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► **To cite this version:**

N. Mangold, E. Dehouck, C Fedo, O. Forni, C Achilles, et al.. Chemical alteration of fine-grained sedimentary rocks at Gale crater. *Icarus*, 2019, 321, pp.619-631. 10.1016/j.icarus.2018.11.004 . hal-02342460

HAL Id: hal-02342460

<https://hal.science/hal-02342460>

Submitted on 31 Oct 2019

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1 **Chemical alteration of fine-grained sedimentary rocks at Gale crater**

2

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22 *Abstract*

23 From Sol 750 to 1550, the Curiosity rover documented >100 m thick stack of fine-grained
24 sedimentary rocks making up part of the Murray formation, at the base of Mt Sharp, Gale crater.
25 Here, we use data collected by the ChemCam instrument to estimate the level of chemical
26 weathering in these sedimentary rocks. Both the Chemical Index of Alteration (CIA) and the
27 Weathering Index Scale (WIS) indicate a progressive increase in alteration up section, reaching
28 values of CIA of 63 and WIS of 25%. The increase in CIA and WIS values is coupled with a decrease
29 in calcium abundance, suggesting partial dissolution of Ca-bearing minerals (clinopyroxene and
30 plagioclase). Mineralogy from the CheMin X-ray diffraction instrument indicates a decrease in
31 mafic minerals compared with previously analyzed strata and a significant proportion of
32 phyllosilicates consistent with this interpretation. These observations suggest that the sediments
33 were predominantly altered in an open system, before or during their emplacement, contrasting
34 with the rock-dominated conditions inferred in sedimentary deposits analyzed at Yellowknife Bay.

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36 **1. Introduction**

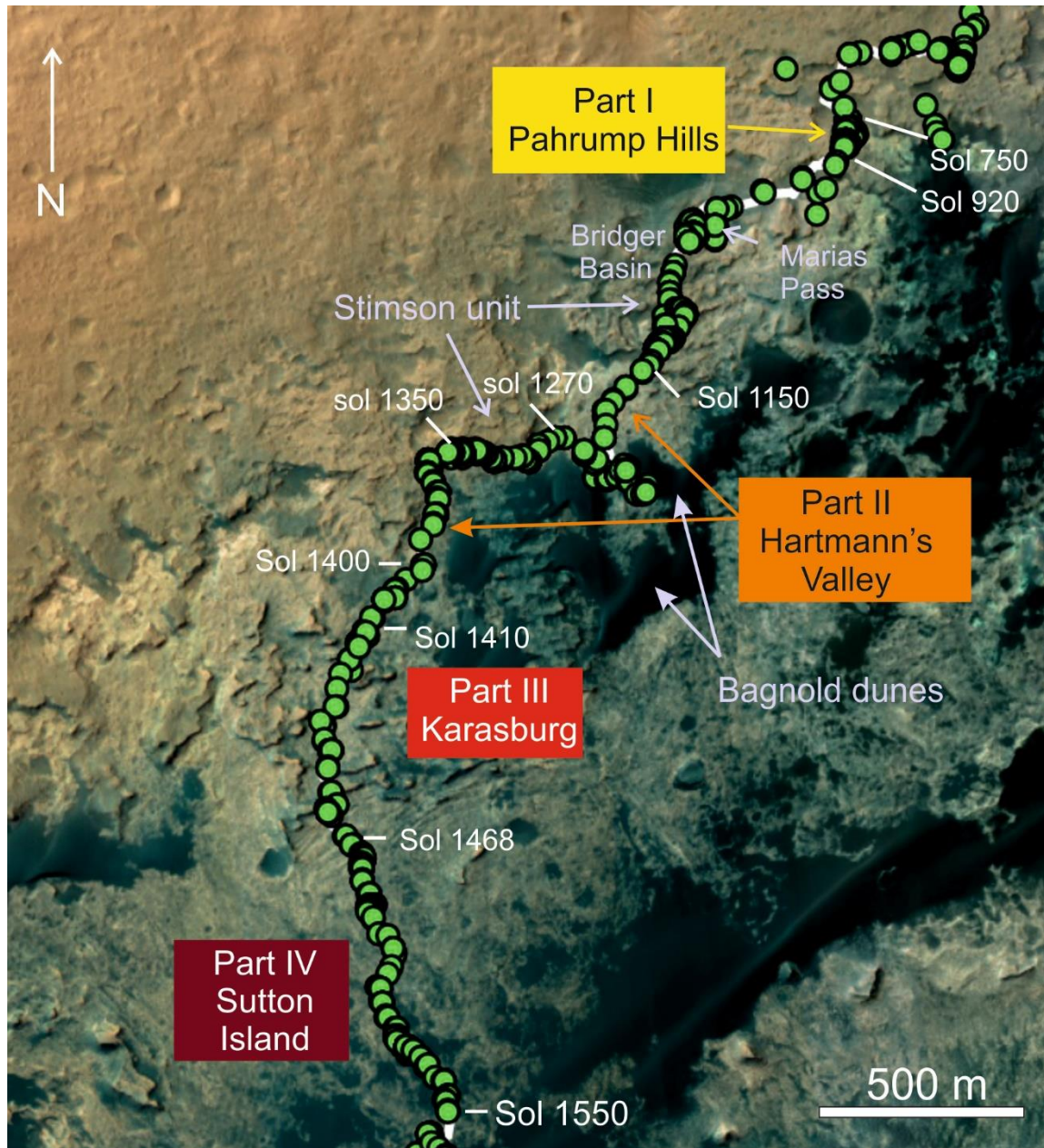
37 Gale crater was formed in the ancient crust of Mars close to the topographic dichotomy boundary
38 ca. 3.6-3.7 Ga, i.e. near the Late Noachian-Early Hesperian boundary (Anderson and Bell, 2010, Le
39 Deit et al., 2013). The site was selected as a landing site for the Mars Science Laboratory (MSL)
40 Curiosity rover for its ~5 km-high central mound composed of sedimentary deposits, where clay
41 and sulfate mineral signatures were detected by orbital spectrometry in the lower units (Milliken
42 et al., 2010). Although the Curiosity rover has not yet accessed the orbitally-detected clay
43 minerals, previous analyses from the CheMin X-Ray diffractometer (XRD) showed that clay
44 minerals were present in the lacustrine mudstones at Yellowknife Bay (Vaniman et al., 2014,
45 Bristow et al., 2015). These rocks are part of a succession which also includes fluvial sandstones
46 and conglomerates (Grotzinger et al., 2014). After 750 Sols (martian days) and 9-km of traverse,
47 clay minerals were detected again by CheMin in the sedimentary rocks of the Murray formation,
48 the basal unit of the Mt Sharp group, informally named the Pahrump Hills member (Rampe et al.,
49 2017, Bristow et al., 2017).

50 At Pahrump Hills, variations in chemistry and mineralogy over a ~10-meter high sequence
51 indicated transitions from hematite-dominated to magnetite-dominated mineralogical
52 assemblages interpreted as due either to diagenetic episodes from multiple influxes of
53 groundwater (Rampe et al., 2017) or variations in pH and Eh of the lake waters (Hurowitz et al.,
54 2017). From Sol 750 to Sol 1550, the rover traversed six more km of various terrains, dominated
55 by the Murray formation totaling ~130 m of stratigraphy, enabling a more in-depth assessment
56 of this sedimentary unit (Fig. 1).

57 Our study focuses on the chemical indices of alteration through the whole column of Murray
58 sedimentary rocks in the studied sol range (750-1550). Data collected by the ChemCam
59 instrument suite are used to assess the chemistry of Murray rocks. The ChemCam Laser Induced
60 Breakdown Spectrometer (LIBS) provides elemental analyses of rocks and soils from their ablation
61 by a pulsed laser, and the Remote Micro-Imager (RMI) provides images for context and textural
62 analysis (Wiens et al., 2012, Maurice et al., 2012). CheMin X-ray diffraction data (Blake et al.,
63 2012) are used to assess locally the mineralogy of the Murray formation in order to better
64 understand the type of aqueous alteration observed on these sedimentary rocks. Results of both
65 instruments are consistent, showing that significant aqueous alteration took place at Gale crater.
66 We discuss further the origin and significance of this alteration.

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70 **Fig. 1:** (a) Context orbital image of the Curiosity traverse from the Bradbury Landing Site to sol 1600. (b)

71 High Resolution Imaging Science Experiment (HiRISE) color image of Gale crater along the traverse of the

72 rover showing the areas of Murray formation outcrops analyzed in this study. The Murray formation was

73 divided in four members: (1) Pahrump Hills (sols 750-920); (2) Hartmann's Valley (sols 1170-1250 and sols

74 1350-1400); (3) Karasburg (sols 1410-1467); (4) Sutton Island (sols 1468-1550). Green disks indicate

75 locations of ChemCam analyses along the rover traverse.

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79 **2. Dataset**

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81 *2.1 Chemcam data*

82 The ChemCam instrument suite consists of a LIBS coupled with the RMI which provides images
83 for context and textural analysis (Wiens et al., 2012, Maurice et al., 2012). The LIBS technique
84 uses a pulsed laser to ablate the rocks, producing a plasma. The atomic emission spectrum of the
85 plasma is analyzed by three spectrometers in the spectral ranges of the ultra-violet (240–342 nm),
86 visible violet (382–469 nm), and visible/near-infrared (474–906 nm). By using ChemCam LIBS
87 spectra, the emission lines of all major elements (Si, Al, K, Na, Ca, Mg, Fe, Ti), as well as several
88 minor and trace elements (Li, Mn, Cr, Zn, Ni, Sr, Rb, Ba, Cu) can be identified (e.g., Ollila et al.,
89 2014). Volatiles and halogens (O, S, P, H, Cl, F) can also be identified if present in sufficient
90 proportions (i.e., >1-10% depending on elements, Meslin et al., 2013, Forni et al., 2015). Carbon
91 is detected in all targets analyzed and corresponds to the contribution of atmospheric carbon
92 dioxide (Meslin et al., 2013).

