 ± 0.2 wt% and 12.1 ± 0.2 wt% for the Emerson and Naukluft
 591 plateaus, respectively.

592 Both the Emerson and Naukluft plateaus have a unimodal distribution for MgO, with a
 593 peak at ~7.5 wt%, while the Greenheugh pediment capping unit has a bimodal distribution with
 594 peaks at 7 wt% and 9 wt%. All localities have a right tail to MgO concentrations of >17 wt%,
 595 generating a slight right skew in the datasets (Fig. 6).

596 The distribution in CaO is also the same between the Emerson and Naukluft plateaus in
 597 that both are unimodal with a dominant peak at 6.4 wt%. Meanwhile, the Greenheugh pediment

598 capping unit has a bimodal distribution with the 6.4 wt% peak shared with the other localities, in
599 addition to a different, more dominant peak at 4.0 wt%.

600 MnO has a unimodal distribution for all localities with a strong peak at 0.2 wt%,
601 however, outliers exist to high concentrations at the Emerson plateau (up to 2.6 wt%) and
602 Greenheugh capping unit (4.0 wt% in target Aegis_post_2730). The outlying MnO values at the
603 Greenheugh pediment capping unit are more numerous than those of the Emerson plateau, and
604 all occur within the Edinburgh interval bedrock targets, generating a mean concentration that is
605 ~1.6x that of the Emerson and Naukluft plateaus.

606 To summarize, the Stimson formation at the Greenheugh pediment capping unit is
607 situated within the compositional range of earlier Stimson units, according to the standard
608 deviation of the dataset. However, statistically significant variations in the geochemical
609 distribution of the dataset exist, as is evident in the mean, equivalence test results, and the
610 histograms of major element oxides. The Greenheugh pediment capping unit has a higher
611 abundance of MgO and MnO, and lower abundances of SiO₂, Al₂O₃, and CaO relative to the
612 Stimson formation at the Emerson and Naukluft plateaus that may relate to different abundances
613 of sandstone components such as mafic minerals, felsic minerals, or sandstone cement.

614 *3.3 Geochemical variation associated with diagenetic features and the unconformity at the* 615 *Greenheugh capping unit.*

616 Calcium sulfate veins across the Stimson formation show a decrease in all major oxide
617 compositions and total sum of oxides, except for CaO that can reach values up to 50 wt%
618 (Bedford et al., 2020a). The two calcium sulfate veins targeted on the Greenheugh pediment
619 capping unit were both situated in the Edinburgh interval and as a result of both veins being very
620 thin (<1 mm wide), the analyses of these veins only show a slight increase in CaO compared to
621 the average of the Edinburgh interval. Calcium sulfate can also be a cement component. For
622 Stimson analyses across all localities with total sum of oxides less than 90 wt%, 23 analyses out
623 of 191 share a similar geochemical trend to the calcium sulfate mineral veins, suggesting that
624 calcium sulfate cement is present in these ChemCam observation points. For the Greenheugh
625 pediment capping unit, 11 observation points that targeted bedrock have geochemical signatures
626 of calcium sulfate cement, suggesting that it is a small component of the sandstone and is
627 variable in its distribution.

628 Large concretions (>2 cm) in the Stimson formation preserve sedimentological features
629 such as laminations and therefore likely represent parts of the sandstone that have been
630 preferentially cemented (Banham et al., 2018). To conduct a closer investigation of how the
631 sandstone cement of the Stimson formation varies across the Greenheugh capping unit, we have
632 isolated the compositions of these large concretions and plotted them alongside the geochemical
633 abundances of the non-concretionary bedrock and small concretionary bedrock (S.5). Similar to
634 the results of Bedford et al. (2020) for the Emerson and Naukluft plateaus, no statistically
635 significant geochemical variation is present across concretions (Fig. S.5).

636 To investigate how the geochemistry of the Greenheugh pediment capping unit varies
637 with respect to the basal Siccar Point Group unconformity, and in turn how it may vary for the
638 immediately underlying Carolyn Shoemaker formation, we have classed the bedrock data in
639 relation to the stratigraphic levels just above (Stimson Unconf.), and just below (CS Unconf.) the
640 unconformity (Fig. 7 and 8). In addition to the targets closest to the unconformity, we also

641 compare the results of analyses acquired at the base of the pediment around the Hutton drill hole
 642 (CS Hutton) and the analyses acquired farther above the unconformity in the Stimson formation
 643 stratigraphic intervals (Glean Beag interval, Ladder interval, and Edinburgh interval). As the
 644 previous literature has suggested that the unconformity may have been a conduit for diagenetic
 645 fluids to travel and alter the underlying Carolyn Shoemaker formation (Achilles et al., 2020;
 646 Fraeman et al., 2020; Horgan et al., 2020; Rampe et al., 2020), we have not restricted the
 647 bedrock totals for this analysis, to allow for geochemical variation associated with secondary
 648 phases that formed through water-rock interactions to be more apparent. The mean compositions
 649 and their associated standard errors of the Stimson formation and Carolyn Shoemaker formation
 650 are also shown for comparison (Fig. 7). Figure 7 shows that the Carolyn Shoemaker formation
 651 leading up to the unconformity is depleted in SiO₂ and H but enriched in CaO relative to the
 652 mean composition of the Carolyn Shoemaker formation. The Stimson formation just above the
 653 unconformity (Stimson Unconf.) has a unique geochemistry that is low in MgO, Na₂O, and
 654 MnO, low in total sum of oxides, but high in CaO and H relative to the mean composition of the
 655 Stimson formation. These geochemical characteristics of the base of the Stimson formation
 656 indicate that it may have either been affected by diagenetic fluids, been derived from a different
 657 sediment provenance, or represents a complex mixture of both processes.

658 *3.4 The distribution of halogens across the Stimson formation.*

659 For the bulk rock dataset, all Stimson formation localities have a unimodal distribution of
 660 H peak area about 0.004, however, the Stimson formation at the Greenheugh pediment has a
 661 bimodal distribution with a second modal peak of H peak abundances at a value of 0.008 (Fig.
 662 6). This contrasts significantly with the right-skewed unimodal distributions of the Emerson and
 663 Naukluft plateaus. Looking at the distribution of the H data in relation to the stratigraphic
 664 intervals at the Greenheugh pediment (Fig. 8A), these peak area values are divided between the
 665 Gleann Beag interval that contains concretions and is situated close to the unconformity, and the
 666 Ladder and Edinburgh intervals that do not contain notable concretions and are higher in the
 667 stratigraphy (Fig. 8). The Gleann Beag interval shares the modal H peak area with that of the
 668 Stimson formation at the Emerson and Naukluft plateaus, whereas the Ladder and Edinburgh
 669 intervals exclusively have the higher H peak area. ChemCam analyses of the non-concretionary
 670 bedrock have the highest H peak area (0.0052) compared to small concretions (0.0046) and large
 671 concretions (0.0039), however the change in surface roughness between these classifications
 672 likely impacts the H signal (Rapin et al., 2017a; Schröder et al., 2015). As such, we do not
 673 consider the H peak area to differ significantly across the concretions. H peak area is also
 674 variable among the other diagenetic features, with the calcium sulfate veins having the highest H
 675 abundances (mean of 0.01) of all features and bedrock analyzed by ChemCam. Silica-rich halos
 676 also had a slightly higher mean H abundance (0.007) than that of the average Stimson, unaltered
 677 bedrock (0.005). Only MnO has a possible positive correlation with H abundance for targets on
 678 the Edinburgh interval with MnO > 1 wt% (r^2 49%, $p = 0.011$).

679 F detections across the Stimson formation are rare. In total, there are 20 bedrock
 680 observation points (16 non-concretionary bedrock, four small concretion-rich bedrock), and two
 681 halo analyses across all Stimson localities that have F above the limit of detection. Only one
 682 observation point in the Edinburgh interval of the Greenheugh pediment capping unit has F,
 683 contrasting with the high F targets in the underlying Carolyn Shoemaker formation of the Hutton
 684 zone (Dehouck et al., *this issue*; Gasda et al., *this issue*). For the bedrock targets, F has a
 685 potential positive correlation with CaO (r^2 48%, $p = 0.001$; S.3) and shares no correlations with

686 any other major element oxide. F has the largest range in abundances for targets close to the
 687 unconformity (0.7 wt% F), compared to those situated over 1.5 m above (0.2 wt% range in F;
 688 Fig. S.4).

689 3.5 Stimson bedrock cluster analysis results

690 The results of the cluster analysis show that the Stimson formation bedrock dataset can be
 691 divided into 5 clusters (Fig. 9). Cluster 1 is defined as the cluster with the highest mean MgO
 692 content (14.7 ± 1.1 wt%), higher than average MnO (0.7 ± 0.2 wt%) and the lowest Al₂O₃ ($6.9 \pm$
 693 0.4 wt%), CaO (3.6 ± 0.4 wt%), Na₂O (2.3 ± 0.1 wt%), and K₂O (0.3 ± 0.1 wt%) of all clusters
 694 (Table 2). Nineteen data points have a Cluster 1 membership from across the Stimson formation
 695 with eight at the Greenheugh pediment (Fig. 9, 10). Cluster 1 is most similar to Cluster 2 and is
 696 differentiated by a similarity level of 36.3 (see Bedford (2019) for the methods to calculate
 697 similarity level). Cluster 2 has a moderately high abundance of MgO (8.6 ± 0.1 wt%) with the
 698 greatest variation in TiO₂ (standard deviation of 0.21 wt%) and the greatest maximum TiO₂ (2.18
 699 wt%) of all clusters. Cluster 2 differs from Cluster 1 by a slightly greater mean abundance of
 700 Al₂O₃ (9.7 ± 0.1 wt%) and CaO (5.6 ± 0.1 wt%), and a lower abundance of MgO (Table 2). In
 701 total, 372 data points have a Cluster 2 membership across all localities. It is the largest cluster,
 702 comparable in size to Cluster 3 and is the most representative of the Greenheugh pediment
 703 average composition as it is situated at the focal composition for this locality (Fig. 10, 12).

704 The next closest cluster is Cluster 3 with a similarity to Clusters 1 and 2 of 28.3. Cluster 3
 705 is differentiated by Clusters 1 and 2 by having on average higher abundances of SiO₂ (48.5 ± 0.2
 706 wt%), Al₂O₃ (13.1 ± 0.1 wt%), CaO (6.0 ± 1.8 wt%), and lower abundances of FeO_T (18.7 ± 0.1
 707 wt%) and MgO (6.5 ± 0.1 wt%). Cluster 3 has a total of 330 observation points with the majority
 708 situated on the Naukluft plateau (Fig. 10). Clusters 2 and 3 are the largest clusters in the dataset
 709 and as such are situated within the density contours of the Stimson bulk rock (Fig. 12). Similar to
 710 the results of Bedford et al. (2020a) for the main clusters 2 and 3, the standard deviations are
 711 higher than for the other clusters, which could indicate that some of the data points are not well
 712 bound to these clusters. However, the bimodal distribution of SiO₂ and Na₂O of the Naukluft
 713 plateau (Fig. 6), which contains the majority of Cluster 3 memberships, shows peak abundances
 714 of 45 wt% and 48 wt% SiO₂, and 2.7 wt% and 3.4 wt% Na₂O that align with the Clusters 2 and 3
 715 averages, respectively. The variation of Al₂O₃ between the Emerson plateau and Greenheugh
 716 pediment localities that share a peak of 10 wt% Al₂O₃ also differs from the main peak
 717 composition for Al₂O₃ of the more Cluster 3-rich Naukluft plateau (13 wt%), suggesting a more
 718 bimodal distribution between all Stimson localities for this major element oxide. As such, this
 719 indicates that Clusters 2 and 3 are an inherent part of the dataset and accurately represent the
 720 most common sandstone components across all localities, despite the overlap of some points.
 721 Furthermore, as Clusters 2 and 3 derive from a total of 702 points, the overlap of <10 % of
 722 datapoints does not significantly skew the analysis of these two clusters.

723 The two most distinctive clusters are Clusters 4 and 5 (Fig. 9). Cluster 4 consists of 15
 724 data points, distributed relatively equally between localities (four at Emerson, five at Naukluft,
 725 and six at Greenheugh, Fig. 10). Cluster 4 is defined as having the highest abundances of SiO₂
 726 (54.50 ± 0.84 wt%), Al₂O₃ (17.97 ± 0.61 wt%), Na₂O (5.74 ± 0.34 wt%), and higher than
 727 average K₂O for the Stimson formation (0.97 ± 0.16 wt%). Cluster 5 only has five data points,
 728 one at the Emerson plateau and 4 in the Edinburgh interval of the Greenheugh capping unit and
 729 is defined by its high MnO concentration (2.92 ± 0.19 wt%) and high K₂O concentration similar

730 to Cluster 4 (1.0 ± 0.3 wt%). Aside from the MnO and K₂O concentration of Cluster 5, it is very
 731 similar in composition to the MgO, FeO_T, and TiO₂-rich Clusters 1 and 2.

732 **4 Discussion**

733 *4.1 Geochemical variation associated with mineral sorting or sediment source*

734 The sedimentary architecture of the Stimson formation is interpreted as resembling dry
 735 aeolian sand dunes (Banham et al., 2018, 2021, *this issue*). In a dry aeolian environment, the
 736 coarsest and densest grains are rolled across the surface as they are transported by surface creep,
 737 while the majority of sand-sized particles are temporarily lifted across the surface and
 738 redeposited elsewhere as they travel via saltation. Meanwhile, grains finer than sand are
 739 suspended in the wind. As such, mud to silt-sized grains and platy minerals such as
 740 phyllosilicates are not likely to be deposited in a dry aeolian bedform (Kocurek et al., 2012),
 741 supporting that the phyllosilicates at the Greenheugh capping unit are likely to be authigenic.
 742 Sediments that are transported and deposited in dry aeolian environments are also subjected to
 743 physical abrasion and less chemical weathering (Thomas et al., 2005). As such, sediment grains
 744 that are less dense with a shape that is efficient at transportation by saltation or surface creep, and
 745 that are resistant to physical weathering are more likely to be transported farther in a dry aeolian
 746 environment and concentrated in distal deposits relative to those which do not meet these
 747 criteria.

748 The geochemical trends of the majority of ChemCam data between mafic and felsic
 749 minerals, and the similarity in composition between concretionary and non-concretionary
 750 bedrock, also suggests that alteration was likely minimal away from the unconformity for the
 751 Greenheugh pediment capping unit. Minimal alteration at Greenheugh is further supported by the
 752 presence of 8 wt% olivine in the Edinburgh drilled sample (Rampe et al., 2020), which is only
 753 slightly less than the olivine abundances for the active aeolian sands at the Bagnold Dunes (e.g.,
 754 9.6 wt% at Ogunquit Beach) (Rampe et al., 2018). This, in addition to the low degree of
 755 chemical weathering during transportation for dry aeolian systems (Jiang et al., 2019; Zhu et al.,
 756 2009), suggests that the geochemistry of the Stimson formation at the Greenheugh pediment
 757 capping unit is likely to be representative of the mineralogy of the dune sands at the time of
 758 deposition.

759 *4.1.1. The relationship of the CheMin mineralogy to the ChemCam derived clusters*

760 The crystal chemistries of the Edinburgh sample are not yet available, so we will
 761 compare our geochemical results to the crystal chemistries of the Big Sky drilled sample which
 762 is considered the least affected by alteration of the Emerson and Naukluft bedrock samples (Yen
 763 et al., 2017). As olivine is no longer present at the Emerson and Naukluft plateaus due to
 764 diagenesis (Bedford et al., 2020a; Hausrath et al., 2018; Siebach et al., 2017), we use the olivine
 765 crystal chemistry of the Gobabeb sample from the Bagnold dunes. (Achilles et al., 2017;
 766 Morrison et al., 2018b). The primary igneous minerals present in the non-halo related drilled
 767 samples Big Sky and Okoruso drilled at the Emerson and Naukluft plateaus respectively are
 768 plagioclase feldspar (36.5 ± 1.8 wt% Big Sky, 27.2 ± 1.7 wt% Okoruso), K-feldspar (1.1 ± 0.6
 769 wt% Big Sky, 1.9 ± 0.7 wt% Okoruso), and pyroxene (25.4 ± 2.3 wt% Big Sky, 20.7 ± 1.1 wt%
 770 Okoruso) (Yen et al., 2017). The primary igneous minerals of the Edinburgh drilled sample from
 771 the Greenheugh pediment capping unit also contains plagioclase feldspar (28.9 wt%), K-feldspar

772 (3.4 wt%), and pyroxene (20.2 wt%), in addition to olivine (8.4 wt%) (Rampe et al., 2020). On a
 773 plot of $\text{Al}_2\text{O}_3 + \text{Na}_2\text{O} + \text{K}_2\text{O}$ versus $\text{MgO} + \text{FeO}_T$, all five clusters are situated along a trendline
 774 that extends from Stimson feldspar mineral endmembers (plagioclase feldspar and K-feldspar) in
 775 the top left of the plot, to mafic mineral endmembers (pyroxene and olivine) situated in the
 776 bottom right (Fig. 12A). Clusters 1 and 2, in addition to Cluster 5 are situated closer to the mafic
 777 mineral endmember of the plot. The Greenheugh pediment average composition is most similar
 778 to Cluster 2 (Fig. 11A). The geochemical composition of Cluster 2 also relates strongly to the
 779 lower SiO_2 , Al_2O_3 , Na_2O , and K_2O , and higher MgO dominant peak compositions of the
 780 Emerson plateau and Greenheugh capping unit compositions, and the lesser of the bimodal peak
 781 compositions for the Naukluft plateau (Fig. 6). This geochemical relationship of Cluster 2 with
 782 one of the bimodal peak compositions and greater chemical affinity to mafic minerals suggests a
 783 high proportion of mafic minerals and their diagenetic products being targeted by the ChemCam
 784 laser. Cluster 2 is most prevalent in the Greenheugh capping unit (55% total datapoints) followed
 785 by the Emerson plateau (46.2% total datapoints), with the fewest data at the Naukluft plateau
 786 (27.5% total data points; Fig. 10), supporting the greater influence of this cluster on the
 787 geochemical distribution present in the histograms.