93

94 Quantification of major elements is achieved by multivariate analyses using a range of emission
95 lines and comparing them to 450 standards analyzed in a testbed (Clegg et al., 2017). The
96 ChemCam laser beam focuses to a small spot, typically ~350-550 μm in diameter (medium sand
97 in size) at distances of 2–5 m. LIBS data are usually collected in lines of 5 to 20 points or matrices
98 of 3 by 3 to 5 by 5 points. A series of 30 laser shots (or more to investigate the rock deeper) are
99 used to collect spectra at a given point (Fig. 2). However, to avoid dust contamination, the first 5
100 spectra are removed before processing for quantification, and the last 25 spectra are averaged to
101 give the mean chemistry of this point (Fig. 2).

102

103 In the case of large grains, such as phenocrysts or diagenetic concretions, the small footprint of
104 the individual LIBS observations allows individual mineral chemistry to be obtained (e.g., Nachon
105 et al., 2014, Sautter et al., 2015, Rapin et al., 2016). In contrast, in the case of
106 mudstones/siltstones (<62 μm) and fine-grained sandstones (<250 μm), each point provides the
107 average chemical composition of multiple grains (Mangold et al., 2017). A single point therefore
108 provides a composition representative of the bulk chemistry. Here, the composition is averaged
109 across all points in a given target (with a minimum of four points), further increasing the
110 representativeness of the calculated bulk chemistry. In addition, this approach allows us not to
111 bias the dataset with targets over which many points have been analyzed compared to targets
112 with fewer points (e.g., Figure 2, Tab. 1).

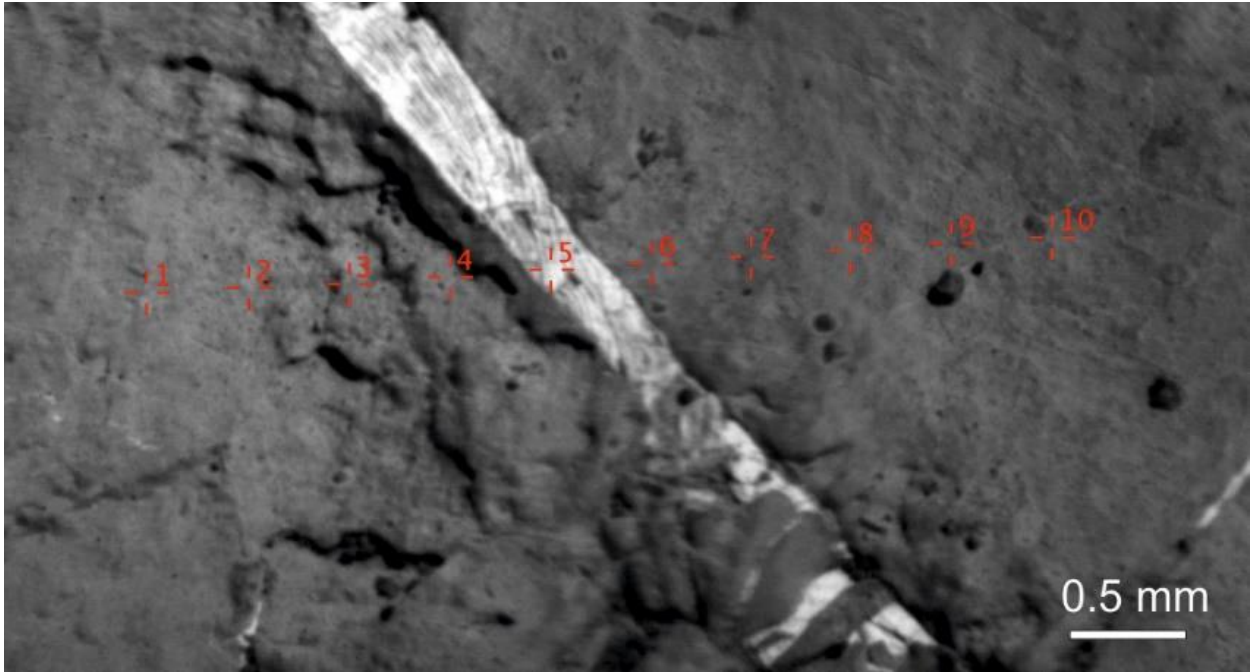
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114 This approach was previously applied to the mudstones and sandstones of Yellowknife Bay, where
115 low deviations around mean values were observed over >500 analysis points collected (Mangold
116 et al., 2015). At that location, an analysis of laser ablation cavities was done for both sandstones
117 and mudstones (Arvidson et al., 2014). Results show that mudstones pits range from 0.2 to 0.6
118 mm, well above the grain size in these fine-grained rocks (<62 μm). Thus, each analysis point can
119 be used as a bulk rock analysis for mudstones. Pits in sandstones are often larger, from 0.4 to 0.8
120 mm in the sandstones analyzed at Yellowknife Bay (Arvidson et al., 2014). At this scale, the laser
121 spot on fine-grained to medium sandstones (<0.5 mm) still enable good average estimations,
122 explaining for instance that the Gillespie lake sandstones did not show significantly larger
123 standard deviations than the Sheepbed mudstones (Mangold et al., 2015). Over the 205 targets
124 studied hereafter, only 5 correspond to medium to coarse sandstones, while the other 200
125 correspond to mudstones and fine-grained sandstones for which the average of four points is
126 sufficient to provide a representative bulk chemistry.

127
128 A strength of LIBS is its ability to exclude points that probed diagenetic features instead of the
129 bedrock itself. All points on resolvable diagenetic features have been removed by a systematic
130 cross-analysis of RMI images, as, for example, point 5 in figure 2. The chemistry corresponding to
131 point 5 (Tab. 1) displays a low total wt.% of oxides because this point is enriched in sulfur, which
132 is an element that is difficult to detect, and so, it is not taken into account in the sum of major
133 element oxides like most volatiles (H, C, P, etc.). The majority of points with low totals were
134 removed after the RMI analyses. Many individual points have still low totals that we interpret as
135 diagenetic features too small for visual determination. There is no definitive method to define the
136 threshold (in total oxides wt.%) between the presence and absence of volatiles due to diagenetic
137 phases, because the bedrock itself can display various abundances of volatiles (such as hydrogen
138 inside clays, etc.). We choose here to use the maximum total of the major element wt.% oxides
139 observed for targets for which diagenetic features were actually identified on images, found as
140 ~90 wt.% in a couple of examples. While some amount of volatiles may still be present in targets
141 with major element totals above 90 wt. %, this technique enables us to minimize the effect of
142 diagenetic features enriched in volatiles, and thus provides a composition as close as possible to
143 the bulk rock unaffected by diagenetic episodes.

144 By applying these criteria, 1660 points belonging to 205 different targets were retained in our
145 analysis. The 205 bulk compositions were then normalized to total weight oxides of 100% to
146 obtain volatile-free compositions (Tab. S1), which are useful for comparisons with other datasets,
147 e.g., Mars average crust composition (Tab. S2). Note that this normalization has no consequence
148 on the ternary diagrams shown later, which plots elements relative to the others.

149

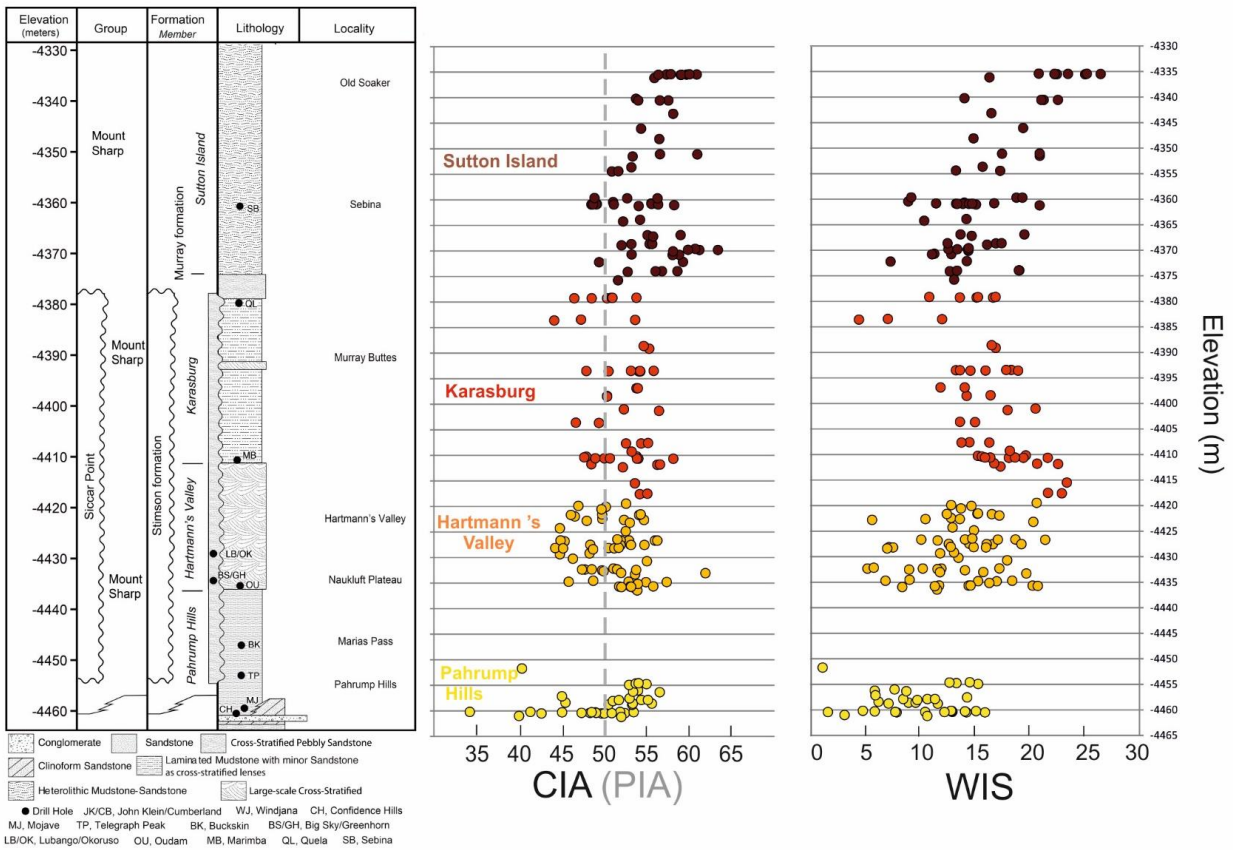


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152 **Fig. 2:** Example of a RMI image of a Murray formation target (Upper Hadlock, sol 1505) crossed by a light-
153 toned vein sampled on point 5. Table 1 displays the chemical results of this target.

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Fig. 3: (a) Stratigraphic column of the Murray formation highlighting the four areas of Murray used in this study. The gap above Pahrump Hills is related to the predominance of Si-rich outcrops of Murray formation and of the overlying Stimson formation. The stratigraphic framework shown here was established and refined through the efforts of the MSL sedimentology/stratigraphy working group (Fedó et al., 2017). (b) CIA plotted in filled circles along the stratigraphy for the four areas studied. PIA values are plotted next to CIA values in open circles. (c) WIS along the stratigraphy. WIS values calculated for starting composition corresponding to the average Mars crust. Starting from the average of Gale crater conglomerates would translate the diagram by 6% to the left.

173 2.2 *CheMin data*

174
175 CheMin collects X-Ray Diffraction (XRD) data using Co K α radiation in transmission geometry
176 (Blake et al., 2012). More details of instrument operation and sample analysis conditions can be
177 found in in Bristow et al. (2018). Abundances of crystalline phases were determined by Rietveld
178 analysis using Jade software. The results presented here are products of modeling that
179 incorporate measured standards of clay minerals in addition to structures of crystalline phases
180 (see Bristow et al., 2018 for more details). The abundances of amorphous components and poorly
181 crystalline clay minerals are determined using the program FULLPAT (Chipera and Bish, 2002).

182 **3. Geologic context and stratigraphy**

183
184 From landing to sol 750, the traversed terrains are associated with the Bradbury group (Fig. 1).
185 This formation consists of fluvial sandstones and conglomerates with local exposures of lacustrine
186 mudstones. At Yellowknife Bay, the Sheepbed member of the Bradbury group corresponds to
187 mudstones with basaltic chemical composition that are overlain by coarse sandstones divided
188 into three members with basaltic composition as well (McLennan et al., 2014, Mangold et al.,
189 2015, Anderson et al., 2015). At Kimberley, sedimentary rocks are potassic (up to 5 wt.% of K₂O)
190 and consist of fine-grained sandstones and mudstones overlying conglomerates (Le Deit et al.,
191 2017). In the Bradbury group, rocks are diverse and heterogeneous in grain size, in agreement
192 with rocks derived mainly from alluvial/deltaic environments (Grotzinger et al., 2015), but
193 relationships with the Mt. Sharp sequence are unclear due to the predominance of regolith and
194 lack of contacts. Starting sol 750, at Pahrump Hills, rocks correspond to well-defined outcrops at
195 the base of Mount Sharp that are exposed nearly continuously from there onto Mount Sharp on
196 the orbital images, and have been named the Murray formation. As a consequence of this
197 continuity and near-horizontal bedding, the stratigraphic section is described with elevation as a
198 proxy for geologic time, allowing us to localize targets analyzed along the section (Fig. 3).

199
200 For the purpose of this study, the Murray formation was divided into four members from their
201 geographic location and textural analysis (Fig. 1, Fedo et al., 2017). Part I of our study corresponds
202 to the base of the Murray formation that was analyzed at the location named Pahrump Hills, a
203 ~10-m high outcrop (sols 750-920, area I on Figs. 1 and 3) composed of interbedded lacustrine
204 laminated mudstones and fluvial fine- to coarse-grained sandstones with local cross-bedding
205 (Grotzinger et al., 2015). This location was analyzed with 36 ChemCam bedrock targets and 3 drill
206 samples analyzed by CheMin (Confidence Hills, Mojave, Telegraph Peak, Figure 3).

207
208 After Pahrump Hills, the rover crossed hilly terrains of Marias Pass (sols 990-1050) and Bridger
209 Basin (sols 1050-1150). At these locations, ChemCam analyses of the Murray formation were

210 discontinuous because the Murray formation was partially covered by lags and unconformably
211 overlain by eolian cross-bedded fine-grained sandstones of the Stimson formation (Banham et al.,
212 2016, 2018). Eolian sandstones from the Stimson formation formed well after the Murray
213 formation (Banham et al., 2018), and are not included in this work for this reason. Outcrops of
214 the Murray formation in these areas are locally enriched in silica (>60 wt.% SiO₂). XRD showed
215 the presence of various phases of silica, including tridymite, which were interpreted as detrital
216 components of silicic volcanic rocks in the drill target Buckskin (Morris et al., 2016). Light-toned
217 halos (enriched in SiO₂ up to 80 wt.%) are frequently observed, suggesting a diagenetic episode
218 due to Si-rich fluid circulation and related cementation (Frydevang et al., 2017). To avoid the bias
219 that would be introduced by these diagenetically imprinted rocks on the weathering indices, we
220 chose to exclude these few outcrops of the Murray formation from our study. This explains the
221 gap in observation points between elevations -4450 and -4437 m in Fig. 3. Neither ChemCam
222 data, nor CheMin mineralogy data of the Murray formation strata above this area indicate any
223 further silica enrichment in bedrock apart from diagenetic halos (Bristow et al., 2018).

224 Part II of Murray formation (Fig. 2) is referred to as Hartmann's Valley member hereafter. The
225 Hartmann's Valley member includes two areas at similar elevation (sols 1150-1270 and sols 1350-
226 1400) separated by the Naukluft plateau composed of the overlying Stimson formation. Rocks in
227 these two areas of Murray formation have a similar texture, with silt to very fine-sand sized grains,
228 and local meter-scale cross-beds, interpreted as potential fluvial and eolian deposits (Fedo et al.,
229 2017) (Fig. 3). The Hartmann's Valley member was analyzed using 63 ChemCam targets and 1 drill
230 sample delivered to CheMin (Oudam, Figure 3).

231 Part III of our study is the Karasburg member (sols 1410-1468) which starts approximately at the
232 southern edge of the plateau composed of the Stimson unit in a location with remaining residual
233 buttes of the Stimson formation discontinuously capping the Murray formation (at a location
234 named Murray buttes). The Karasburg member is composed of regularly mm-scale laminated
235 mudstones interpreted as lacustrine deposits (Fedo et al., 2017). 46 ChemCam targets of the
236 Karasburg member were analyzed as well as 2 drill holes (Marimba and Quela, Fig. 3).

237 Part IV of our study is the Sutton Island member (sols 1468-1550). Targets of this member are
238 located in terrains with gentle slopes going upward, along Mt Sharp ascent route. In this area, the
239 Murray formation consists of more heterolithic mudstones and sandstones (Figs. 1 and 3).
240 Interpretations of the facies suggest that rocks were deposited in lacustrine and lacustrine-margin
241 environments (Fedo et al., 2017) with evidence in the form of desiccation cracks indicating
242 episodes of at least partial drying out (Stein et al., 2018). The Sutton Island member was analyzed
243 using 60 ChemCam targets and 1 drill hole (Sebina, Fig. 3). A drill attempt, at Precipice, close to
244 the desiccation cracks, was unsuccessful due to a mechanical fault.

245 A detailed description of stratigraphy and sedimentary facies is not the aim of this study. Further
246 work on the facies will be the subject of detailed articles in the near-future. Nevertheless, we
247 have analyzed the texture of targets using RMI images. A large majority of targets analyzed for
248 this study are fine-grained sediments, with few or no grains visible at the usual scale of RMI
249 images, i.e. 120-300 μm resolution for 3 pixel features, at 2 to 5 m distance (see Tab. 1 in Mangold
250 et al., 2017). One exception is a horizon in the Pahrump Hills member where medium to coarse
251 sandstones are observed in 5 targets. Independent, more in-depth assessments of the
252 stratigraphy also point to the predominance of fine-grained sediments in Murray formation,
253 either mudstones or fine-grained sandstones (Fedo et al., 2017, Rivera-Hernandez et al., 2018).

254 255 **4. Chemical weathering indices**

256 257 *4.1 The Chemical Index of Alteration (CIA)*

258
259 On Earth, many studies have attempted to estimate the degree of weathering based on chemical
260 compositions of soils and sedimentary rocks. Weathering indices are based on the differential
261 mobility of elements during alteration processes. Therefore, they reflect open-system weathering
262 rather than isochemical alteration (Parker, 1970, Nesbitt and Young, 1982, Fedo et al., 1995,
263 Meunier et al., 2013).

264
265 The Chemical Index of Alteration (CIA, see Methods) is the most commonly used index (Nesbitt
266 and Young, 1982). It is based on the leaching of Ca, Na and K relative to Al. Al is an immobile major
267 element in supergene alteration, thus taken as a reference. The CIA is calculated using the
268 following relationship: $\text{CIA} = [\text{Al}_2\text{O}_3 / (\text{Al}_2\text{O}_3 + \text{Na}_2\text{O} + \text{CaO}^* + \text{K}_2\text{O})] \times 100$, calculated in molar
269 proportions. CaO^* corresponds to CaO from silicates excluding CaO from carbonates, sulfates and
270 phosphates. The filtering procedure explained in section 2 enables us to minimize the effect of
271 these volatile-rich phases such as sulfates and phosphates, if present. Still, this correction is
272 conservative, which means that our value of CaO is higher than the true CaO^* . As a result, and
273 because CaO is in the denominator of the equation, our calculated CIA values is underestimated
274 compared to the actual CIA of each target.