788 Cluster 1 plots closer to the mafic mineral endmembers than Cluster 2 and likely
 789 represents a mixture of mafic minerals (Fig. 11A). Few observation points ($n = 17$) have Cluster
 790 1 memberships suggesting that it is rare for the mafic end members to be targeted on their own,
 791 though it is present at each Stimson locality in relatively equal proportions (6 points at Emerson,
 792 5 points at Naukluft, and 6 points at Greenheugh). An image analysis of Cluster 1 and 2
 793 memberships (Fig. 13) show that Cluster 1 and 2 points likely relate to ChemCam targets that
 794 have sampled dark-toned and fine-grained parts of the sandstone. As such, the fine grain size and
 795 association of mafic minerals with the formation of the sandstone cement may have decreased
 796 the likelihood of a mafic endmember being targeted with the ChemCam laser, instead preferring
 797 the more mixed composition of Cluster 2.

798 The five ChemCam observation points with a Cluster 5 membership also show a
 799 geochemical composition which is closer towards the mafic mineral endmembers (Fig. 12A), but
 800 with high abundances of MnO (Fig. 11B). MnO can be present as a trace element in mafic
 801 minerals such as olivine and pyroxene, though not at the high abundances present in Cluster 5,
 802 suggesting that this cluster may relate to the local remobilization of Mn and Fe from mafic
 803 minerals that contain trace amounts of MnO in their crystal structure. As such, we interpret
 804 Clusters 1 and 2 as being indicative of the relative abundance of mafic minerals initially present
 805 within the deposited aeolian sands, a proportion of which was later altered to sandstone cement
 806 during early diagenesis, creating Cluster 5.

807 Clusters 3 and 4 are, in turn, situated closer to the feldspar mineralogical endmember,
 808 with Cluster 3 more representative of the geochemistries of the Emerson and Naukluft plateaus
 809 (Fig. 12A). Our Clusters 3 and 4 are geochemically similar to Clusters 3 and 5 of Bedford et al.
 810 (2020a) that were interpreted to represent more felsic minerals relative to mafic minerals and
 811 cement targeted by the ChemCam laser. The ChemCam RMIs show that the LIBS spots with
 812 Cluster 3 and 4 memberships have preferentially targeted light-toned mineral grains in the
 813 sandstone, further supporting a felsic nature for these clusters (Fig. 12B). Cluster 3 more strongly
 814 influences the bulk composition of the Emerson and Naukluft Stimson formation, skewing the
 815 focal composition away from Cluster 2 and causing a greater number of contour intervals to
 816 extend around the Cluster 3 composition (Fig. 11A). Looking at how the Cluster memberships

817 vary with location (Fig. 10), Cluster 3 is most abundant at the Naukluft plateau (n = 173),
 818 followed by the Emerson plateau (n = 101), with the lowest abundance of Cluster 3 present at the
 819 Greenheugh pediment capping unit (n = 56). The average composition of Cluster 3 also
 820 correlates to the peak composition of the Stimson formation that is situated at higher SiO₂,
 821 Al₂O₃, and Na₂O, and lower MgO and FeO_T, particularly at the Emerson plateau where Cluster 3
 822 is most abundant (Fig. 6). As such, we interpret Clusters 3 and 4 as being representative of the
 823 proportion of felsic minerals that were present in the initially deposited Stimson aeolian sand
 824 dunes.

825 *4.1.2. Implications for sediment sorting and provenance of the Stimson dune sands*

826 Basaltic sediments transported in aeolian environments on Earth in localities such as the
 827 Dyngjúsundur sand sheet, Iceland, are largely derived of volcanic glass, olivine, pyroxene,
 828 feldspar, and lithic grains (Baratoux et al., 2011; Cornwall et al., 2015; Mangold et al., 2011).
 829 These basaltic components are also present in the active aeolian sands on Mars, including those
 830 of the active Bagnold dune fields in Gale crater (Achilles et al., 2017; Lapotre et al., 2018;
 831 Rampe et al., 2018). Of these basaltic components, feldspar and augite are the least likely to be
 832 transported far as their cleavage planes and more tabular/elongate shapes make them both
 833 vulnerable to physical abrasion and less efficient at transportation by saltation and surface creep
 834 in aeolian environments (Baratoux et al., 2011; Cornwall et al., 2015; Mangold et al., 2011;
 835 Mountney et al., 2004). Olivine has a higher density (from forsterite at 3.29 g/cm³ to fayalite at
 836 4.39 g/cm³) than most basaltic materials which could decrease the efficacy in which it can be
 837 transported far in aeolian settings, particularly at low wind speeds (Fedo et al., 2015). However,
 838 its similar Mohs hardness to quartz and conchoidal fractures promoting rapid rounding of the
 839 olivine grains can make it more likely to persist in the aeolian transport of basaltic sands relative
 840 to feldspar and pyroxene, provided the sands are heterogeneous (Cornwall et al., 2015). The most
 841 resilient basaltic components in aeolian sediments are volcanic glass and polycrystalline basalt
 842 grains. Hence, if pyroxene and feldspar are present in distal aeolian sands they are likely
 843 entrained within polycrystalline basaltic grains (Cornwall et al., 2015). Geochemical trends
 844 relating to an increase in MgO downwind in the Dyngjúsundur sand sheet, Iceland, supports an
 845 increase in olivine minerals (Baratoux et al., 2011; Sara, 2017; Mangold et al., 2011).

846 The previous cluster analysis by Bedford et al. (2020a) hypothesized that physical grain
 847 segregation was the main cause of geochemical variation between the Emerson and Naukluft
 848 plateaus. In addition to the geochemical variation that related to mineralogical variation, a more
 849 local sand source for the Naukluft plateau was further supported by the presence of coarse
 850 feldspar grains which are less abundant at the Emerson plateau (Bedford et al., 2020a; Fig. 3d),
 851 and the geochemical results supported the interpretation of a southwest-northeast net sediment
 852 transport direction determined through cross-bed dip directions (Banham et al., 2018). The
 853 Greenheugh pediment capping unit has more MgO relative to the other Stimson localities (Fig. 6,
 854 11), in addition to a greater abundance of Clusters 2 and 5 (Fig. 11), indicative of higher
 855 abundances of mafic minerals. This is supported by the mineralogical results from CheMin that
 856 show a relatively high concentration of olivine (Rampe et al., 2020), indicating that the
 857 Greenheugh capping unit may be enriched in mafic components as a result of mineral sorting
 858 during transportation assuming that the sands were derived from the same sediment source. If
 859 this is the case, the Greenheugh capping unit would have been deposited farther from the source
 860 than the Stimson formation at the Emerson and Naukluft plateaus that respectively show an
 861 increase in the felsic Clusters 3 and 4 (Fig. 10). Cross-set dip directions of the Stimson formation

862 at the Gleann Beag interval of the Greenheugh pediment shares the northeast dip direction of the
863 cross-sets examined at the Emerson and Naukluft plateaus. However, the cross-set dip directions
864 of the olivine-rich Edinburgh interval are indicative of an east-west net sediment transport
865 direction, different to the southwest-northeast transport direction of the earlier Stimson localities
866 and Gleann Beag interval (Banham et al., *this issue*). Cross-set dip directions in the Ladder
867 interval show dip directions to the south, which indicates a sediment transport direction to the
868 south. These variable dip directions show that the wind regime driving transportation at the time
869 of deposition of the Greenheugh pediment was also variable (Banham et al., *this issue*), making
870 the possibility of detecting geochemical variation relating to physical grain segregation with
871 transportation more complex.

872 It is possible that sediment had accumulated down slope of the present-day pediment and
873 was then remobilized after the sediment transport ratio changed, resulting in the reversal of the
874 dune at the Ladder interval and the change in net sediment transport direction for the Edinburgh
875 interval. This would maintain that the sediments at the pediment were transported farther from
876 their source than those at the Emerson and Naukluft plateaus. Despite the variations in net
877 sediment transport directions across the Greenheugh pediment intervals, the ChemCam data do
878 not show a statistically significant difference in geochemistry for the major element oxides in
879 these different stratigraphic intervals of the Greenheugh pediment capping unit away from the
880 unconformity (Fig. 7). However, the grain size of the Stimson sandstone at the Greenheugh
881 pediment capping unit is approximately the same as that of the Emerson plateau (Banham et al.,
882 *this issue*) which is not anticipated for a more distal deposit. Furthermore, the presence of
883 pyroxene and feldspar within the Stimson sandstone of the Greenheugh capping unit suggests a
884 relatively immature aeolian sandstone (Cornwall et al., 2015), supporting that the higher
885 abundance of olivine at the Greenheugh pediment likely relates to an olivine-rich local sediment
886 source, not transportation. In the ~10 km long Dyngjusandur sand sheet of Iceland, the
887 mineralogy of the aeolian sediments show strong contributions from the source rocks of the
888 surrounding volcanoes (Baratoux et al., 2011; Sara, 2017; Mountney et al., 2004). Feldspar
889 grains are diverse in this sand sheet, representing several volcanic source rocks (Sara, 2017), and
890 the incorporation of pumice into the northeast portion of the sand sheet from the 1875 eruptives
891 of the nearby Askja volcano shows how readily aeolian processes can incorporate local bedrock
892 materials (Baratoux et al., 2011; Sara, 2017; Mountney et al., 2004). As such, it is possible that
893 the ~2.5 km distance that separates the Greenheugh pediment from the Emerson and Naukluft
894 plateaus and the different wind regimes at the Greenheugh pediment were sufficient to result in a
895 change in the main sediment source at that locality. We favor the hypothesis that a more olivine-
896 rich sediment source is responsible for the geochemical difference of the Greenheugh pediment
897 capping unit from that of the Emerson and Naukluft plateaus.

898 It is widely accepted that the present topographic expression of Mt Sharp formed as a
899 result of aeolian erosion after the deposition and lithification of the Mt Sharp group (Day et al.,
900 2016; Grotzinger et al., 2015; Steele et al., 2018). Orbital investigations of the units farther up
901 the slopes of Mt Sharp show several olivine-bearing marker beds to the south and southeast of
902 the Greenheugh capping unit (Rudolph et al., 2019). These marker beds could provide an
903 adequate source for the olivine-rich sandstones preserved in the Greenheugh pediment capping
904 unit, with the Stimson formation at the Emerson and Naukluft plateaus likely derived more from
905 a Bradbury sandstone source (Bedford et al., 2020a).

906 4.2 *The contribution of sediment recycling to the Stimson formation at the Greenheugh*
907 *pediment*

908 Sedimentological evidence for sediment recycling has been identified at several locations
909 in Gale crater (Edgett et al., 2020) including the basal Stimson formation at the Marias Pass
910 locality (Newsom et al., 2018). Several angular clasts are present in the basal Stimson formation
911 at Marias Pass <4 mm wide, and decrease in abundance with stratigraphic height in the outcrop
912 (Newsom et al., 2018). These angular clasts were interpreted as either a mixture of Murray
913 mudstone intraclasts and calcium sulfate vein fragments or Murray clasts coated in calcium
914 sulfate according to their largely CaO-rich geochemistry and light tonation (Newsom et al.,
915 2018). Away from the unconformity, the geochemistry of the Stimson sandstone shared
916 similarities to that of the basaltic sandstone from the Bradbury group, suggesting that they may
917 either have been derived from a similar regional sediment source, or have been recycled from
918 Bradbury group sandstones to the southwest of the Naukluft plateau (Bedford et al., 2020a).

919 Like the basal Stimson Missoula outcrop at Marias Pass, the basal Stimson targets of the
920 Greenheugh pediment capping unit have a relatively lighter color compared to the overlying dark
921 grey Stimson sandstone, and they contain some intraclasts (Fig. 13). However, there are some
922 important differences in the physical and chemical properties between these units. First, the
923 intraclasts are not as bright as those of the Missoula outcrop of Marias Pass, indicating that they
924 may not have been overprinted by calcium sulfates to the same extent (Fig. 13D). Second, the
925 underlying Carolyn Shoemaker formation is actually SiO₂-poor relative to average (Fig. 8)
926 indicative of silica-remobilization, not precipitation like that which formed the silica-rich halos
927 of the Emerson and Naukluft plateaus.

928 Although ChemCam did not directly analyze a mudstone intraclast at the Greenheugh
929 pediment, it is possible that smaller grains of the Carolyn Shoemaker formation may have been
930 recycled into the aeolian Stimson deposits (Fig. 13D; Banham et al., *this issue*), in addition to
931 any diagenesis that could be the cause of the lighter color of the basal Stimson targets,
932 complicating the geochemistry (Fig. 13A and 13B). To delineate the geochemical trends of
933 diagenesis from those of sediment recycling of the underlying Murray formation into the aeolian
934 deposits preserved at the Greenheugh pediment, we have generated mixing lines between the
935 Carolyn Shoemaker formation and Stimson formation. Results show that at least four
936 observation points from basal Stimson targets are situated within 1 standard deviation of the
937 trendline indicative of mixing with between 50% to 90% of Carolyn Shoemaker bedrock (Fig.
938 13A and 13B). Looking at the post-ChemCam LIBS MAHLI image, the points with a more
939 Shoemaker-like chemistry appear to have hit a greater proportion of light-toned angular grains.
940 Four basal Stimson observation points have pure Stimson chemistries, and the rest of the basal
941 Stimson observation points show geochemical signatures indicative of calcium sulfates. For the
942 observation points that largely lie along the mixing line, they do show some depletions in Na₂O
943 and K₂O relative to the trendline indicating that these points are not completely free of diagenetic
944 overprint (see supplementary material S6). H is also elevated in these unconformity targets but is
945 present at average Stimson values for the other Gleann Beag interval ChemCam targets above
946 the basal Stimson targets (Fig. 9). This indicates that Carolyn Shoemaker grains were
947 incorporated into the basal Stimson formation at the Greenheugh pediment which was later
948 subjected to diagenetic fluids that remobilized the alkali elements, and precipitated calcium
949 sulfate cement and iron oxides.

950 Mt Sharp group mudstone intraclasts preserved at the base of the Stimson formation at
 951 Marias Pass and the Greenheugh pediment supports that sediment recycling of the immediately
 952 underlying bedrock occurred. Due to grains finer than fine sand being winnowed away in dry
 953 aeolian environments and transported in suspension, it is possible that the lacustrine mudstone of
 954 the Mt Sharp group was rapidly abraded and transported away from the Stimson sand dunes
 955 which is why geochemical signatures of the Mt Sharp group are not preserved far from the
 956 unconformity. The sorting of fine-grained volcanoclastic material from basaltic sand grains in
 957 aeolian bedforms is seen in fluvio-aeolian sedimentary systems in Iceland, where fine-grained
 958 palagonite tuff is not preserved in the well-sorted aeolian ripples, but is present in the proximal
 959 fluvial deposits (Bedford et al., 2020b), supporting the hypothesis that grain size is likely a factor
 960 in the Mt Sharp group mudstone's preservation in the Stimson formation.