275
276 In addition to the CIA, the Plagioclase Index of Alteration (PIA) (Fedo et al., 1995) is also calculated:
277 $\text{PIA} = (\text{Al}_2\text{O}_3 - \text{K}_2\text{O}) / (\text{Al}_2\text{O}_3 + \text{CaO}^* + \text{Na}_2\text{O} - \text{K}_2\text{O})$. This index helps to remove the effect of potential K-
278 enrichment during burial and diagenesis (Fedo et al., 1995). CIA and PIA indices are represented
279 in Fig. 3. PIA values are slightly higher than CIA values at Sutton Island by one or two CIA units at
280 maximum, but overall PIA values are close to the CIA values of the corresponding targets. Thus,
281 we focus most of the description on CIA values hereafter.

282

283 The point-to-point variations in these figures represents variations from one bedrock target to
284 another bedrock target (thus from compositions averaged across an entire raster and not from
285 one analysis point to another analysis point). In both indices, igneous rocks have CIA from 35
286 (ultramafic) to 50 (granite), and fully weathered rocks reach 100. A strength of the CIA is that it
287 can be plotted as a vertical axis on a A–CN–K ternary diagram (Fig. 4a). This diagram is useful for
288 comparing parent rocks and their weathering products because unaltered rocks and primary
289 minerals plot with $CIA \leq 50$ below the plagioclase-K-feldspar join (50-50 line in Fig. 4a), and
290 weathering products plot on a systematic trend predicted by thermodynamic and kinetic
291 considerations. The A-CNK-FM diagram, where the alteration line joins the middle of the A-CNK
292 line to the FM end-member, is provided for comparison (Fig. 4b).

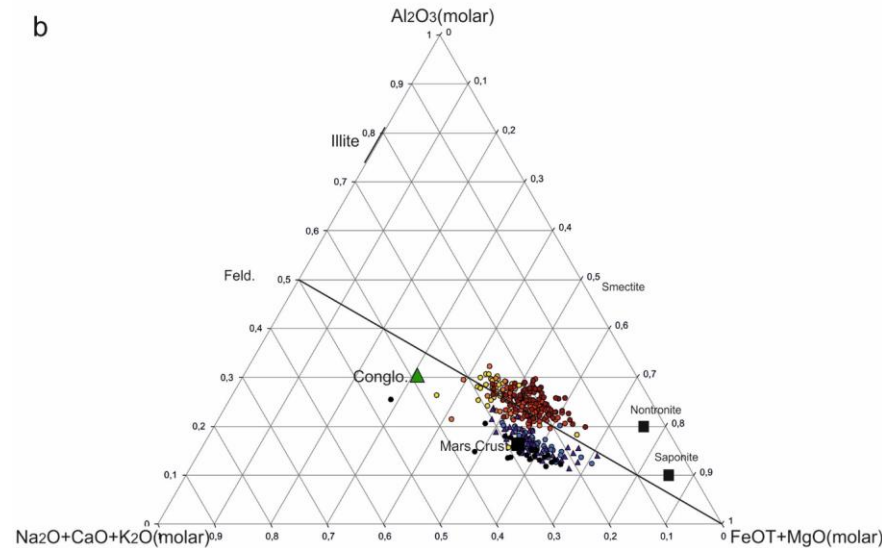
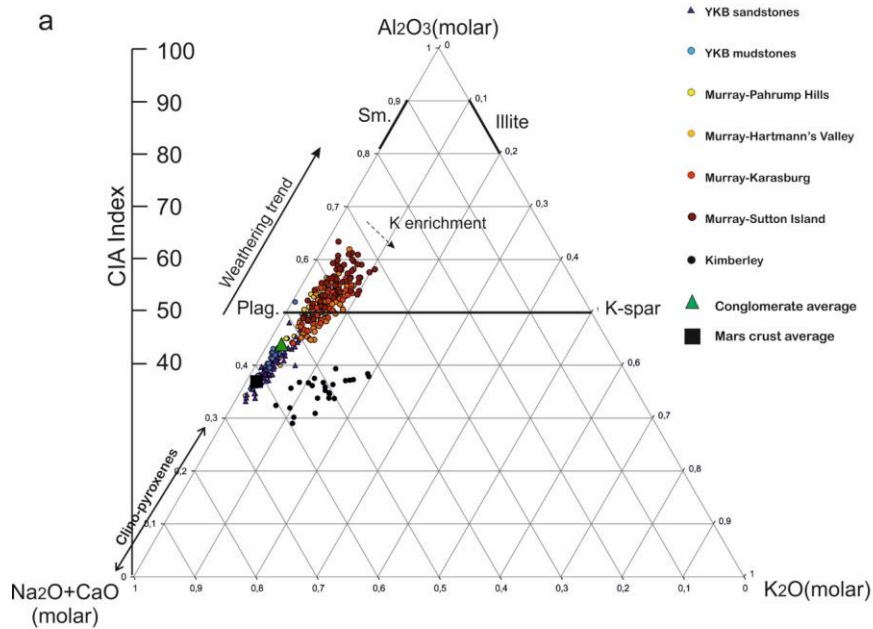
293
294 Figure 4 displays two possible average source rocks for the sediments in the rocks of the Murray
295 formation: (i) the widely-used average Mars crust composition (Taylor and McLennan, 2009) (Tab.
296 S2) with a basaltic composition and a CIA of ~ 38 , and (ii) the average composition of
297 conglomerates analyzed by ChemCam at Gale crater, representing the least altered component
298 of the sedimentary rocks analyzed at Gale crater (Mangold et al., 2016). Conglomerates are more
299 felsic than Mars average crustal composition, explaining the higher CIA of ~ 45 . At Yellowknife Bay,
300 lacustrine mudstones (Sheepbed member) and fluvial sandstones have CIA values between 35
301 and 45 close to these two potential source rocks, a result that has been interpreted as a lack of
302 open-system weathering (McLennan et al., 2014). In contrast, the CIA values of most Murray rocks
303 plot well above 45 for all outcrops (Figs. 3 and 4).

304
305 CIA values display variations between the four areas considered (Fig. 3). The lowermost Pahrump
306 Hills member appears slightly altered with an average of 49.8 ± 5.1 , in good agreement with values
307 of ~ 50 reported there using APXS data (Hurowitz et al., 2017). The lowermost CIA values
308 correspond to the group of coarser sandstones identified in this area. Hartmann's Valley member
309 has CIA values similar to Pahrump Hills member (except one point at 61), with an average of
310 51.0 ± 3.7 .

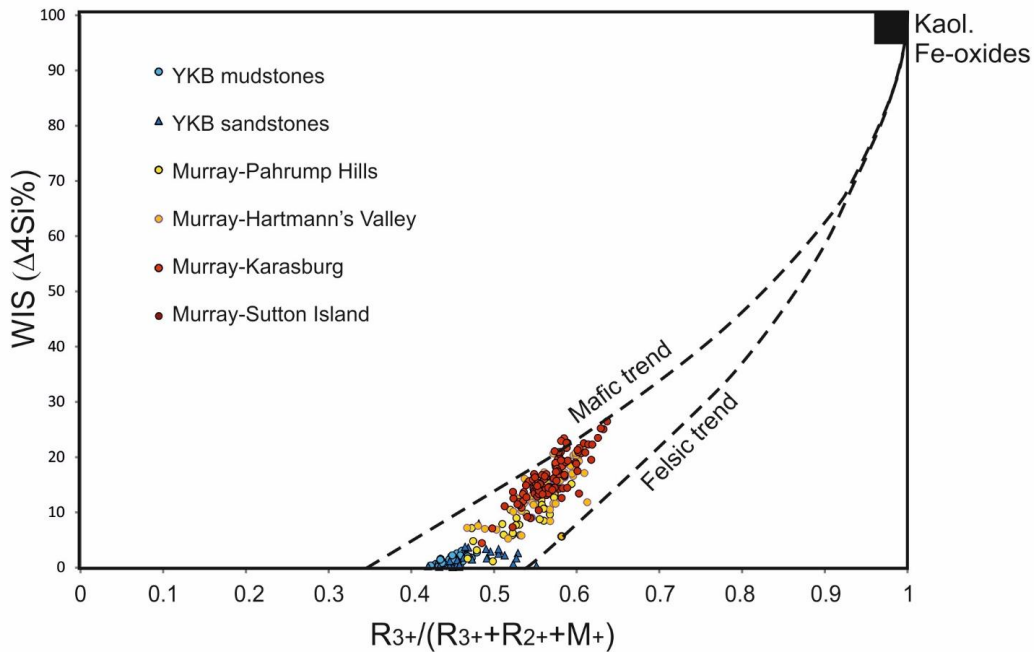
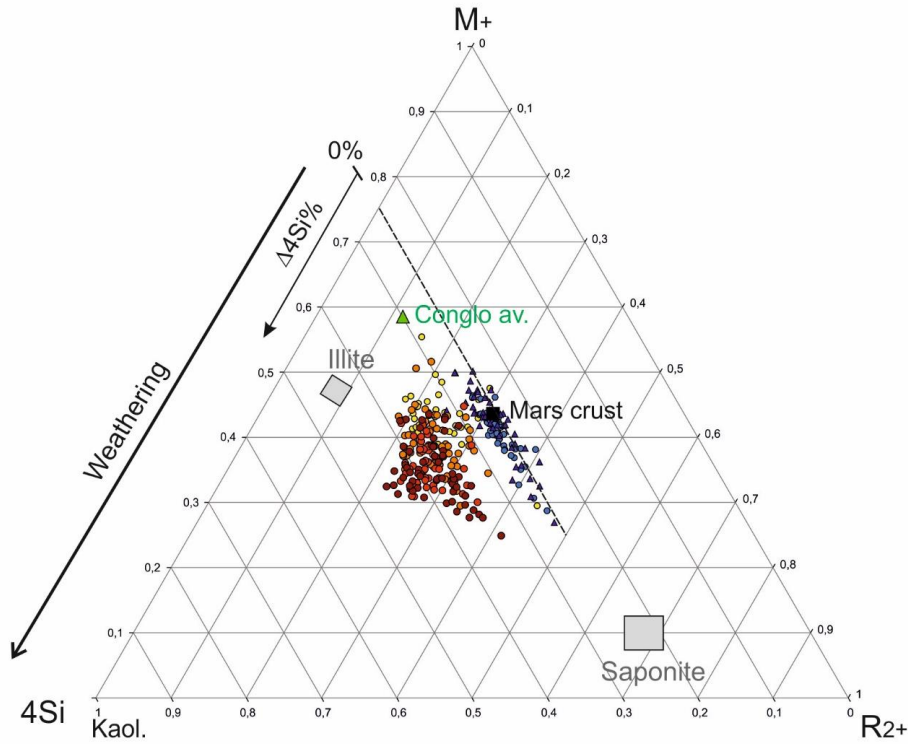
311
312 The Karasburg member displays CIA values that are predominantly >50 , with an average of
313 52.1 ± 3.2 , reaching up to 57. A difference with the underlying group is the much lower amount of
314 values below 50. The Sutton Island member reports the highest group of CIA values with an
315 average of 55.5 ± 3.6 and several targets above 60, reaching up to 63. Overall, CIA values are
316 increasing going up stratigraphy, and are above 50 for a large majority of the targets of the two
317 upper members. The fact that a majority of points are above a CIA of 50 suggests that significant
318 aqueous alteration took place.