961 *4.3 Aqueous alteration and cementation of the Stimson formation at the Greenheugh*
 962 *pediment.*

963 4.3.1 Lithification of the Stimson formation and early diagenesis

964 The Stimson sand sea was likely a dry aeolian deposit (Banham et al., 2021), so the first
 965 measurable instance of water-sediment interaction would have occurred after deposition of the
 966 dune sands, during the interaction of the sediments with the cementing fluid which lithified the
 967 formation in what we define as the early stage diagenesis. In this section, we will discuss
 968 whether this cement-forming diagenesis occurred at the same time under the same conditions
 969 across all localities through a comparison of the mineralogy of Thorpe et al. (*this issue*) and
 970 ChemCam geochemistry.

971 The secondary minerals present in the non-halo Big Sky and Okoruso drill samples,
 972 drilled at the Emerson and Naukluft plateaus respectively, include iron oxides magnetite ($10.3 \pm$
 973 0.6 wt% in Big Sky, 11.2 ± 0.7 wt% in Okoruso) and hematite (3.0 ± 0.5 wt% in Big Sky, $0.7 \pm$
 974 0.4 wt% in Okoruso), and minor (<2 wt%) abundances of calcium sulfates, quartz, and
 975 fluorapatite (Morrison et al., 2018; Yen et al., 2017). Both the Emerson and Naukluft plateaus
 976 also contain 15-20 wt% of an X-ray amorphous component with a moderate SiO_2 and FeO_T
 977 composition for Gale crater which likely formed from the aqueous alteration of basaltic
 978 sediments (Smith et al., 2021). Secondary minerals at the Greenheugh pediment capping unit
 979 include magnetite (10 wt%), hematite (0.4 wt%) and phyllosilicates (7 ± 3 wt%), and 20 wt%
 980 amorphous component (Rampe et al., 2020). When comparing the mineralogy of these
 981 sandstones to the ChemCam data, the secondary minerals and amorphous component that derive
 982 the diagenetic cement were hypothesized in Bedford et al. (2020a) to have contributed to the
 983 FeO_T - and MgO -rich Clusters 1 and 2, and the TiO_2 -rich Cluster 4 of that study. As the MgO
 984 geochemistry of the Stimson formation at these sites is similar to that of the olivine-bearing
 985 active Bagnold dune sands and the chemistry of the bedrock comprised of small concretions is
 986 indistinguishable from non-concretionary bedrock, it was hypothesized that the cement forming
 987 diagenesis at these Stimson localities was extensive enough to convert all olivine to diagenetic
 988 cement, but was relatively isochemical, preserving the geochemical trends that related to mineral
 989 sorting in the ancient dune field and sediment source characteristics (Fig. 11; Bedford et al.,
 990 2020a). Similar to the results of Bedford et al. (2020a), our cluster analysis, which now includes
 991 the Greenheugh capping unit and MnO data, shows MgO - and FeO_T -rich Clusters 1 and 2,
 992 respectively. These clusters are compositionally equivalent to Clusters 1 and 2 of Bedford et al.
 993 (2020a) with the exception of SiO_2 that is on-average ~ 4 wt% lower and Al_2O_3 which is ~ 2 wt%

994 lower in the new clusters, likely a result of the higher abundance of mafic minerals at the
995 Stimson formation at the Greenheugh capping unit, but not at the Emerson and Naukluft
996 plateaus. This similarity in composition of the new Stimson Clusters 1 and 2 to the old ones
997 maintains that Clusters 1 and 2 are compositionally representative of the primary igneous
998 mineralogy of the pre-lithified sands that is present across all localities, even though the
999 secondary alteration minerals and amorphous component composition varies.

1000 Hydrous alteration modelling of the Stimson formation at the Emerson and Naukluft
1001 plateaus suggests that the sandstone cement which contains the secondary minerals and
1002 amorphous component primarily formed through the dissolution of olivine under slightly acidic
1003 to near neutral pH conditions (pH 6–8) during burial diagenesis (Hausrath et al., 2018). The pH
1004 was largely constrained by the presence of abundant magnetite (10.3 wt%) as a lower pH would
1005 inhibit the precipitation of authigenic magnetite (Hausrath et al., 2018). The similar abundance of
1006 magnetite to the Emerson and Naukluft plateaus further suggests that olivine at the Greenheugh
1007 capping unit also underwent dissolution at a pH > 6 to precipitate magnetite, which given that 8
1008 wt% olivine is still present at the Greenheugh pediment, indicates a much higher abundance of
1009 olivine in the originally deposited aeolian sands (Rampe et al., 2020). Furthermore, as olivine is
1010 one of the most reactive basaltic minerals, particularly in aqueous fluids with a pH < 8 (Babechuk
1011 et al., 2014; Eggleton et al., 1987; Nesbitt et al., 1992; Pokrovsky et al., 2000), the high
1012 abundance of olivine in the sandstone of the Greenheugh pediment capping suggests that this
1013 sandstone has undergone less interaction with diagenetic fluids than the Stimson formation at the
1014 Emerson and Naukluft plateaus.

1015 The presence of 7 wt% phyllosilicates at the Greenheugh pediment and none detected at
1016 the Emerson and Naukluft plateaus further suggests that the early diagenetic conditions may
1017 have been slightly different at this locality even though the clusters are compositionally
1018 equivalent. Magnetite (4.3–11.3 wt%), phyllosilicates (10–22 wt%), and olivine (1.8–4.9 wt%)
1019 have been detected in the Bradbury group basaltic fluviolacustrine deposits, the John Klein and
1020 Cumberland drilled samples of the Yellowknife Bay mudstones (Vaniman et al., 2014), and the
1021 Windjana drilled sample of the Kimberley formation (Treiman et al., 2016). Hydrous alteration
1022 modelling of the formation conditions of the phyllosilicates at Yellowknife Bay by Bridges et al.
1023 (2015) suggests that these phyllosilicates formed through an inhomogeneous dissolution of ~70
1024 wt% amorphous component, ~20 wt% olivine, and 10 wt% host rock in an open-system with a
1025 neutral to alkaline fluid at a pH 7.5–12. A comparison of the Stimson formation sandstone
1026 geochemistry to that of the Yellowknife Bay mudstone and sandstone showed that they are both
1027 basaltic and likely derived from a similar sediment provenance (Bedford et al., 2020a), as such,
1028 the modelling results of Bridges et al. (2015) are applicable to the Stimson formation. Thereby,
1029 the formation of the phyllosilicates at the Greenheugh capping unit likely occurred under more
1030 alkaline conditions (pH 7.5–12) compared to the early diagenetic conditions of the Emerson and
1031 Naukluft plateaus (pH 6–8; Hausrath et al., 2018).

1032 In the ChemCam data, H can be used to trace the abundance of hydrated phases such as
1033 phyllosilicates across sedimentary units (Thomas et al., 2020). H at the Greenheugh capping unit
1034 is notably bimodal at H peak abundances of 0.003 and 0.005 (Fig. 6, 7). The Edinburgh and
1035 Ladder intervals of the Greenheugh pediment capping derive the higher H values (0.005),
1036 supporting the presence of phyllosilicates in these stratigraphic units. Meanwhile, the lower H
1037 value at the Greenheugh capping unit consists of data solely within the Glean Beag interval and
1038 is equivalent to the peak H value at the Emerson and Naukluft plateaus (0.003), suggesting

1039 minimal phyllosilicates. This indicates that the phyllosilicates are stratigraphically confined to
1040 the Edinburgh and Ladder intervals (Fig. 8) which could result from spatial diagenetic variability
1041 in the Stimson formation at the Greenheugh capping unit relative to the unconformity. This
1042 spatial variability in phyllosilicate abundances with the unconformity is also present in the
1043 underlying Carolyn Shoemaker formation, which has less phyllosilicates in the Hutton drilled
1044 sample compared to other drill holes away from the unconformity (Thorpe et al., *this issue*).

1045 Evidence to support spatial diagenetic variability in the Stimson formation is present with
1046 the distribution of the concretions. On average, concretions are more abundant and greater in size
1047 with proximity to the unconformity for the Stimson formation at the Greenheugh pediment (Fig.
1048 5), and elsewhere (Banham et al., 2018, *this issue*). This relationship of the concretions with the
1049 unconformity suggests that their formation may relate to the presence of a perched aquifer at the
1050 base of the Stimson formation, or that it could indicate a greater abundance of minerals that acted
1051 as a nucleus for concretion formation (Banham et al., 2018; Chan et al., 2012; Potter-McIntyre et
1052 al., 2014). With the exception of the basal Stimson formation that includes mixing of mudstone
1053 intraclasts and alteration by a later diagenetic fluid, the Stimson formation is relatively
1054 geochemically uniform (Bedford et al., 2020a; Siebach et al., 2017; Thompson et al., *this issue*),
1055 with large-scale variations only relating to late-stage diagenetic features (Bedford et al., 2020a;
1056 Yen et al., 2017). As such, a greater concentration of minerals at the unconformity for which
1057 these concretions could have nucleated seems unlikely. However, the Mt Sharp group units that
1058 underly the Stimson formation are well-cemented, phyllosilicate-bearing mudstones which
1059 makes them more likely to act as an aquiclude and promote the formation of a perched aquifer
1060 which in turn promoted the formation of the concretions. If the perched aquifer is the cause of
1061 the concretion formation, this would in turn support that the diagenetic fluids responsible for
1062 cementing the Stimson formation were derived from near-surface waters that accumulated
1063 towards the unconformity. The Greenheugh pediment is fan shaped and extends from Gediz
1064 Vallis whose morphology suggests that it may have formed from fluvial processes (Anderson et
1065 al., 2010; Thomson et al., 2011; Wiens et al., 2020), providing a potential source for the surficial
1066 water that lithified the Stimson formation at the Greenheugh pediment and potentially further
1067 downslope.

1068 With the addition of MnO data to the Stimson formation and, therefore, the cluster
1069 analysis, there is now a MnO-rich Cluster 5 that consists of high-MnO observation points (mean
1070 of 2.9 wt%) from three host rock ChemCam targets from the Edinburgh interval of the
1071 Greenheugh pediment capping unit (Fig. 13B). Additional MnO-rich host rock targets in the
1072 Edinburgh interval are also present for observation points that have ChemCam totals <90 wt%
1073 that are excluded from the cluster analysis. The ChemCam observation points with high-MnO
1074 concentrations targeted fine grained, dark-toned sandstone that is not associated with any
1075 obvious feature and are sporadically distributed spatially across the sandstone. MnO
1076 concentrations are >1 wt% for these data points and there are no correlations with other major
1077 element oxides. Similar observations of sporadic enrichments in MnO were detected in the
1078 Glenelg and Gillespie Lake sandstones of the aforementioned Yellowknife Bay formation (Lanza
1079 et al., 2014); note that these Mn abundances were later recalibrated (Gasda et al., 2021). The
1080 cause of these high MnO-points were hypothesized to be either detrital, from a sediment source
1081 that experienced oxidizing aqueous conditions, or diagenetic, either forming during rapidly
1082 oxidizing, but localized conditions during lithification of the sandstones, or from later diagenetic
1083 fluids moving through the sandstones (Lanza et al., 2014). As the Greenheugh capping unit has

1084 minimal evidence for late diagenesis, unlike the older sedimentary units of Yellowknife Bay, it is
1085 most likely to be detrital or formed during cementation. Mn is a common trace element in
1086 olivine, substituting for Fe^{2+} . As such, the dissolution of olivine and formation of magnetite
1087 under isochemical conditions may have precipitated a late-stage Mn-bearing mineral or
1088 amorphous component if aqueous fluids were minimal and the system was closed at the scale of
1089 the stratigraphic interval.

1090 4.3.2. The formation of alteration features across the Stimson formation

1091 Based on the Mastcam and Navcam images of the Stimson formation at the Greenheugh
1092 capping unit (Fig. 3, 4), in addition to the plots of data classifications across all Stimson
1093 localities (Fig. 5), it is evident that the Stimson formation at the Greenheugh pediment has not
1094 experienced the same later diagenetic events as those at the Emerson and Naukluft plateaus. The
1095 Greenheugh pediment lacks silica-rich halos, raised ridges, and has very few occurrences of
1096 calcium sulfate mineral veins which are much thinner than those sampled and studied at the
1097 Emerson and Naukluft plateaus (Fig. 4). Erosionally resistant polygons - polygonal diagenetic
1098 features associated with preferential cementation around fractures as a result of groundwater
1099 recharge - are also present to a lesser degree at the Greenheugh capping unit compared to the
1100 other Stimson localities and with a different morphology (Kronyak et al., 2019b).

1101 The lack of fractures and their associated diagenetic features, in addition to the different
1102 morphology of erosion resistant polygonal ridges at the Greenheugh pediment capping unit
1103 suggests different potential scenarios: 1) that at this location, groundwater flow was not as
1104 pervasive (Kronyak et al., 2019b), 2) that the Stimson formation at the Greenheugh capping unit
1105 experienced less compaction compared to that at the Emerson and Naukluft plateaus, or 3) that
1106 the Greenheugh pediment exhibits different material properties such as rock hardness and grain
1107 size. The Greenheugh pediment is situated at an elevation of -4150 m, which is 200 m higher in
1108 elevation than the Stimson formation at the Emerson and Naukluft plateaus (Fig. 1,2). As water
1109 is most likely to collect at topographical lows, it is possible that there was less groundwater
1110 available to the Greenheugh pediment to either create the well-formed, large polygons of the
1111 Emerson and Naukluft plateaus, or to generate sufficient fluid overpressure and hydrofracturing,
1112 providing support to scenario 1.

1113 Another factor which may have impacted the amount of groundwater available to the
1114 Greenheugh pediment capping unit is the time in which it was deposited and lithified. Although
1115 the Stimson formation at the pediment shares the common sedimentary architecture of the
1116 Stimson formation investigated earlier in the mission, and all outcrops are situated above the
1117 basal Siccar Point group unconformity, it is not clear whether these outcrops were lithified at the
1118 same time. While the sedimentary architecture indicates sediment accumulation under arid
1119 conditions (hence the absence of damp interdunes), water would have percolated through the
1120 sediments post-deposition to cement the strata. This could have happened at depth where
1121 subsurface water could not interact with the depositional surface (sensu Kocurek and Haveholm,
1122 1993) or could have happened after the terminal phase of the dune field's evolution. The sources
1123 of cementing fluid are likely to be different between the earlier Stimson localities and the
1124 Greenheugh pediment based on the different secondary minerals. The fan-shaped morphology of
1125 the Greenheugh pediment itself at the base of Gediz Vallis suggests that it may relate to surface
1126 waters flowing down and eroding the slope of Mt Sharp (Bryk et al., 2019). Meanwhile, the
1127 Stimson formation at the Emerson and Naukluft plateaus may have been cemented by rising

1128 groundwaters (Banham et al., 2018). Hence, the Greenheugh pediment capping unit may have
1129 been cemented at a different time, potentially one in which there was less groundwater
1130 availability in Gale crater.

1131 Alternatively, the difference in elevation could have resulted in less overburden to
1132 compact the sediments at the Greenheugh pediment capping unit, supporting scenario 2,
1133 however, it is not known to what extent the Stimson formation was buried at either location.
1134 Several sedimentary float are present on top of and close the Stimson formation at the Emerson
1135 and Naukluft plateaus (Wiens et al., 2020). Some of these float potentially include clasts of
1136 Stimson bedrock and their location on top of the Stimson formation itself suggests that additional
1137 sedimentary units did at some time overlie the Stimson formation at these localities in the past
1138 (Wiens et al., 2020). But as these deposits exist as float and not as a coherent outcrop, we cannot
1139 constrain with any certainty how much burial, and therefore compaction, occurred for the
1140 Stimson formation at the Emerson and Naukluft plateaus relative to the Greenheugh capping
1141 unit.

1142 4.3.3. Diagenesis at the unconformity

1143 Changes in geochemistry and mineralogy in the underlying Mt Sharp group and
1144 immediately overlying Stimson formation have been detected along the basal Siccar Point group
1145 unconformity indicating that this geological boundary may have provided a conduit for later
1146 diagenetic fluids (Achilles et al., 2020; Horgan et al., 2020; Newsom et al., 2018; Rudolph et al.,
1147 *this issue*; Thomson et al., *this issue*; Yen et al., 2021). At the Greenheugh pediment, Stimson
1148 and Carolyn Shoemaker bedrock close to the unconformity exhibit differences in geochemistry,
1149 and for the Carolyn Shoemaker formation mineralogy (Dehouck et al., *this issue*; Thompson et
1150 al., *this issue*; Thorpe et al., *this issue*). The basal Stimson formation is lower in MgO, Na₂O, and
1151 MnO relative to the mean composition of the Stimson formation, low in total sum of oxides, but
1152 high in CaO and H relative to the average Stimson formation (Fig. 7). The high CaO, H, and low
1153 totals are indicative of calcium sulfate addition, which is also supported by the mixing plots (Fig.
1154 13, S.6), and the high abundance of sulfates detected by APXS (Thompson et al., *this issue*). As
1155 this unique Stimson geochemistry is only observed for targets that are approximately 10 cm and
1156 50 cm from the unconformity surface, it suggests that this diagenesis did not permeate far into
1157 the overlying Stimson formation, indicating that it was lithified prior to the alteration.

1158 Calcium sulfate cements were also identified in the basal Stimson outcrop of the Marias
1159 Pass locality according to a lighter coloration of the bedrock and geochemical compositions that
1160 are high in CaO and low for all other major element oxides and totals (Newsom et al., 2018). At
1161 the Naukluft plateau, rover images also showed evidence of either silica-halo alteration or
1162 calcium sulfates concentrated at the Murray formation immediately below the unconformity (Fig.
1163 14). This indicates that the diagenesis that occurred at the unconformity between the Greenheugh
1164 pediment capping unit was widespread and also occurred along the unconformity at other
1165 Stimson localities.

1166 A possible scenario that could result in the precipitation of calcium sulfates and a
1167 decrease in MgO and alkali abundances includes acid-sulfate alteration, such as that which is
1168 hypothesized for the Murray formation of Marias Pass, the Oudam drilled sample in the Murray
1169 formation at the Naukluft plateau (Achilles et al., 2020), the alteration halos of the Emerson and
1170 Naukluft Plateaus (Hausrath et al., 2018; Yen et al., 2017, 2021), and in some places on Vera

1171 Rubin ridge (Rampe et al., 2020b). Acid-sulfate alteration can preferentially alter phenocrysts
1172 such as olivine, pyroxene, and feldspars over volcanic glass, leaving behind a silica residue, iron
1173 oxides, and sulfates and remobilizing MgO and the alkali elements (McCollom, et al., 2013a;
1174 McCollom et al., 2013b; Tosca et al., 2004). However, unlike the potentially acid-sulfate altered
1175 mudstones and sandstones of Marias Pass, the basal Stimson formation at the Greenheugh
1176 capping unit does not have an enrichment in SiO₂ and TiO₂ (Fig. 7), indicating that any acid-
1177 sulfate alteration at this locality may have been less extensive than that which modified the
1178 geochemistry and mineralogy of the rocks at the Emerson and Naukluft plateaus.

1179 vThe alteration of Stimson sandstones at the unconformity suggests that either a weakly
1180 oxic, sulfate-bearing fluid or fewer resurgences of late diagenetic sulfate-bearing groundwater
1181 fluids may have migrated along the unconformity, permeating the lithified sandstone directly
1182 above and the mudstone below. These fluids may have not been able to permeate as extensively
1183 into the upper Stimson formation at the Greenheugh capping unit and create polygonal fractures
1184 due to the greater elevation resulting in less groundwater availability. The difference in
1185 sandstone cement between the Emerson and Naukluft plateaus and the Greenheugh pediment
1186 capping unit may have also been a factor in impeding the formation of polygons and decreasing
1187 the permeability of the Stimson sandstone at the Greenheugh capping unit. The Big Sky and
1188 Okoruso drilled samples of the Emerson and Naukluft plateaus both contain iron oxides
1189 (magnetite and hematite), some calcium sulfates (bassanite), and an amorphous component
1190 within the sandstone cement (Yen et al., 2017). The Edinburgh drilled sample from the
1191 Edinburgh interval of the Greenheugh pediment capping unit contains phyllosilicates in addition
1192 to magnetite and an amorphous component (Rampe et al., 2020). Phyllosilicates decrease the
1193 permeability of a sandstone, which may have further impacted fluid migration at this locality and
1194 the material properties of the Stimson formation at the Edinburgh interval compared to that at the
1195 Emerson and Naukluft plateaus.

1196 The Carolyn Shoemaker formation that underlies the unconformity at the Greenheugh
1197 pediment shows geochemical differences relative to the average Carolyn Shoemaker formation
1198 that are indicative of alteration (Fig. 7, 13; Dehouck et al., *this issue*; Gasda et al., *this issue*).
1199 Furthermore, the Hutton drill location in the Carolyn Shoemaker formation contains 8.7 wt%
1200 iron oxides, 4.7 wt% cristobalite and 5.2 wt% Opal-CT, silica polymorphs that can form as a
1201 result of the dissolution of mafic minerals (Thorpe et al., *this issue*). The immediately underlying
1202 Carolyn Shoemaker bedrock around the Hutton drill site is lower in oxide totals (94.9 wt%
1203 compared to 97.3 wt%), and higher in CaO compared to average Carolyn Shoemaker bedrock
1204 (Fig. 7). This can be indicative of calcium sulfate cement which has been largely detected by the
1205 ChemCam instrument and correlated to light-toned cements in MAHLI images acquired in the
1206 traverse leading up to the Greenheugh pediment (Gasda et al., *this issue*; Nelleson et al., 2021).
1207 However, the mineralogy of the Hutton drilled sample acquired below the capping unit only
1208 contains 0.6 wt% anhydrite (Thorpe et al., *this issue*), which is some of the lowest abundances of
1209 crystalline calcium sulfate detected in the Carolyn Shoemaker formation since the drilled
1210 samples of the Pahrump Hills (Rampe et al., 2017), and sulfur is not detected here with APXS
1211 (Thompson et al., *this issue*). F abundances also increase in the Carolyn Shoemaker formation
1212 below the unconformity at the Hutton drill site (Dehouck et al., *this issue*; Fig. 8B), including for
1213 alteration features such as veins and raised ridges (Gasda et al., *this issue*). This correlated with
1214 the increase in CaO concentrations suggesting the presence of fluorapatite which is confirmed in
1215 the CheMin results of the Hutton drilled sample (Thorpe et al., *this issue*).

1216 The current hypothesis for the distinctive geochemistry and mineralogy of the Hutton
1217 drill hole and surrounding sediments is that warm or acidic diagenetic fluids circulated here and
1218 concentrated below the capping unit (Dehouck et al., *this issue*; Gasda et al., *this issue*; Forni et
1219 al., 2021; 2016). However, these F-rich diagenetic fluids do not appear to have permeated into
1220 the overlying Stimson formation. Fluorine is a fluid-mobile element which is in high abundance
1221 in the Murray formation bedrock and diagenetic features below the Stimson formation at the
1222 Marias Pass locality and in the Carolyn Shoemaker formation's Hutton Zone of Glen Torridon
1223 (Forni et al., 2021; 2016). However, above the unconformity, fluorine detections are sparse in the
1224 Stimson formation, with only one detection in the pediment capping unit (Fig. 8B; Forni et al.,
1225 2021). Meanwhile, the rare detections of fluorine in the Stimson sandstone instead suggests that
1226 these may instead relate to detrital phases, similar to what is hypothesized for the fluorine phases
1227 detected in the Bradbury group sandstones (Forni et al., 2013). The lack of fluorine detected in
1228 the basal Stimson that also contains intraclasts of the underlying mudstones further supports that
1229 the alteration of the Carolyn Shoemaker formation around the Hutton drill hole occurred after the
1230 formation of the overlying Stimson formation.

1231 5 Conclusions

1232 The ChemCam geochemical data of the Greenheugh pediment capping unit suggests that
1233 this unit is a part of the Stimson formation investigated at the Emerson plateau, Naukluft plateau,
1234 and Murray Buttes localities. However, minor but statistically significant geochemical variations
1235 exist for SiO₂, MgO, MnO, Al₂O₃, and Na₂O. These slight geochemical differences are indicative
1236 of higher abundances of mafic minerals and their diagenetic products at the Stimson formation of
1237 the Greenheugh capping unit, which is supported by mineralogical data from CheMin. The
1238 higher abundance of mafic minerals preserved at the Greenheugh pediment capping unit is
1239 unlikely to relate to physical grain segregation with transportation across all Stimson localities
1240 due to variable cross-set dip directions indicative of seasonal wind variations which complicate
1241 the geochemical record of net sediment transport at this locality. Similarities in grain size and the
1242 high abundances of pyroxene and plagioclase feldspar in addition to olivine is also indicative that
1243 this aeolian sandstone has not undergone significant transport from its source (Cornwall et al.,
1244 2015). Instead, we hypothesize that the sediments of the Stimson formation at the Greenheugh
1245 pediment capping unit are predominately sourced from a nearby olivine-rich unit which may
1246 relate to the olivine-bearing marker beds that have been detected close to the pediment, farther
1247 up the slopes of Mt Sharp (Rudolph et al., 2019).

1248 While the Stimson aeolian sand dunes migrated in the Hesperian, they were also able to
1249 erode and incorporate clasts from the underlying Mt Sharp group into the base of the sand dunes,
1250 evidenced by the ChemCam geochemical data and images of the basal Stimson unit at the
1251 Greenheugh pediment. However, due to the fine-grained nature of the Mt Sharp group
1252 mudstones, it is unlikely that mudstones were preserved for long in the Stimson sand dunes and
1253 were instead winnowed away into suspension during dune migration. Alternatively, the
1254 intraclasts within the basal Stimson formation may relate to the onset of aeolian deposition as a
1255 buried lag deposit, similar to what is hypothesized for the Murray intraclasts preserved in the
1256 Stimson formation at Marias Pass (Newsom et al., 2018). Either scenario provides additional
1257 evidence for sediment recycling in Gale crater by aeolian processes.

1258 After the deposition of the Stimson sand dunes, their sediments interacted with surface
1259 waters forming a sandstone cement through the dissolution of the most reactive components,

1260 likely volcanic glass and olivine. These fluids formed a perched aquifer at the base of the
1261 aeolian sandstone at all localities, forming the concretionary bedrock. Geochemically, the
1262 sandstone cement of the Stimson formation is difficult to distinguish from that of the Emerson
1263 and Naukluft plateaus with the exception of MnO, however, differences in secondary mineralogy
1264 and the persistence of olivine in the Greenheugh pediment capping unit suggest that the early
1265 diagenetic fluid was more alkaline than that which cemented the Stimson formation at other
1266 localities. This difference in early diagenetic fluid chemistry could relate to the Greenheugh
1267 pediment capping unit having a different fluid source, and/or being cemented at a different time
1268 in the geological record of the Stimson sand sea.

1269 Once the Stimson formation was lithified, several diagenetic events likely relating to
1270 upwelling groundwater occurred to form erosion resistant polygons, silica-rich halos, calcium
1271 sulfate veins, and alteration along the unconformity. These later diagenetic events were more
1272 prevalent in the Stimson formation preserved at the Emerson and Naukluft plateaus, suggesting a
1273 greater availability of groundwater fluids at these sites than the Greenheugh pediment. This
1274 greater availability of groundwater at the Emerson and Naukluft plateaus could relate to their
1275 lower elevation in Gale crater, greater burial, or a difference in age between these Stimson
1276 deposits (i.e., Emerson and Naukluft Stimson are older than the Greenheugh capping unit and
1277 were cemented at a time when there was more groundwater present in Gale crater). One of the
1278 last episodes of diagenesis to occur likely relates to the unconformity, which is shared across all
1279 localities. The diagenesis below the unconformity involved fluorine-rich, potentially warm fluids
1280 that did not permeate into the overlying Stimson formation. The diagenesis along the
1281 unconformity suggests that acidic, sulfate-bearing fluids migrated along the unconformity and
1282 altered the basal Stimson formation, supporting the hypotheses of previous studies.

1283 Overall, the ancient aeolian environment under which these Stimson sandstones were
1284 deposited was dynamic, recycling nearby material as the dunes migrated across the surface.
1285 These results support that liquid water was stable at the surface in the Hesperian and was
1286 available as groundwater for multiple diagenetic events after deposition along bedrock
1287 weaknesses and the basal Stimson formation in Gale crater.

1288

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1297 the Planetary Data System (PDS). The geochemical data can be found at Wiens (2013a). RMI
1298 mosaics can be found at Wiens (2013b). The derived data which includes the ChemCam targets
1299 used in the study, target classifications, and clustering results, can be found at Bedford (2022).

1300

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1806

1807 **Figure Headings**

1808 **Figure 1.** Map of the Stimson formation localities investigated to date along the Curiosity
 1809 rover's traverse. The labelled, colored units show the present-day extent of the Stimson
 1810 formation (Siccar Point group) at the Emerson Plateau (blue), Naukluft Plateau (green), Murray
 1811 Buttes (yellow), and Greenheugh pediment capping unit (orange). The solid white line shows the
 1812 traverse, and the yellow waypoints mark key drill sites in the Mt Sharp group mudstones and
 1813 Siccar Point group Stimson formation sandstones. The black streaks oriented from the NE to the
 1814 SW derive the active Bagnold dune field.

1815 **Figure 2.** Stratigraphic column of the geological units investigated by the MSL Curiosity rover
 1816 in Gale crater, Mars. The Stimson formation sandstones are represented as tan-colored blocks.
 1817 Dots are drill hole samples with those discussed in this paper labelled as BK (Buckskin), Big Sky
 1818 (BS), Greenhorn (GH), Lubango (LB), Okoruso (OK), Oudam (OU), Hutton (HU), Edinburgh
 1819 (EB). Mt Sharp group drill samples are labelled purple, Stimson formation drills are labelled red.
 1820 Image credit is courtesy of the MSL sedimentology and stratigraphy group.

1821 **Figure 3.** Mastcam mosaics of the Stimson formation at A) the top of the Greenheugh pediment
 1822 (mcam14517, sol 2702), B) side view of the Greenheugh pediment (mcam14007, sol 2683), C)
 1823 Williams outcrop at the Emerson plateau showing cross-bedding and a vertical light-toned halo
 1824 (mcam004777, sol 1087), D) Naukluft plateau at the unconformity (dashed white line) of the
 1825 Murray formation and the Stimson formation (mcam0597, sol 1275). MAHLI images of the
 1826 bedrock drill targets for the Stimson formation at each locality are also provided in the insets: a)
 1827 is of the Edinburgh target of the Edinburgh interval (sol 2703, after DRT), b) The Machir Bay
 1828 target of the Ladder interval (sol 2699, after DRT), c) The target Big Sky (sol 1114, after DRT),
 1829 and d) the target Okoruso (sol 1330, after DRT). Image credits for the Mastcam mosaics:
 1830 NASA/JPL-Caltech/MSSS.

1831 **Figure 4.** ChemCam RMI image mosaics, MAHLI images, and Navcam images showing the
 1832 diversity of alteration features within the Stimson formation at each locality.

1833 **Figure 5. A.** Distribution of ChemCam classifications between each Stimson locality where each
 1834 count equals to one ChemCam observation point. Bedrock low totals is the number of bedrock
 1835 observation points that had total sum of oxides <90 wt%. **B.** Distribution of ChemCam
 1836 classifications between each Greenheugh stratigraphic interval (excluding the basal Stimson
 1837 unit). Observation points that targeted float rocks and soils at these localities were not included.
 1838 Sampling bias will have affected the number of ChemCam points on alteration features relative
 1839 to those of bedrock, particularly for the silica-rich halos, as they were intentionally prioritized in
 1840 the Emerson and Naukluft plateaus.

1841 **Figure 6.** Histograms of ChemCam major element oxides for the bulk rock dataset at each
 1842 Stimson locality. These data exclude those outside the 90-105 wt% range for total sum of oxides.
 1843 Binning was selected based on the Minitab default setting which defines the histogram intervals
 1844 according to the number of observations and the maximum and minimum values in the dataset.
 1845 For our dataset, the number of intervals range between 21 and 34. The black vertical line shows
 1846 the mean value of the major element oxide at each locality. The y-axis shows the number of data

1847 points in each bin (frequency). The histogram for MnO was restricted to 1 wt% but has outliers
 1848 to higher values which skews the mean, particularly for Greenheugh.

1849 **Figure 7.** Boxplots of geochemical variation associated with the unconformity for the bedrock
 1850 datasets of the underlying Carolyn Shoemaker formation and overlying Stimson formation. FeO_T
 1851 and TiO₂ showed little variation and were not included in the plots. Dashed lines show the mean
 1852 composition of the Stimson formation at Greenheugh (red) and the Carolyn Shoemaker
 1853 formation (blue). Dotted lines show the uncertainty around the respective sample means through
 1854 2x the standard error.

1855 **Figure 8. A.** H peak area data distribution for the Greenheugh pediment capping unit according
 1856 to the different stratigraphic intervals. The concretion-rich Hutton interval has the lowest peak H
 1857 abundance while the non-concretionary Edinburgh and Ladder Intervals have the higher H
 1858 abundance. **B.** F variation with distance from the unconformity for the Stimson formation and the
 1859 underlying Mt Sharp group. Alteration features (halos, concretions, raised ridges, veins) and
 1860 alteration-free bedrock are shown. **C.** Annotated Mastcam image (mcam_14083) of the side of
 1861 the Greenheugh pediment showing the division between the concretionary Glean Beag Interval
 1862 and the Edinburgh Interval that has fewer concretions concentrated towards the base. Image
 1863 credit for the Mastcam mosaic used in C: NASA/JPL-Caltech/MSSS.