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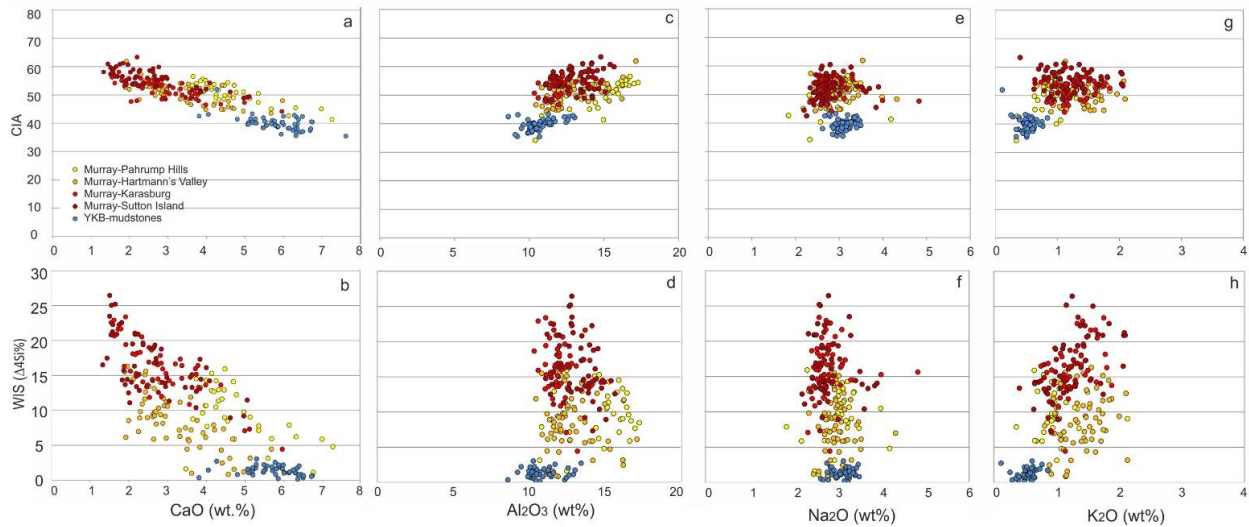


322
323 **Fig. 4:** (a) A-CN-K ternary diagram (molar values) with CIA values plotted on the vertical axis. Targets display
324 many points above the 50-50 join, suggesting a limited contribution of primary minerals. Mars crust (black
325 square) and average of Gale crater conglomerate (green triangle) represent potential source rock
326 compositions with CIA values typical of unaltered rocks. Note that the drift towards higher potassium of
327 Murray targets is correlated with higher CIA values. (b) Ternary molar A-CN-K-FM diagram of the different
328 sedimentary rocks of Gale crater. The line joining FM to the middle of the A-CN-K join indicates the
329 boundary between altered and non-altered rocks, the latter being below the line. As in Fig. 4a most Murray
330 rocks plot above the alteration line though the evolution shown does not provide a clear trend suggesting
331 a mixing of alteration phases.



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335

336 **Fig. 5:** (a) Ternary diagram of $M^+/R^{2+}/4Si$ showing the difference between potential source rocks
337 composition and actual Murray formation composition to obtain the values of the WIS (difference of $\Delta 4Si$
338 in %. see Methods). (b) Diagram showing $R^{3+}/(R^{3+}+R^{2+}+M^+)$ vs $\Delta 4Si\%$ compared to the empirical trends
339 observed for terrestrial mafic and felsic rocks.



340
 341
 342 **Fig. 6** : (a) Diagram plotting CIA vs CaO abundance showing a correlation between high CIA and low CaO.
 343 (b) Diagram plotting WIS and CaO abundance showing a similar relationship. (c) and (d) Same diagrams
 344 with Al₂O₃. (e) and (f) Same diagrams with Na₂O. (g) and (h) Same diagrams with K₂O. No obvious variations
 345 are observed with increasing weathering indices compared to that observed for CaO.

346
 347
 348
 349

350 4.2 The Weathering Intensity Scale (WIS)

351
 352 Most other weathering indices have usually good correlations with the CIA (Yang et al., 2006).
 353 However, the CIA does not take into account Si variations. For this reason, we have also used the
 354 Weathering Intensity Scale (WIS) (Meunier et al., 2013). The main components of the WIS are (1)
 355 $4Si$, which is the molar proportion of Si⁴⁺ divided by 4 to refer to the formula of phyllosilicates; (2)
 356 M^+ , which is the sum of the molar proportion of alkali and earth alkali elements ($Na^+ + K^+ + 2Ca^{2+}$);
 357 (3) R^{2+} , which groups the bivalent elements ($Mg^{2+} + Fe^{2+} + Mn^{2+}$), and (4) R^{3+} which groups the
 358 trivalent cations ($Al^{3+} + Fe^{3+}$). This method enables to sort the mobile elements (M^+ and R^{2+}) from
 359 the elements stable in soils (R^{3+}). For the R^{2+} , we neglect the effect of Mn²⁺ because it is not
 360 systematically taken into account in the quantification of major elements by ChemCam. High Mn
 361 values (>3%) have been observed locally on Mars in fracture fills (Lanza et al., 2014), but average
 362 values are ca. 0.4% corresponding to a minor shift from the much larger contribution of Fe and
 363 Mg (totaling >25 wt.% for most targets).

364 The M^+ , $4Si$ and R^{2+} amounts are first normalized to 100% (see Meunier et al., 2013) and plotted
 365 in a ternary diagram (Fig. 5a). The $\Delta 4Si\%$ (referred to as WIS for simplicity) is then a parameter

366 varying from 0 to 100% measured by the difference between the compositions of unweathered
367 and weathered samples : $\Delta 4Si\% = ((4Si_{\text{altered rock}} - 4Si_{\text{unaltered parent rock}}) * 100) / (100 - 4Si_{\text{unaltered parent rock}})$.
368 It can be visualized on the ternary diagram by the distance from the reference unaltered source
369 rock (Fig. 5a). Any index aiming to measure weathering must also take into account the
370 progressive concentration of trivalent chemical components (R^{3+}) (Meunier et al., 2013). The
371 second diagram (Fig. 5b) plots the $\Delta 4Si\%$ obtained versus the increase in trivalent cations
372 calculated from the ratio $R^{3+} / (R^{3+} + R^{2+} + M^+)$.

373
374 The R^{2+} and R^{3+} in the WIS calculation requires an estimation of the Fe^{2+}/Fe^{3+} ratio, which is not
375 directly accessible from ChemCam data. It was recognized as a weakness of the method (Meunier
376 et al., 2013). CheMin XRD data have been used locally to estimate this ratio in the Murray
377 mudstones, at the drill hole Marimba (Tab. 2). Result shows a strong predominance of the ferric
378 oxides with Fe^{2+}/Fe^{3+} (molar) = $0.00301/0.0509 = 0.059$. Given large assumptions on the mineralogy
379 of ferrous minerals and the fact that the amorphous component cannot be determined by this
380 method, we have used a conservative value of Fe^{2+}/Fe^{3+} of 0.1. Comparison of chemistry and
381 mineralogy in previous drill holes has argued for the presence of ferrihydrite in the amorphous
382 component (Dehouck et al., 2017) suggesting a large part of the iron of this component is indeed
383 ferric, so it would not change this ratio significantly. We have also performed tests at various
384 Fe^{2+}/Fe^{3+} ratio to estimate the impact of these variations to the WIS, showing that the difference
385 in $\Delta 4Si\%$ is minor whatever the Fe^{2+}/Fe^{3+} ratio taken (Fig. S1).

386
387 The ternary diagram using the three first components $M^+ - 4Si - R^{2+}$ shows that the targets from the
388 Karasburg and Sutton Island members plot away from the two potential source rocks yielding WIS
389 values of up to 26% at Sutton Island using Mars crust composition as the initial source, or 20%
390 taking the conglomerate average composition as the source. These results are consistent with
391 those using the CIA although the WIS calculation includes SiO_2 . The diagram plotting the WIS
392 versus the trivalent cations shows a progressive increase similar to the empirical trends observed
393 for terrestrial felsic and mafic rocks. The trends are significant in both diagrams although the
394 alteration is relatively limited and far from reaching the kaolinite end-member. In contrast,
395 Bradbury group mudstones (Sheepbed member) do not show any substantial increase in the WIS
396 (Fig. 5), in agreement with their low CIA (Fig. 4). For simplicity, we have used the same
397 Fe^{2+}/Fe^{3+} ratio for all targets, although it creates an overestimation of the X-axis of the unaltered
398 targets, which are naturally more ferrous. This assumption explains why the trend provided by all
399 targets is slightly shifted between felsic and mafic from the empirical lines drawn in Fig. 5
400 (bottom).