1864 **Figure 9.** Dendrogram of the Cluster analysis results for Stimson across all localities.

1865 **Figure 10.** Pie charts showing the distribution of the clusters across Stimson localities. The red
 1866 and blue wedges correspond to the relative abundances of mafic minerals present at each locality.

1867 **Figure 11.** Biplots showing the geochemistry of the Stimson formation at the Greenheugh
 1868 pediment capping unit compared to that of the earlier Stimson formations, the geochemical
 1869 compositions of the clusters, and the chemical compositions of the primary igneous minerals
 1870 identified in the Big Sky drilled sample of the Stimson sandstone (Yen et al., 2017), and the
 1871 Gobabeb scooped sample of the active aeolian Bagnold dunes (Achilles et al., 2017). Bulk
 1872 Stimson compositions are shown as density contours in addition to a linear regression model
 1873 (dashed black line). Cluster compositions also show 1 standard deviation. **A. A**
 1874 Al₂O₃+Na₂O+K₂O vs MgO + FeO_T plot to show the variation in chemistry that relates to mafic
 1875 and felsic minerals. **B.** A MnO vs MgO plot to show the unusual chemistry of Cluster 5

1876 **Figure 12.** Annotated ChemCam RMI mosaics of targets **A.** Mowry (sol 1080) of the Emerson
 1877 Plateau, **B.** Bentiaba (sol 1320) of the Naukluft plateau, **C.** Lowther Hills (sol 2697) of the
 1878 Gleann Beag interval, and **D.** Dunedin (sol 2720) of the Edinburgh Interval. Colored circles show
 1879 the cluster memberships of the associated ChemCam observation points.

1880 **Figure 13: A.** and **B.** show geochemical plots of the ChemCam targets immediately above and
 1881 below the unconformity. Each point is an observation point composition with green points
 1882 related to the Carolyn Shoemaker formation (target names are Ariundle, Shannochie,
 1883 Corriecravie, Hutton), and orange and gray points related to the Stimson formation (target names
 1884 are Huttons section and Gleann Beag). Black arrow shows the geochemical trend relating to Ca-
 1885 sulfates. The solid line represents a mixing line between the average composition of the Stimson
 1886 formation at the Greenheugh pediment and the bulk Carolyn Shoemaker formation (Dehouck et

1887 al., *this issue*). Percentages along the mixing line show the percentage of Stimson (e.g., 25% is
 1888 25% Stimson, 75% Murray). Dashed lines represent 1 standard deviation of this mixing line. **C.**
 1889 and **D.** show the MAHLI images of Stimson targets near the unconformity at the Greenheugh
 1890 pediment capping unit. Yellow arrows point to angular mudstone clasts in the Stimson formation.
 1891 White arrows point to light-toned, spherical grains that may also relate to rounded mudstone
 1892 clasts (Banham et al., *this issue*).

1893 **Figure 14.** Mastcam image of light-toned alteration features in the underlying Murray formation
 1894 pooling at the unconformity with the Stimson formation and breaching through the fractures.
 1895 Image was taken at the Naukluft plateau (mcam05931, sol 1267). Image credit: NASA/JPL-
 1896 Caltech/MSSS.

1897 **Table 1.** Basic statistics (mean, standard error of the mean (SE Mean), standard deviation
 1898 (StDev), minimum (Min), median (Med), and maximum (Max)) for the bulk rock data at each
 1899 locality. Mean and median values are highlighted based on whether they represent the largest
 1900 value (red) or the lowest value (blue), taking into consideration the standard error of the mean.
 1901 Both standard deviation and error are provided as standard deviation is representative of the
 1902 dispersion of the individual data values to the mean, while the standard error of the mean
 1903 considers the number of points in the dataset and measures how far the sample mean of the data
 1904 is from the true population.

1905 **Table 2.** Mean composition, standard error of the mean (+/-), and standard deviation (std) of the
 1906 derived clusters. Colors highlight the highest abundances (red), the lowest (blue), and the
 1907 gradation in between for each of the five clusters.
 1908

Figure 1.

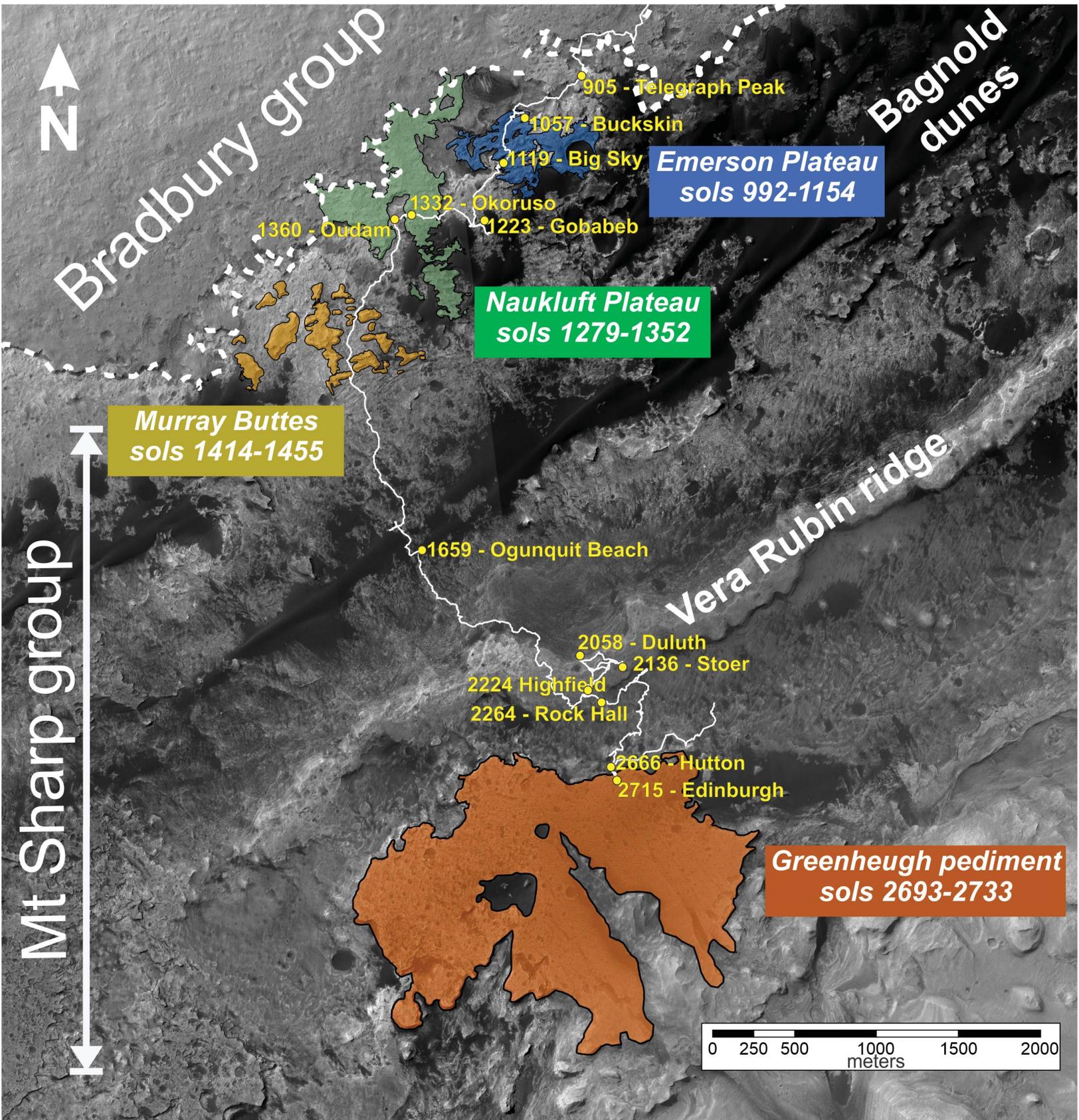


Figure 2.

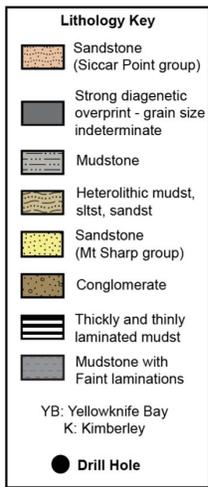
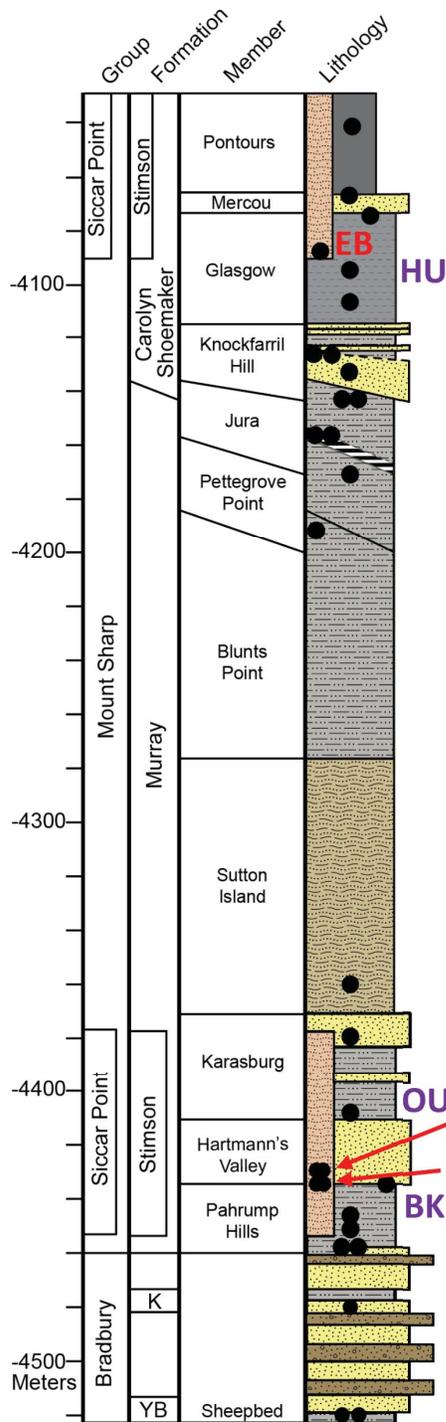


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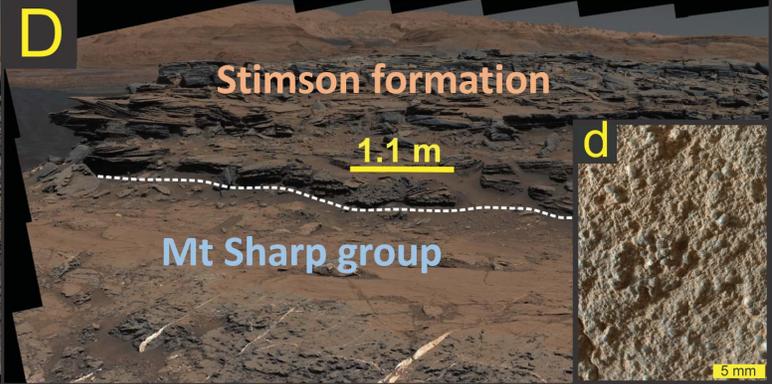
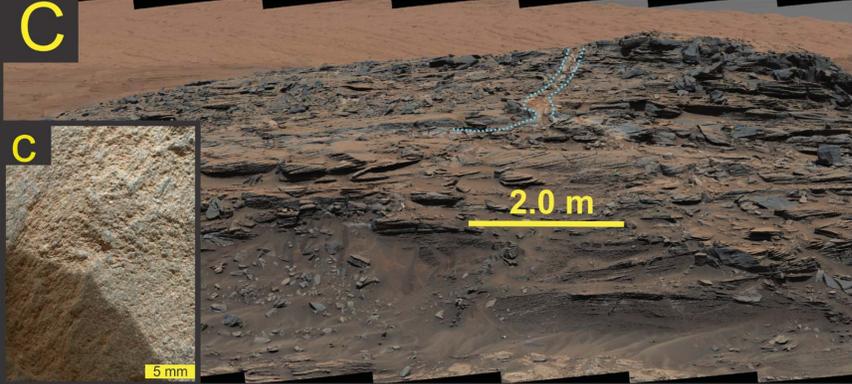
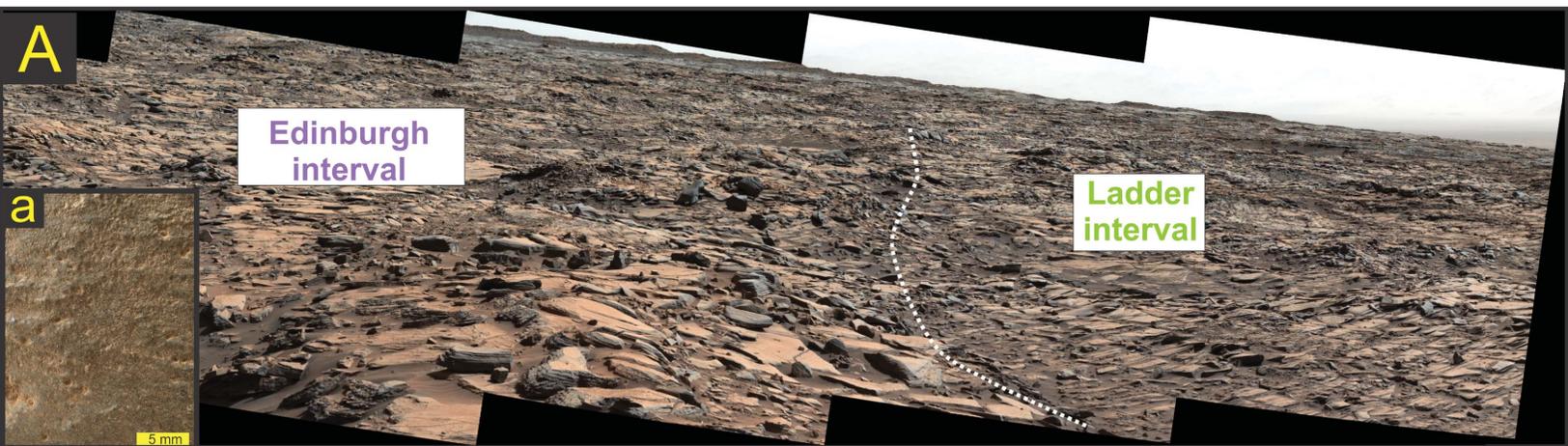


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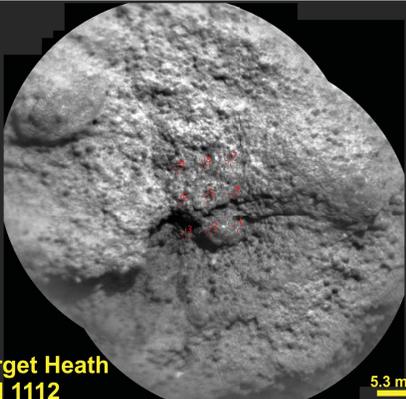
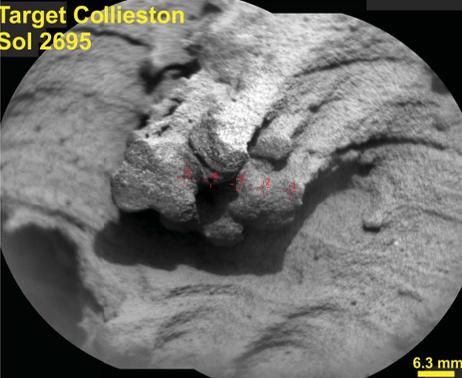
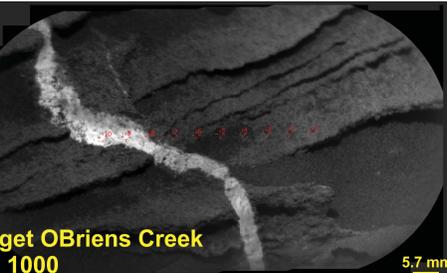
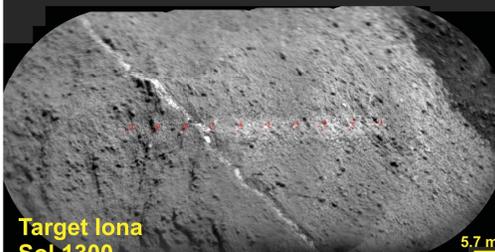
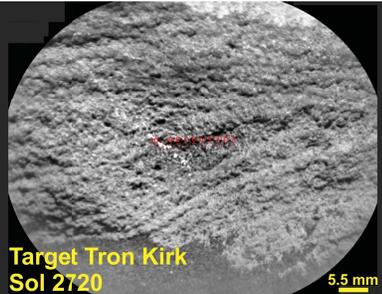
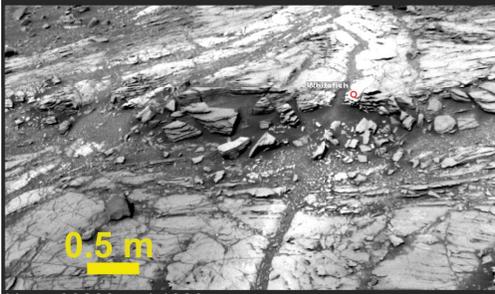
	Emerson plateau	Naukluft plateau	Greenheugh
Concretions	 <p>Target Heath Sol 1112</p> <p>5.3 mm</p>	<p>Sol 1278 – Target Maieberg Focus merge product from ~ 4 cm standoff</p>  <p>1278MH0001700000500011R0c</p> <p>1 cm</p>	<p>Target Collieston Sol 2695</p>  <p>6.3 mm</p>
Veins	 <p>Target OBriens Creek Sol 1000</p> <p>5.7 mm</p>	 <p>Target Iona Sol 1300</p> <p>5.7 mm</p>	 <p>Target Tron Kirk Sol 2720</p> <p>5.5 mm</p>
Halos	 <p>0.5 m</p> <p>Ncam00263, sol 1093</p>	 <p>0.5 m</p> <p>Ncam00353, sol 1317</p>	<p>No halos detected</p>

Figure 5.