401

402 *4.3 Chemical variations in relation to alteration indices*

403

404 Leaching of sodium, calcium, or potassium, or a combination of these elements, is needed to
405 explain CIA values greater than 50. The CaO abundance is low in all Murray formation (2-5 wt.%,
406 Tab. S1) compared to average Mars crust (at ~7 wt.%, Tab. S2). Calcium is also the only cation
407 that decreases significantly with higher CIA and WIS (Fig. 6). CaO abundance is as low as 1.5-2.5
408 wt.% for targets with CIA>55, by comparison with rocks displaying CIA<50 that have CaO>4 wt.%
409 (Fig. 6a). CaO shows a progressive decrease from Pahrump Hills (4.8±1.2 wt.% in average, see Tab.
410 S1) to Hartmann's Valley (3.3±1.0 wt.%) then Karasburg (2.8±0.9 wt.%) and Sutton Island (2.6±1.0
411 wt.%). In contrast to CaO, no obvious trend is observed with the other elements used for the CIA
412 calculation (Fig. 6c, 6e, 6g), although K₂O displays a slight increase with CIA. Al₂O₃ displays a
413 variability in abundance that may be related to natural variations in clastic sedimentation
414 processes. These variations do not show a correlation with the highest CIA values, showing that
415 these variations in Al are not the cause of the high CIA values, which is based instead on trends in
416 cation abundances within each rock.

417
418 Similarly, values of WIS>20% correspond to low abundance of CaO (<2.5 wt.%, Fig. 6b), and not
419 of other elements, confirming the trend observed with the CIA with an index less dependent on
420 calcium. A comparison between the CIA and the WIS (Fig. 3) also shows that the Karasburg and
421 Sutton Island members are those where the alteration is the most developed for both alteration
422 indices. The WIS exhibits slightly larger variations, indicating a diversity that could be related to
423 local variations in the intensity of alteration.

424

425

426 **5. Results from mineralogy**

427

428 CheMin analyzed three drill samples at Pahrump Hills (Confidence Hills, Mojave and Telegraph
429 Peak, Rampe et al., 2017), one at Hartmann's Valley (Oudam), and three in the two upper
430 members (Marimba, Quela and Sebina) where both the CIA and WIS are the highest (Fig. 3, Tab.
431 3). In these latter three drill samples, CheMin data indicate the presence of abundant clay
432 minerals (~16 to 28 wt.% of the whole rock) (Tab. 3). Clay minerals in these three samples are
433 interpreted as a mixing of phases such as di-octahedral smectites (with Al³⁺ and Fe³⁺) and tri-
434 octahedral smectites (with Mg²⁺). The presence of these phases is consistent with the enhanced
435 alteration observed from chemistry (Bristow et al., 2018). By comparison, drill samples in the
436 Pahrump Hills and Hartmann's Valley members do not contain large proportions of phyllosilicates
437 (~3%), suggesting a lower imprint of alteration – at this point, either from detrital input or *in situ*
438 alteration, or both -, in agreement with the lower CIA and WIS in these areas.

439 The three uppermost drill samples also display a lower proportion of primary minerals (olivine,
440 pyroxene, plagioclase and alkali feldspar) than the previous drill samples. Plagioclase is the most
441 abundant primary mineral everywhere, but it is present in lower proportion in the three upper

442 drill holes (10 to 14 wt.%) than in the four lower drill ones (20 to 27 wt.%) (Tab. 3). Mafic minerals
443 are in very low proportion in the three upper drill holes, with no olivine and pyroxenes
444 representing only ~0.7 wt.% at Marimba, 2.7 wt.% at Quela and 2.8 wt.% at Sebina, compared to
445 5 to 13 wt.% of pyroxenes (and 1% of olivine) in the four previous drill holes. These trends are still
446 valid when removing the amorphous component from the balance, i.e. the low abundance of
447 primary minerals in the three upper drills is not due to a higher proportion of amorphous
448 component. Thus, CheMin data display an increase in clay mineral abundances going up section
449 where the alteration indices are the highest.

450 6. Discussion

451 6.1 Alteration versus provenance effects

452 The increase in clay minerals, decrease in primary mafic minerals and decrease in CaO abundance
453 are all specific to the two upper members (Karasburg and Sutton Island), which display the highest
454 alteration indices, thus favoring a role of enhanced alteration up section. Before discussing the
455 origin of the potential alteration (sections 6.2 and 6.3), we discuss hereafter potential links
456 between mineralogy and chemistry to understand if these trends are related to provenance or
457 alteration, or both.

460 A potential explanation of the low calcium abundance up section is that this element could have
461 been leached from Ca-bearing minerals, such as plagioclase and clinopyroxene. We can test this
462 hypothesis by comparing the mineralogy and chemistry of the relevant locations. It must be
463 recalled, nevertheless, that drill holes are not numerous, and so, do not enable a statistically
464 representative comparison between chemistry and mineralogy.

465 Plagioclase is the dominant primary mineral in all drill samples, but with lower proportions in the
466 three upper drill holes. We can estimate the variations in CaO abundance linked to these
467 variations of mineralogy, assuming a plagioclase stoichiometry of An₄₀, as estimated among the
468 Pahrump Hills member drill holes (Morrison et al., 2017). The CaO abundance due to 20-27 wt.%
469 of plagioclase observed from Confidence Hills to Oudam corresponds to an abundance of CaO of
470 1.7-2.3 wt.%. By comparison, the 10 to 14 wt.% plagioclase observed in the three upper drill
471 samples represents 0.8-1.2 wt.% of CaO. Thus, net losses of CaO are in the range of 0.5-1.5 wt.%.
472 These values are in agreement with the decrease in CaO abundance recorded by ChemCam, from
473 the Pahrump Hills (4.8 wt.%) to the Sutton Island member (2.6 wt.%), although not sufficient to
474 explain all the CaO loss.

475
476 These differences in composition may be due to variations in provenance rather than alteration.
477 In such a hypothesis, a lower proportion in plagioclase and alkali feldspar in the three upper drill

478 holes should be related to more mafic source rock(s). However, the very low proportion of
479 pyroxene and lack of olivine does not argue for a more mafic source rock there, thus ruling out
480 this possibility. The lower amount of plagioclase could instead be related to a partial dissolution
481 of this mineral as highlighted by the higher CIA values, which is the most sensitive index to this
482 process. Assuming a partial dissolution of plagioclase, this process should be visible by a coupled
483 decrease in sodium, which is not clearly observed (Fig. 6). Nevertheless, plagioclase minerals are
484 known to dissolve incongruently, which means that the ions in the mineral lattice do not dissolve
485 according to their stoichiometry, enabling calcium to be leached preferentially (e.g., Wilson,
486 2004). Note that a lower plagioclase abundance due to provenance effects would lead to a
487 corresponding decrease in sodium.

488
489 A striking observation is that the three upper drill holes display the lowest proportion of mafic
490 minerals ever observed by Curiosity (<2.8wt% of pyroxene, no olivine). A decrease in the
491 proportion of clinopyroxene can also contribute to the decrease in CaO abundance recorded by
492 ChemCam. The pyroxene structure in these drill holes is difficult to determine due to its low
493 abundance (Bristow et al., 2018). So, we cannot provide a proper estimate of its contribution to
494 the CaO abundance. Nevertheless, drill holes of the lower Murray members and of the Bradbury
495 group include pigeonite, augite and some proportion of orthopyroxene (Morrison et al., 2018). In
496 the Pahrump Hills member, the Confidence Hills drill hole contains pigeonite and orthopyroxene
497 (with no Ca) accounting for 14 wt.% of the crystalline phases and augite accounting for 12 wt.%
498 of the crystalline phases (Morrison et al., 2018). Note nevertheless that the augite stoichiometry
499 was not computed and assumed to be similar to the Bradbury group with 18 wt.% CaO (Morrison
500 et al., 2018). Accounting for the amorphous component comprising 35 wt.% at Marimba, the
501 abundance of CaO corresponding to augite would then be ~1.4 wt.%. This value is of the same
502 magnitude than the 0.5-1.5 wt.% of CaO loss from plagioclase for a total loss of 1.9-2.9 wt.%,
503 consistent with the difference of CaO abundance (2.2 wt% in average, see Tab. S1) measured by
504 ChemCam between Pahrump Hills and Sutton Island (Fig. 6a, b).

505 The low proportion of mafic minerals in the drill holes from upper stratigraphic members can be
506 due to provenance or sorting effects instead of dissolution. However, pyroxenes are common
507 minerals of the Mars crust, both in Noachian or younger terrains (e.g., Poulet et al., 2009), and
508 were identified in all previous drill samples, in much higher proportions (>10 wt.%) than in the
509 three drill samples considered (Morrison et al., 2018). In addition, the chemistry does not point
510 towards an increase in felsic or alkali components (K, Na) as it was observed in the initial part of
511 the mission (Sautter et al., 2015, Mangold et al., 2016). Indeed, there is no coupled increase in Si,
512 Al and K, and CheMin indicates that feldspars are not in higher proportion (alkali feldspar
513 abundance is actually lower up section in CheMin analyses, Tab. 3). These observations show that
514 a more felsic parent composition is not a valid explanation for the low proportion of mafic
515 minerals. In addition, sorting of grains in fine-grained sedimentary rocks has been shown to

516 increase the proportion of dense, resistant mafic minerals rather than decrease it (Siebach et al.,
517 2017), a conclusion inconsistent with an effect of sorting in the studied locations. Thus, the low
518 proportion of pyroxene in the three drill holes of Karasburg and Sutton Island members suggests
519 that their partial dissolution contributed to the loss of CaO.

520 Dissolution of pyroxenes would also result in Mg and Fe leaching too. While MgO is present at ~5
521 wt.% abundance in the Karasburg and Sutton Island members (Tab. S1), this value is lower than
522 in sedimentary rocks analyzed previously in the mission (~8 wt.% in the Sheepbed member of the
523 Bradbury formation, at Yellowknife Bay, e.g., McLennan et al., 2014). In addition, there are not
524 enough primary minerals remaining to explain the abundance of MgO: the abundance of 2.8 wt.%
525 pyroxene would translate to <1 wt.% MgO. In contrast, magnesium is present in the tri-octahedral
526 clay minerals (saponite) detected at Marimba, Quela and Sebina (Bristow et al., 2018). It is known
527 on Earth that smectites formed during weathering are able to incorporate structural magnesium
528 in octahedral sites while calcium cannot go into these sites (e.g., Nesbitt and Wilson, 1992), and
529 can only be present as interlayer potentially explaining why calcium shows a net loss. By
530 comparison to Mg, Fe is less mobile and is known to remain in soils during weathering. The high
531 proportion of hematite (~6-7 wt.%) in the three drill holes could correspond to a byproduct of
532 alteration (including diagenesis). In addition, both Fe and Mg are potentially present in the
533 amorphous component as well, in phases such as hisingerite and ferrihydrite as it has been
534 deduced from data earlier in the mission (Dehouck et al., 2014, 2017). From Tab. 2, we can deduce
535 that only ~8 wt.% of FeO_T is taken by crystalline phases identified by CheMin (especially
536 hematite), therefore suggesting that the remaining ~10 wt.% are concentrated in the amorphous
537 component.

538 Possible variations in the composition of the amorphous component from one drill sample to
539 another could have an impact on the chemistry analyzed by ChemCam. For instance, Al-rich
540 phases (such as allophane) could have an impact on the CIA calculation by increasing the Al₂O₃
541 abundance, while both Al and Si could have an impact on the WIS. So far, no Al-rich amorphous
542 component has been identified (Dehouck et al., 2017, Morrison et al., 2018). The comparison
543 between chemistry and mineralogy (Tab. 2) points towards an excess of FeO relative to the low
544 abundance of identified mafic minerals, suggesting an iron-rich amorphous component, as in
545 Dehouck et al., 2017. In this case, variations in the proportion of the amorphous component
546 would have little impact on the CIA calculation. Concerning Si, no opal has been detected in the
547 drill holes of the two upper members (Tab. 3), suggesting that these locations are distinct from
548 the high-silica locations analyzed on sols 1000-1100, with the exception of the local presence of
549 Si-rich diagenetic halos that have been analyzed punctually and clearly postdate the rock
550 deposition as they cross also the overlying Stimson unit (Frydenvang et al., 2017). The good
551 correlation between WIS and CIA, calculated using different cations, also points towards the
552 limited influence of amorphous phases on the variability of these indices, although more work on

553 the amorphous component should be done to understand its origin and role in the processes of
554 alteration.

555
556 In summary, with limitations due to the lack of knowledge of the amorphous component and the
557 limited number of drill holes, the high CIA and WIS targets in Karasburg and Sutton Island are
558 consistent with a change in mineralogy related to a significant alteration by aqueous processes
559 with partial dissolution of primary minerals such as pyroxene and plagioclase.

560

561 *6.2 Diagenetic imprint*

562
563 Post-depositional alteration during diagenesis may have modified the mineralogical assemblages.
564 With that regard, another chemical trend observed is the slightly higher K₂O abundance (up to 2
565 wt.%) observed on the ternary diagram trending away from the A-CN axis (Fig. 4a). The Kimberley
566 sandstones are plotted for comparison in Fig. 4a showing that these fluvial sandstones with
567 anomalously high K-spar abundance (Le Deit et al., 2016, Treiman et al., 2016) (up to 5 wt.% K₂O)
568 cannot explain this trend by mixing or source rocks effects because for Murray formation rocks
569 the trend towards higher K₂O is correlated with the higher CIA

570
571 In contrast, illite and interlayered illite/smectite are possible candidates for such an enrichment;
572 they commonly form during burial and diagenesis (e.g., Fedo et al., 1995). It would also be in
573 agreement with modeling of the burial of Mount Sharp sediments (Borlina et al., 2015). CheMin
574 observations do not favor illite as the main phyllosilicate present (Bristow et al., 2018). The K₂O
575 abundance at Marimba is ~1.0 wt.% based on ChemCam data (Tab. S1). Yet, the modeled
576 proportion of sanidine in CheMin pattern is only 2.4±0.6 wt.% at Marimba (and less at Quela and
577 Sebina; Bristow et al., 2018) while jarosite is below detection limit (<1%). The equivalent
578 proportion of K₂O of a sanidine (~14 wt.% as an average of the four lower Murray drills, Morrison
579 et al., 2018) corresponds only to ~0.4 wt% K₂O, thus not accounting for all the potassium present
580 at Marimba. So, a contribution of illite or interlayered illite/smectite is possible, although difficult
581 to demonstrate.

582
583 The Plagioclase Index of Alteration (PIA) was proposed to correct for the addition of K₂O during
584 burial (Fedo et al., 1995) (Fig. 3). PIA values are usually slightly above CIA for Karasburg and Sutton
585 Island, suggesting the actual CIA values before burial were slightly higher, but they remain close
586 to CIA suggesting minor contributions. Thus, variations in K₂O cause minor effects on the CIA, and
587 diagenesis is unlikely to be the origin for the high CIA values.

588

589 *6.3 Origin of the alteration observed*

590 Both alteration indices, CIA and WIS, show evidence for substantial alteration of the Murray
591 formation. The CIA ranges from 50 to 63, strongly favoring that the alteration occurred in an open-
592 system, contrasting with the closed-system alteration observed in Sheepbed member mudstones
593 (Bradbury group at Yellowknife Bay), with CIA of ~ 40 (McLennan et al., 2014). Indeed, closed-
594 system alteration, such as found in local hydrothermal circulation or deep diagenesis, does not
595 increase the CIA index (remaining below the 50-50 line on figure 4). However, these chemical
596 indices usually represent a combination of weathering in the source region, during transport, and
597 in depositional areas (McLennan, 1993, Fedo et al., 1995, Cox et al., 1995, Nesbitt and Young,
598 1996, Yang et al., 2006, Garzanti et al., 2013). Thus, it remains debatable whether this alteration
599 predates or coincides with the deposition of the fine-grained sediments within Gale crater.

600 On one hand, altered source rocks are possible contributors to the high alteration indices. In this
601 case, the clay minerals present in high CIA Murray rocks are predicted to have a detrital origin.
602 Their presence could have been favored by grain-size sorting during depositional processes with
603 limited alteration during lake activity. Alteration of source rocks could have been in the form of
604 crustal or hydrothermal alteration related to the Gale impact crater, either by exhumation of deep
605 crustal rocks or by hydrothermal activity immediately after its formation. However, no high-T
606 secondary minerals indicative of hydrothermal conditions have been detected by CheMin in the
607 three upper drill holes or in previous clay-rich samples (Bristow et al., 2015). Only some of the
608 small fraction (3 wt.%) of clay minerals at Oudam (Hartmann's Valley) may be interpreted as of
609 pyrophyllite/talc type mixed within eolian deposits (Bristow et al., 2018). In contrast, the presence
610 of substantial proportions of di-octahedral phyllosilicates in the three upper drill holes is
611 consistent with an open-system, low-T alteration. Here too, the low proportion of mafic minerals,
612 usually sorted in higher proportion in finer-grained sediments (Siebach et al., 2017) does not favor
613 a pure detrital assemblage as these minerals should have been transported from unaltered
614 sections of the crater rim as observed in the Bradbury group. Thus, we do not favor the scenario
615 of rocks altered hydrothermally before or immediately after Gale crater's formation.
616 Nevertheless, it is still possible that weathering processes postdating Gale crater's formation
617 could have reprocessed hydrothermal clays into the smectites that are observed, or that
618 weathering affected source rocks after the emplacement of Gale crater but prior to the formation
619 of the lake deposits. On the other hand, some observations argue for in-situ weathering linked to
620 fluvial and lacustrine processes. The depletion of calcium interpreted by leaching is consistent
621 with observations of calcium sulfate cements (Newsom et al., 2017, Vaniman et al., 2017). The
622 Old Soaker outcrop (Sutton Island member), displays evidence for desiccation cracks that
623 demonstrate a subaerial exposure of these sediments (Stein et al., 2018). We have no constraint
624 on the frequency, duration, and repetition of such episodes, but the presence of these cracks
625 requires that the lake was shallow enough to permit local subaerial exposure of sediments where
626 additional weathering could have occurred, thus providing a possible explanation for targets that
627 display higher alteration in this section of Murray formation.

628 In summary, the weathering process(es) evidenced in this study could have taken place in the
629 source region, during transport or after deposition, or at these three stages and coevally in the
630 whole basin, and we cannot give more precision about this activity. However, we can exclude the
631 possibility that the observed mineralogy and chemistry consists only of detrital material of
632 hydrothermally altered rocks transported and deposited in a lake without further alteration.