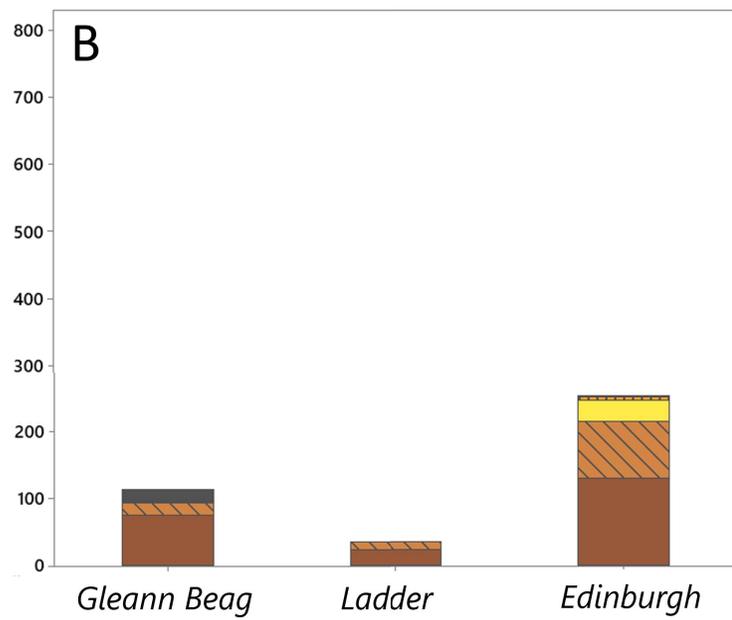
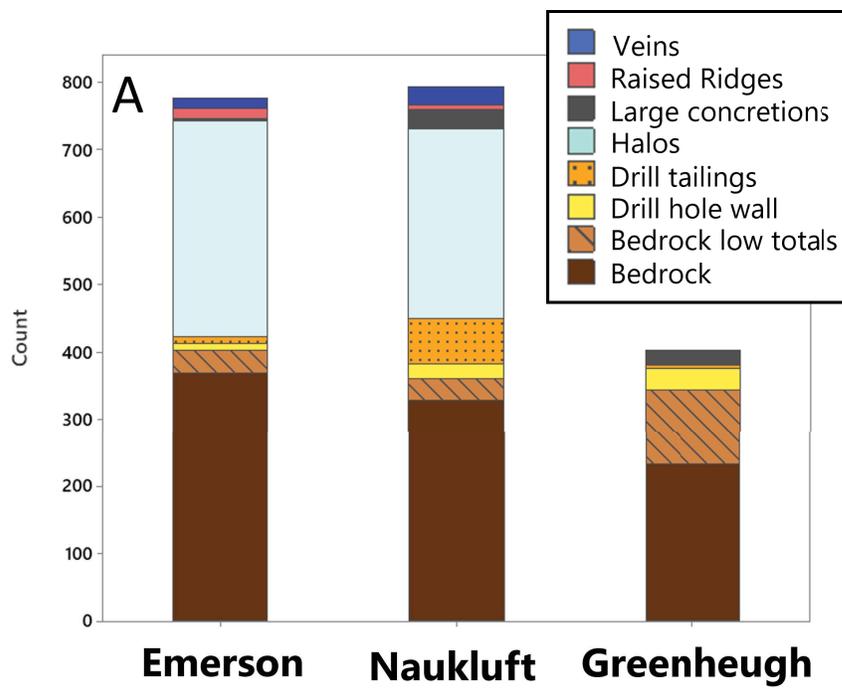
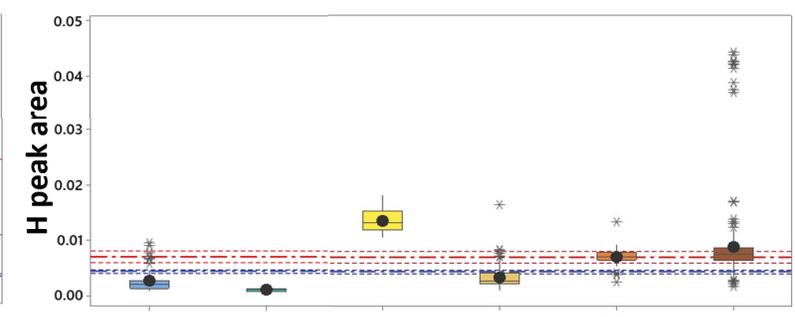
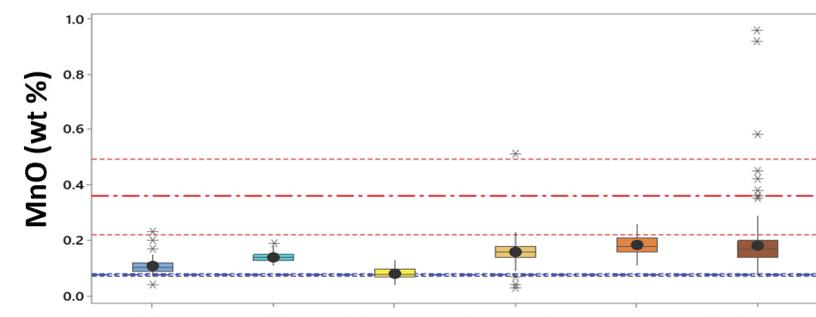
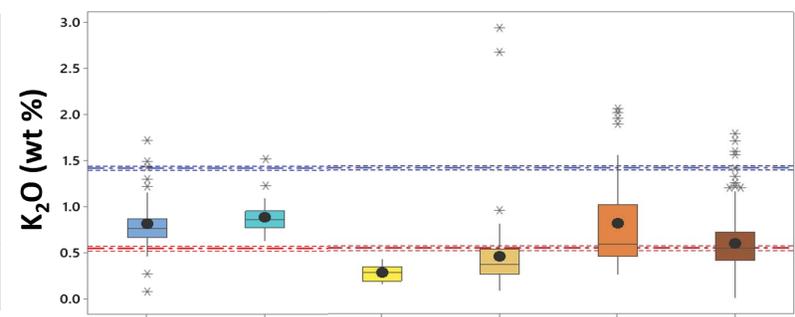
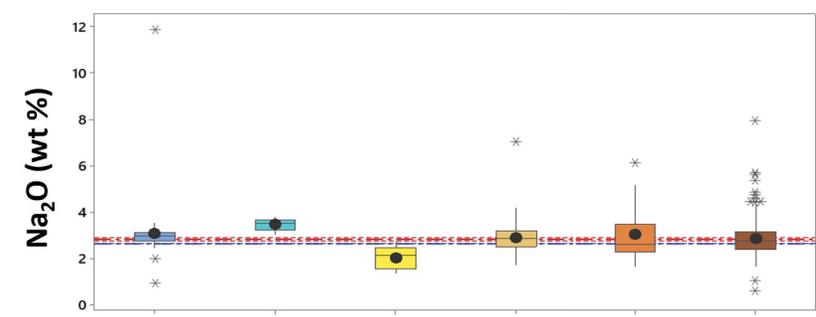
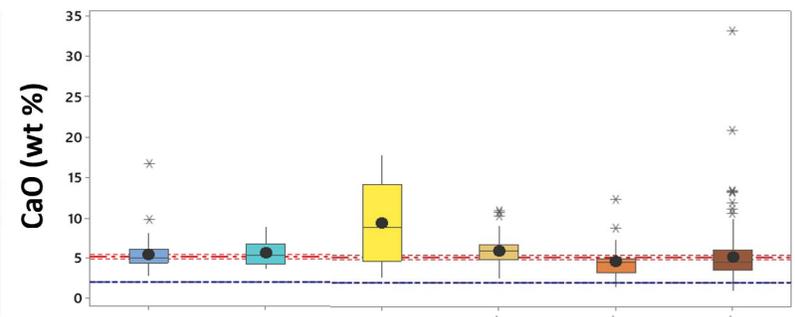
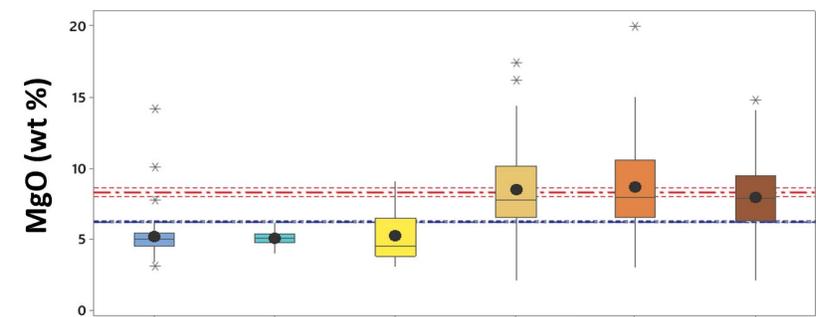
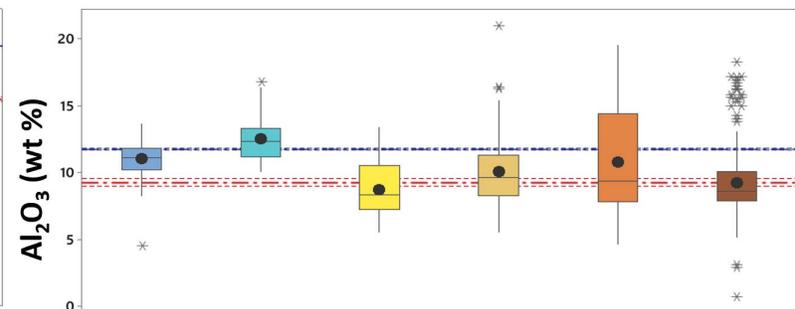
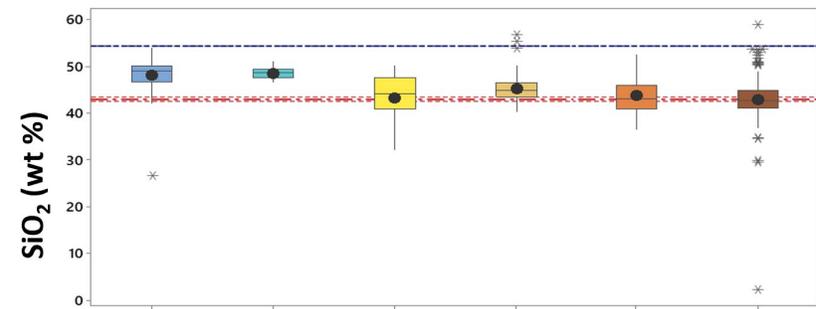


Figure 6.



CS CS Stimson Glean B. Ladder Edinburgh
Hutton Unconf. Unconf. Interval Interval Interval

CS CS Stimson Glean B. Ladder
Hutton Unconf. Unconf. Interval Interval

Figure 7.

Frequency

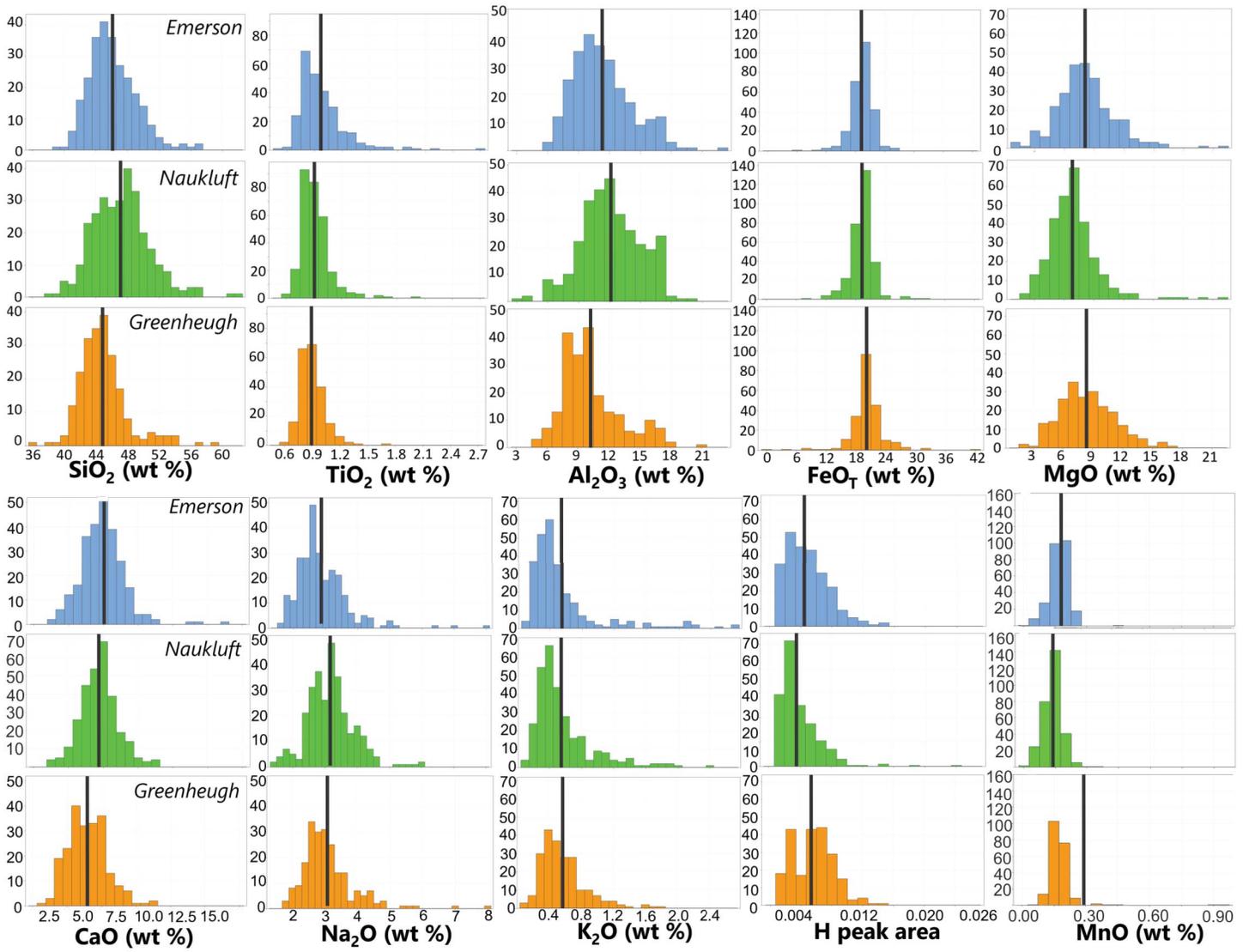


Figure 8.

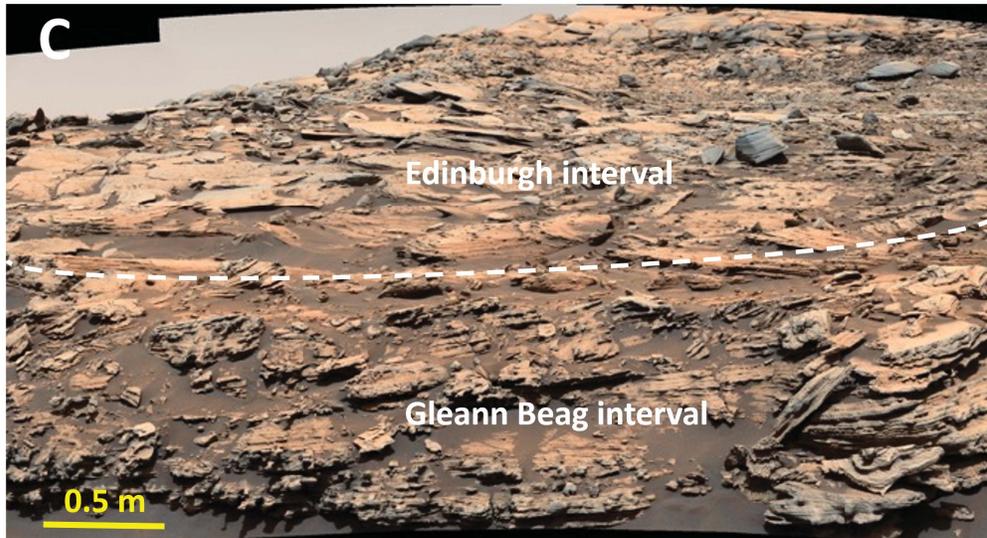
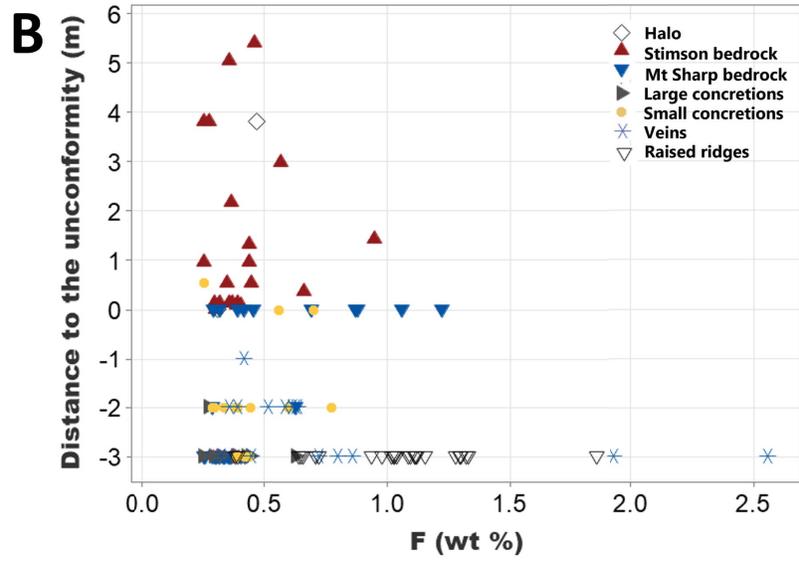
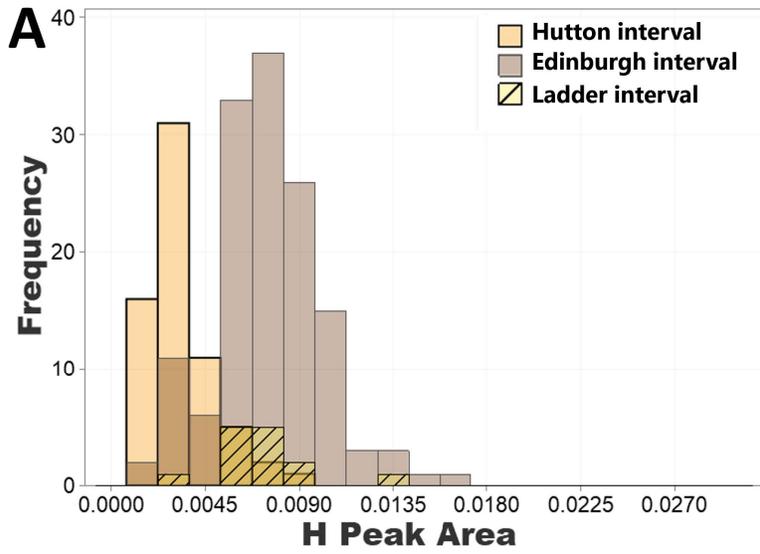


Figure 9.

Dendrogram

Complete Linkage, Euclidean Distance

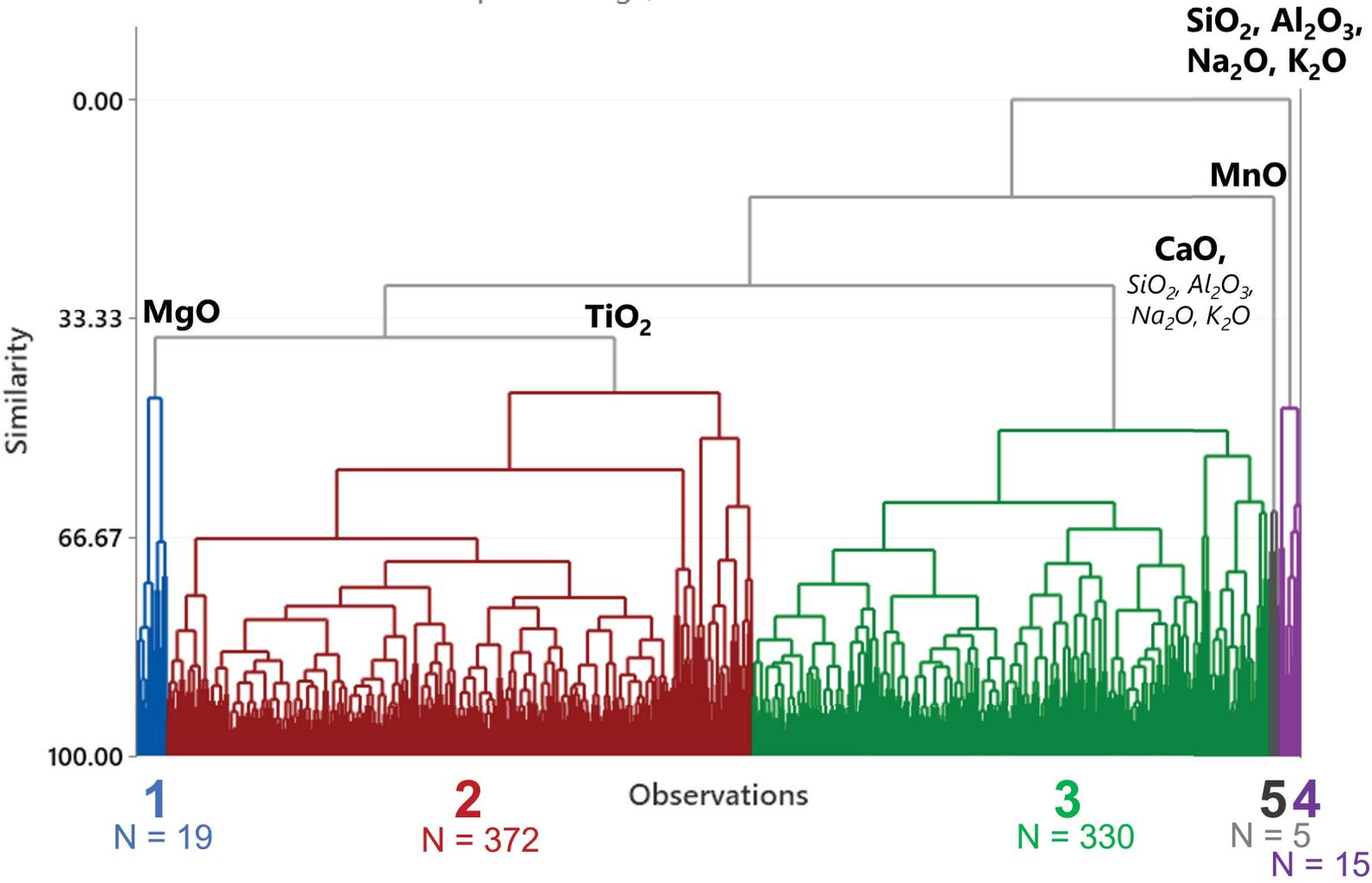
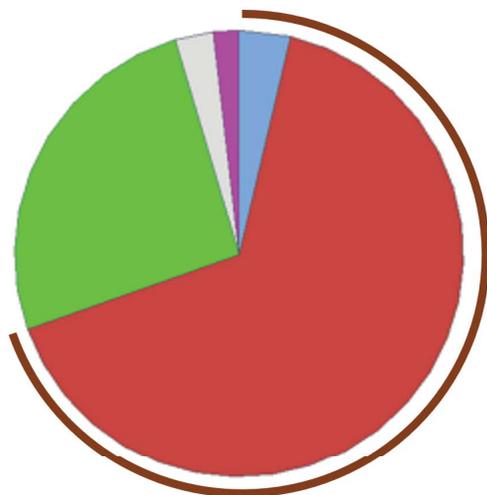


Figure 10.

Greenheugh pediment

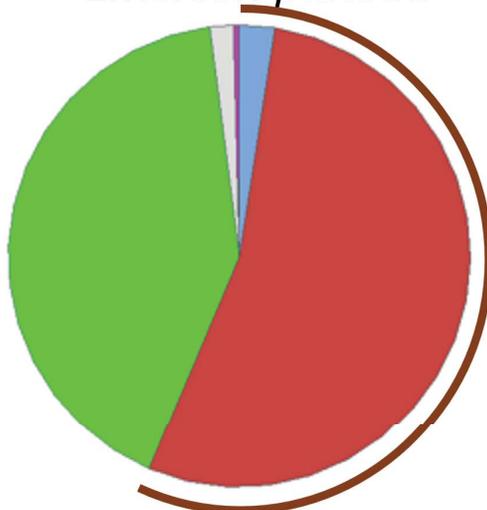


Mafic (69.6%)

Cluster

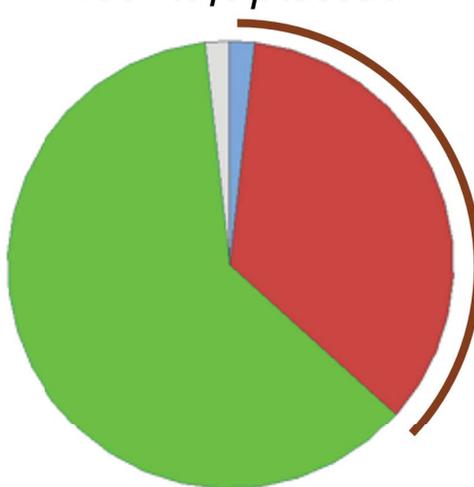
- 1 – high MgO
- 2 – MgO, FeO_T, and TiO₂
- 3 – CaO, SiO₂, Al₂O₃, and alkalis
- 4 – high SiO₂, Al₂O₃, and alkalis
- 5 – high MnO

Emerson plateau



Mafic (56.4%)

Naukluft plateau



Mafic (36.7%)

Figure 11.

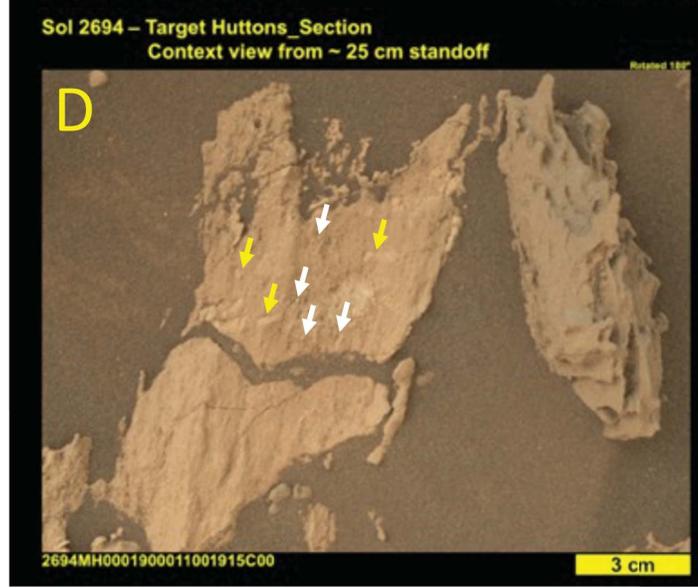
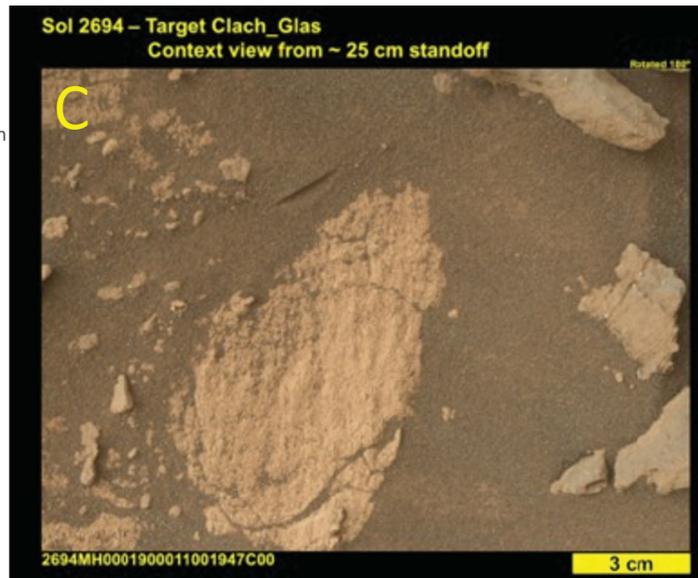
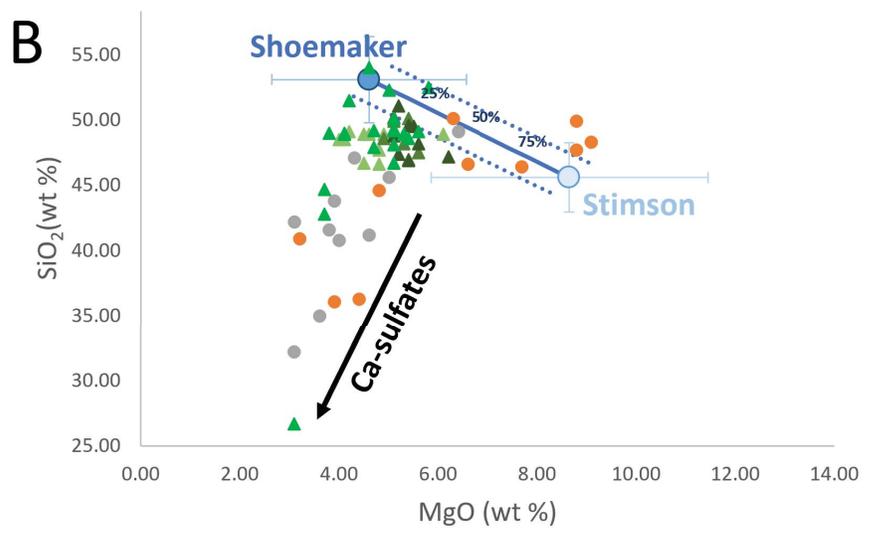
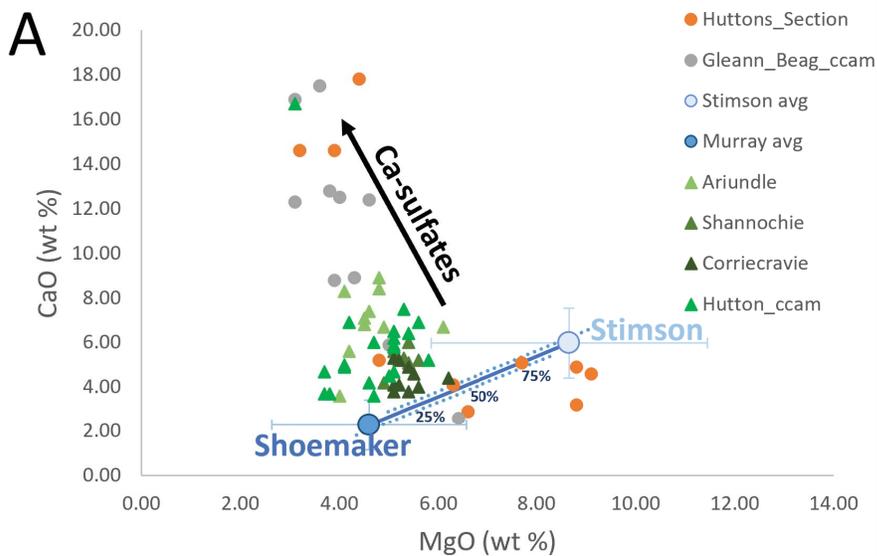


Figure 12.