633

634 *6.4 Climate conditions recorded by alteration indices*

635 Several terrestrial studies have shown the link between climate and variations in alteration
636 indices. Although the WIS has not been studied at the same level of detail as the CIA, maximum
637 values of the WIS of 20-25% are in agreement with conditions required by CIA of ~55-60 (Meunier
638 et al., 2013)(Table 4). Studies in the Arctic (Nesbitt and Young, 2016) and in Antarctica (Salvatore
639 et al., 2013) have shown that a cold and dry climate is unlikely to generate alteration indices >50.
640 Analyses of loess deposits in China and southeastern Europe indicate a CIA index of 60-65, after
641 semi-arid conditions during Pleistocene glacial-interglacial climates, with active weathering
642 during wet seasons over geologically long durations (Yang et al., 2006, Buggle et al., 2011). In
643 contrast, empirical laws extracted from a database of US soils (Sheldon et al., 2002) show that a
644 CIA of ~55-60 would translate into a temperate climate with high annual precipitation of ~600
645 mm. Alteration indices vary a lot according to parameters such as local climate, elevation, bedrock
646 composition and texture, not enabling the determination of a unique environment at a given
647 index value. In particular, felsic rocks such as the terrestrial continental crusts have higher starting
648 values of CIA (unaltered rocks ca. 45-50, e.g. Nesbitt and Young, 1996, Fedo et al., 1995) than
649 starting values for mafic rocks (unaltered rocks ca. 35-40, e.g. Babechuk et al., 2014) as observed
650 for Mars average crust, Sheepbed mudstones (Bradbury group) or soils, which all range from 35
651 to 40 (McLennan et al., 2014) as shown in Figure 4 and Table 4. Thus, at a similar CIA value,
652 alteration is underestimated for mafic rocks (which are found predominantly in Gale crater
653 sediments), supporting the idea that the Murray sediments are significantly altered. For instance,
654 basaltic sediments analyzed in Iceland, a currently wet and cold climate, have been estimated at
655 only 45-50, despite the high rate of precipitation in this region (Thorpe et al., 2017).

656 In summary, climate conditions required to explain our observations involve substantial liquid
657 water at the surface that a cold, dry climate cannot explain, although a specific climate cannot be
658 specified. Previous results from Curiosity indicated the presence of substantial fluvial and
659 lacustrine sedimentation, in what appeared to be relatively cold and dry environment according
660 to geochemical markers, i.e. CIA values at ~40 (Grotzinger et al., 2014, McLennan et al., 2014).
661 The finding of substantial weathering from chemical variations fills a gap in this history, and
662 suggests that the lacustrine activity was coupled with, or was preceded, by a period of open-
663 system weathering.

664 **7. Conclusions**

665 From Sol 750 to 1550, the Curiosity rover documented >130 m of stratigraphy during its 6 km
666 traverse of fine-grained sedimentary rocks of the Murray formation, at the base of Mt Sharp.
667 ChemCam data were used to estimate the level of weathering in these sedimentary rocks. Both
668 the Chemical Index of Alteration (CIA) and the Weathering Index Scale (WIS) indicate a
669 progressive increase in alteration up section, in the range of 55-60, locally reaching CIA values of
670 63 and WIS of 25% contrasting with limited weathering observed in the primary phase of the
671 mission (McLennan et al., 2014). To date, alteration indices of this magnitude have never been
672 observed in-situ on Mars. This enhanced alteration is consistent with abundant clay minerals of
673 both di-octahedral and tri-octahedral nature (Bristow et al., 2018). The increase in CIA and WIS
674 values is coupled with a decrease in calcium abundance, suggesting partial dissolution of
675 clinopyroxene and plagioclase, which both are observed in lower abundances in the drill holes up
676 section. In contrast to deep diagenetic or hydrothermal processes that occur in closed systems,
677 these observations indicate alteration in an open system with the presence of liquid water, at or
678 near the surface.

679 **Acknowledgements**

680 We acknowledge the two reviewers for their insightful comments. This research was carried out with
681 funding from the Centre National d'Etudes Spatiales (CNES) and the Agence Nationale de la Recherche
682 (ANR) under the program ANR-16-CE31-0012 entitled "*Mars-Prime*". Work in the United States was carried
683 out under contract from NASA's Mars Program Office. The data reported in this paper are archived at the
684 Planetary Data System, accessible at <http://pds-geosciences.wustl.edu/missions/msl/index.htm>. J.F.
685 acknowledges the support from the Carlsberg Foundation. Author contributions: N.M. analyzed ChemCam
686 data and wrote the text. E.D., C.F., S.M.M and M.S. provided input on stratigraphy and alteration indices
687 on Earth. E.D., O.F. J.F., O.G., J.H., L.L.D., S.M., P.M., H.E.N., W.R., F.R-H, R.W participated in ChemCam
688 data acquisition and processing. C.A, T.B., S.M., E.R., R.D participated to the processing and analyses of
689 CheMin data.

690
691
692

693 **Supplementary material**

694

695 Supplementary material related to this article can be found in the accompanying file.

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849 **Tab 1:** Example of a single target with 10 observation points (Upper Hadlock, sol 1505). The target average
 850 chemistry is obtained by removing point 5 which targeted a diagenetic feature (see Figure 2), and
 851 averaging the remaining points. Note that the deviation around the average is relatively limited as
 852 expected for homogeneous rocks with small grain size.

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Upper Hadlock	SiO ₂	TiO ₂	Al ₂ O ₃	FeO _T	MgO	CaO	Na ₂ O	K ₂ O	Sum
Point 1	56.4	1.03	12.50	17.0	4.60	2.70	2.52	0.93	97.79
Point 2	55.30	0.94	12.90	18.50	4.20	2.60	3.24	1.56	99.22
Point 3	57.20	0.99	11.80	15.80	4.20	2.30	2.73	1.66	96.71
Point 4	54.60	0.99	11.20	18.10	4.70	2.00	2.25	2.44	96.26
<i>Point 5</i>	<i>1.20</i>	<i>0.20</i>	<i>0.70</i>	<i>4.40</i>	<i>1.50</i>	<i>34.80</i>	<i>0.46</i>	<i>0.01</i>	<i>43.33</i>
Point 6	51.40	0.91	9.20	21.00	4.30	1.60	1.74	2.81	92.88
Point 7	53.30	0.91	10.00	20.60	4.00	1.70	1.85	2.74	95.08
Point 8	55.50	0.94	12.70	18.20	4.00	2.50	2.48	1.17	97.52
Point 9	55.80	0.98	12.50	17.20	4.30	2.50	2.84	1.17	97.22
Point 10	55.70	0.92	11.50	17.70	4.50	2.10	2.06	2.33	96.85
Average (without point 5)	55.02	0.96	11.59	18.23	4.31	2.22	2.41	1.86	96.6
Std Deviation	1.74	0.04	1.27	1.66	0.24	0.39	0.48	0.72	1.79

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858 **Tab. 2:** CheMin quantitative mineralogy at Marimba used for mass balance calculation for estimating the
 859 $\text{Fe}^{2+}/\text{Fe}^{3+}$ ratio for the WIS calculation (from Bristow et al., 2018). The mineral abundances modeled and
 860 presented here were updated compared to results currently available in the Planetary Data System.

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Mineral	Whole rock (%)	FeO (wt%) in pure minerals	Fe ₂ O ₃ (wt%) in pure minerals	FeO (wt%) contribution to sample	Fe ₂ O ₃ (wt%) contribution to sample	Fe ²⁺ (molar) in sample	Fe ³⁺ (molar) in sample
Plagioclase	14.0	N/A	N/A				
Hematite	6.4	0	100	0	6.400	0	0.0401
Ca-sulfates	7.0	N/A	N/A	-	-	-	-
Sanidine	2.4	N/A	N/A	-	-	-	-
Pyroxene*	0.7	30.9	0	0.2163	0	0.00301	0
Jarosite**	0.7	0	47.83	0	0.335	0	0.0021
Quartz**	0.7	N/A	N/A	-	-	-	-
Amorphous	40	?	?	?	?	?	?
Clay minerals***	28	0	5.0	0	1.40	0	0.0088
Whole Rock iron content	99.9	N/A	N/A	0.2163	8.135	0.00301	0.0509

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864 *Pyroxene composition assumed to be similar to the pigeonite from lower Murray drills (Morrison et al.,
 865 2017).

866 **At detection limit. We use here a value of 0.7 wt.%, which is the typical lowest abundance quantifiable.

867 ***Clay minerals assumed with 5% Fe₂O₃ from CheMin diffractometer modeling (Bristow et al., 2018).

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869 **Tab. 3** : Mineral abundances from CheMin data (Rampe et al., 2017, Bristow et al., 2018). Note that Mojave
870 and Telegraph Peaks compositions are strongly modified by diagenetic phases (jarosite, silica). Buckskin is
871 not included, given its presence in the high-silica region where ChemCam targets are excluded from the
872 present work.

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Mineral	Confidence Hills (Pahrump Hills)	Mojave (Pahrump Hills)	Telegraph Peak (Pahrump Hills)	Oudam (Hartmann's Valley)	Marimba (Karasburg)	Quela (Karasburg)	Sebina (Sutton Island)
Plagioclase	20.4	23.5	27.1	27.8	14.0	13.5	10.7
Sanidine	5.0	-	5.2	-	2.4	2.3	1.4
Olivine	1.2	0.2	1.1	-	-	-	-
Pyroxenes	13.8	7.0	7.6	5.3	0.7	2.7	2.8
Quartz	0.7	0.8	0.9	0.7	At detection limite	At detection limit	At detection limit
Hematite	6.8	3.0	1.1	13.9	6.4	7.1	6.9
Magnetite	3.0	3.0	8.2	-	-	-	-
Ca-sulfates	-	-	-	6.3	7.0	5.5	7.4
Apatite	1.3	1.8	1.9	-	-	-	-
Jarosite	1.1	3.1	1.5	-	At detection limit	At detection limit	0.9
Silica	-	-	18.2	-	-	-	-
Clay minerals	7.6	4.7	-	3	28	16	19
Amorphous	39.2	53	27.2	43	40	52	51.1

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877 Table 4: Summary of various CIA values found on Mars with Earth as comparison.

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Rock type	Value	Reference
Fresh basaltic rock, Earth	35-40	Babechuk et al., 2014
Fresh continental crust, Earth	45-50	Nesbitt and Young, 1996
Average Mars crust	37	McLennan et al., 2014
Bradbury formation, Sheepbed member, Mars	35-40	McLennan et al., 2014
Bradbury formation conglomerates, Mars	44	This study
Bradbury formation, Kimberley, Mars	30-40	This study
Murray formation, Pahrump Hills member, Mars	~50	Hurowitz et al., 2017
Murray formation, Sutton island member, Mars	48-63	This study

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