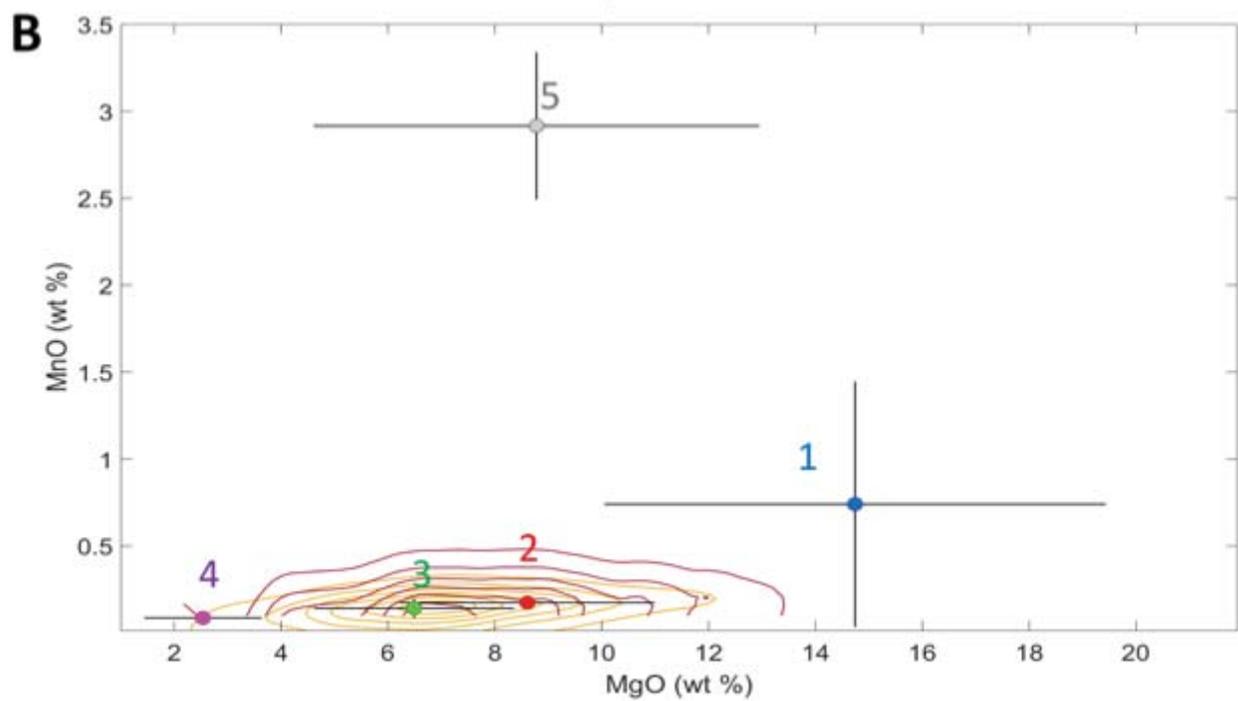
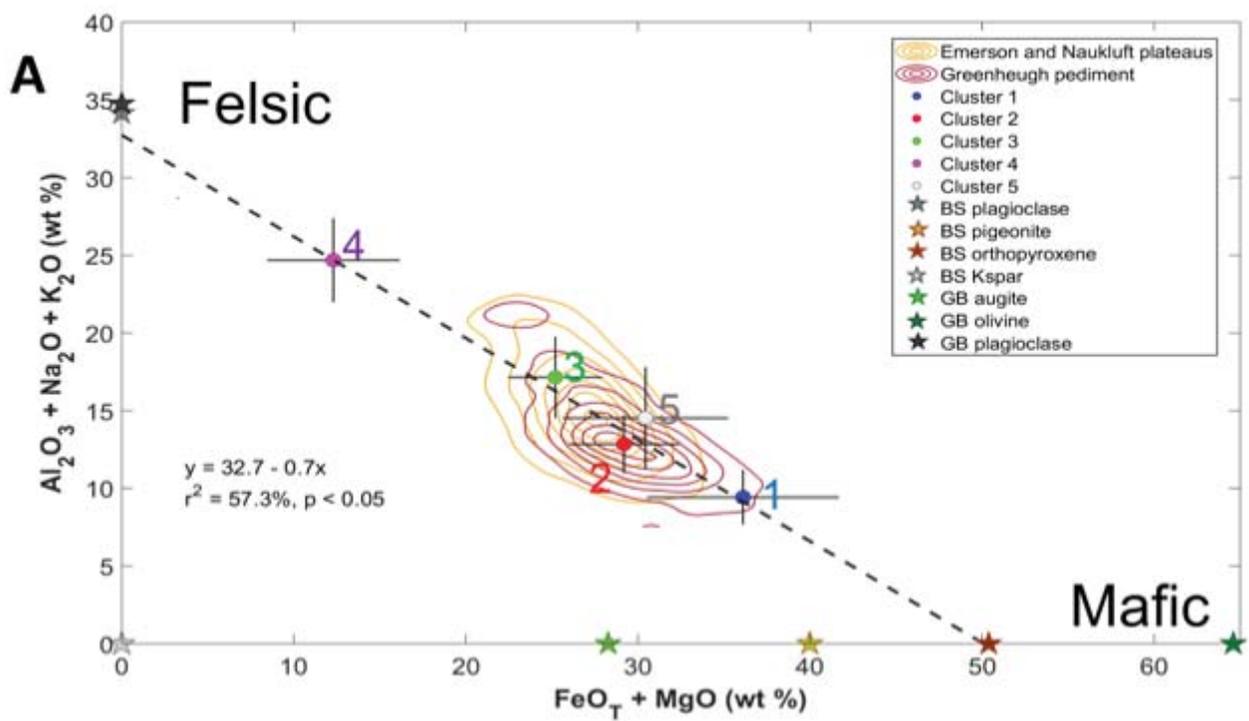
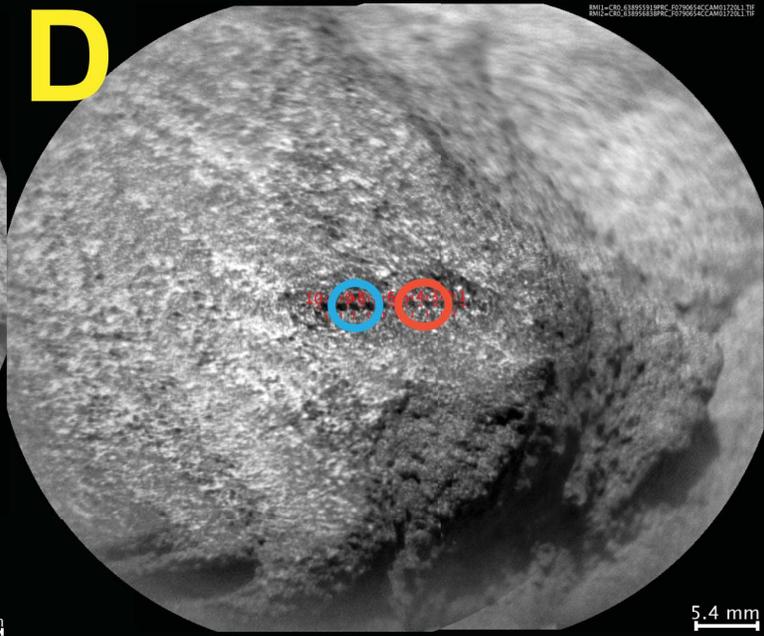
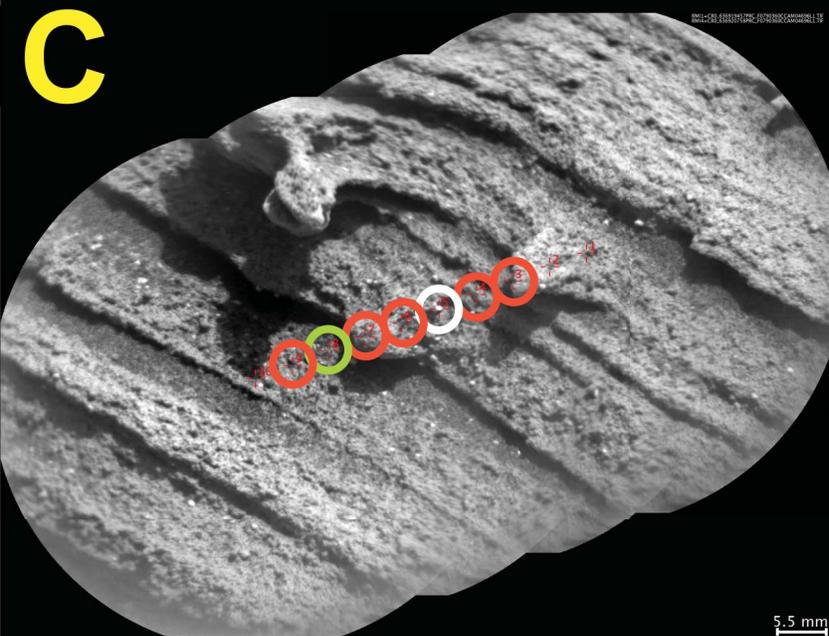
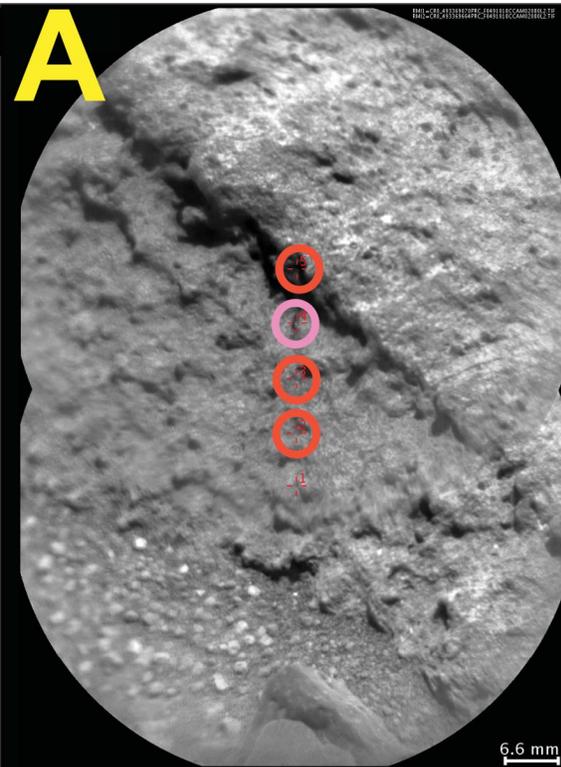
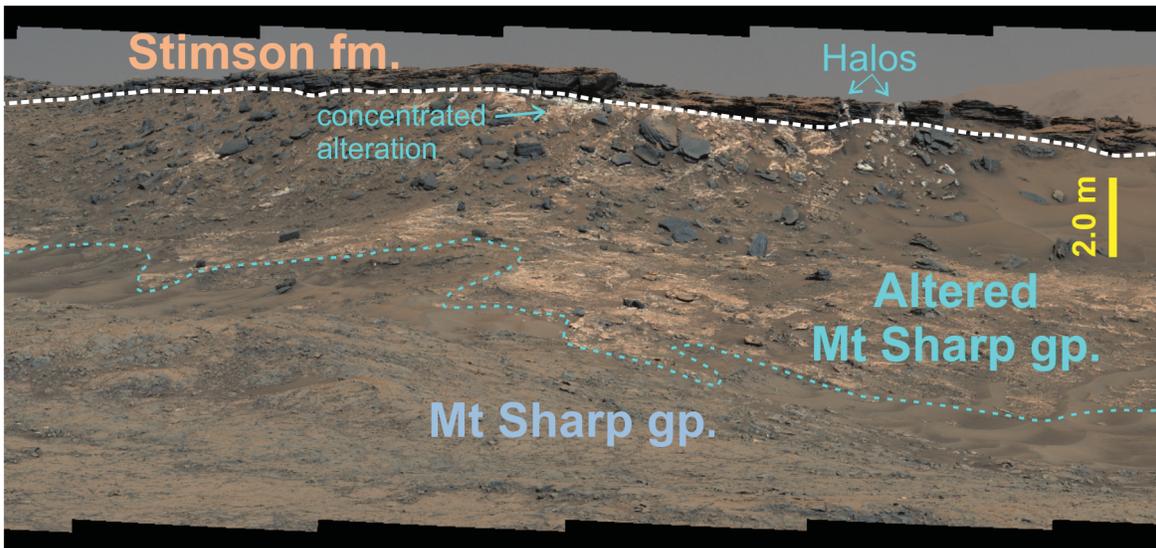


Figure 13.



○ Cluster 1 (mafic)
 ○ Cluster 2 (mafic avg)
 ○ Cluster 3 (felsic avg)
 ○ Cluster 4 (high MnO)
 ○ Cluster 5 (felsic)

Figure 14.



Variables	Locality	90-105 wt% totals					
		Mean	SE	Mean	StDev	Min	Med
SiO ₂	Emerson	46.1	0.2	3.0	38.9	45.6	57.1
	Naukluft	47.2	0.2	3.6	37.6	47.3	61.7
	Greenheugh	44.9	0.2	3.1	35.6	44.5	59.0
TiO ₂	Emerson	1.0	0.0	0.3	0.5	0.9	2.7
	Naukluft	0.9	0.0	0.2	0.6	0.9	2.0
	Greenheugh	0.9	0.0	0.1	0.6	0.9	1.7
Al ₂ O ₃	Emerson	11.3	0.2	2.9	5.7	10.8	22.5
	Naukluft	12.1	0.2	2.9	3.3	12.0	19.6
	Greenheugh	10.3	0.2	2.9	4.6	9.6	21.0
FeO _T	Emerson	19.2	0.1	2.3	6.6	19.5	26.0
	Naukluft	19.3	0.1	2.4	7.8	19.4	32.1
	Greenheugh	20.1	0.3	3.7	0.9	20.3	41.2
MgO	Emerson	8.1	0.2	2.9	0.9	7.7	21.6
	Naukluft	6.8	0.2	2.5	2.0	6.7	22.0
	Greenheugh	8.4	0.2	2.8	2.1	8.1	17.4
CaO	Emerson	6.2	0.1	2.0	1.6	6.2	17.2
	Naukluft	6.0	0.1	1.7	1.5	6.0	11.5
	Greenheugh	5.0	0.1	1.8	1.0	4.9	11.1
Na ₂ O	Emerson	2.9	0.1	0.8	1.5	2.7	8.1
	Naukluft	3.8	0.0	0.7	1.5	3.2	6.0
	Greenheugh	3.1	0.1	0.8	1.7	2.9	8.0
K ₂ O	Emerson	0.6	0.0	0.5	0.1	0.4	2.7
	Naukluft	0.6	0.0	0.4	0.1	0.4	2.4
	Greenheugh	0.6	0.0	0.3	0.1	0.5	1.8
MnO	Emerson	0.2	0.0	0.2	0.0	0.2	2.6

Naukluft	0.1	0.0	0.1	0.0	0.1	0.3
Greenheugh	0.3	0.0	0.6	0.1	0.2	5.5
Sum of OxidesEmerson	95.4	0.2	3.5	90.0	95.1	104.2
Naukluft	96.1	0.2	3.6	90.0	95.6	104.8
Greenheugh	93.5	0.2	2.8	90.0	92.8	102.2

Cluster	SiO ₂ +/- std	TiO ₂ +/- Std	Al ₂ O ₃ +/- std	FeO _T +/- std	MgO +/- std	CaO +/- std	Na ₂ O +/- std	K ₂ O +/- std	MnO +/- std
1	43.9 0.7 2.8	0.8 0.0 0.1	6.9 0.4 1.7	21.4 0.7 3.0	14.7 1.1 4.7	3.6 0.4 1.6	2.3 0.1 0.6	0.3 0.1 0.2	0.7 0.2 0.7
2	43.9 0.1 1.8	1.0 0.0 0.2	9.7 0.1 1.8	20.6 0.1 2.1	8.6 0.1 2.4	5.6 0.1 1.8	2.7 0.0 0.4	0.4 0.0 0.2	0.2 0.0 0.0
3	48.5 0.2 2.8	0.9 0.0 0.2	13.1 0.1 2.5	18.7 0.1 2.0	6.5 0.1 1.9	6.0 0.1 1.8	3.4 0.0 0.7	0.7 0.0 0.4	0.1 0.0 0.1
4	54.5 0.8 3.2	0.9 0.1 0.2	18.0 0.6 2.4	9.8 1.0 3.7	2.5 0.3 1.1	5.7 0.7 2.6	5.7 0.3 1.3	1.0 0.2 0.6	0.1 0.0 0.0
5	42.8 1.2 2.6	0.8 0.0 0.1	10.3 1.4 3.2	21.7 1.1 2.4	8.8 1.9 4.2	4.0 0.9 2.0	3.3 0.3 0.7	1.0 0.3 0.6	2.9 0.2 0.4