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### ► To cite this version:

Frances Rivera-Hernández, Dawn y Sumner, Nicolas Mangold, Kathryn M Stack, Olivier Forni, et al.. Using ChemCam LIBS data to constrain grain size in rocks on Mars: Proof of concept and application to rocks at Yellowknife Bay and Pahrump Hills, Gale crater. *Icarus*, 2019, 321, pp.82-98. 10.1016/j.icarus.2018.10.023 . hal-02933737

**HAL Id: hal-02933737**

**<https://hal.science/hal-02933737>**

Submitted on 8 Sep 2020

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1 **Using ChemCam LIBS data to constrain grain size in rocks on Mars: Proof of concept and**  
2 **application to rocks at Yellowknife Bay and Pahrump Hills, Gale crater**

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49 **Abstract.** Grain size in martian sedimentary rocks can be constrained using point-to-point  
50 chemical variabilities in Laser Induced Breakdown Spectroscopy (LIBS) data from the  
51 ChemCam instrument on the Mars Science Laboratory (MSL) *Curiosity* rover. The diameter of  
52 each point vaporized by the ChemCam laser is in the range of medium to coarse sand in size.  
53 Thus, rocks with grains smaller than the laser spot size produce bulk rock compositions at each  
54 LIBS point and low point-to-point chemical variability among LIBS points. In contrast, analyses  
55 of rocks with grains about the size of the spot or larger contain contributions from individual  
56 grains at each point and often have high point-to-point chemical variability. The Gini index, a  
57 statistical parameter, was used to calculate the point-to-point chemical variability in major-  
58 element oxide compositions derived from the ChemCam LIBS data. The variability of each  
59 oxide was then combined to derive a Gini mean score,  $G_{MEAN}$ , for each LIBS observation. A  
60 standard procedure was developed and validated using observations of sedimentary rocks of  
61 various grain sizes from the Yellowknife Bay formation and the Pahrump Hills member of the  
62 Murray formation in Gale crater. Overall, finer-grained rocks have smaller  $G_{MEAN}$  than coarser-  
63 grained rocks. To calibrate grain size ranges for specific  $G_{MEAN}$  values, the LIBS major-element  
64 oxide compositions were first normalized as a group. Next, grain size estimates based on visual  
65 assessment of high-resolution images were compared to  $G_{MEAN}$  values for the same targets to  
66 create a calibrated scale. This calibrated scale was used to infer the grain size of rocks with  
67 unknown grain size. Overall, the grain sizes predicted for rocks with unknown grain size  
68 overlapped with those of known grain size from the same units and/or bedrock targets. The grain  
69 sizes inferred using the  $G_{MEAN}$  based on ChemCam LIBS data are complimentary to those  
70 determined from images and both techniques can be used to improve interpretations of the  
71 depositional environments of rocks analyzed by *Curiosity* and future Mars missions with LIBS,  
72 such as the Mars 2020 rover.

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75 Keywords: Mars, grain size, sedimentary rocks, LIBS, Mars Science Laboratory

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## 95 1. Introduction

96 The primary goal of the Mars Science Laboratory (MSL) mission is to characterize  
97 habitable environments of early Mars in the sedimentary record of Gale crater (e.g., Grotzinger  
98 et al., 2012). Evaluating past habitability requires reconstructing the conditions of sediment  
99 transport and deposition from sedimentary rocks with an emphasis on interpreting changes in the  
100 ancient environments. Characterizing the size of grains and their distribution in sedimentary  
101 rocks provides information about how the grains were transported, which is crucial for  
102 interpreting depositional environments. For example, laminated mudstones accumulate in  
103 environments with very low flow speeds such as lakes, whereas cross-bedded sandstones and  
104 conglomerates require higher flow speeds and are characteristic of river and beach deposits.  
105 Identifying lacustrine deposits is important as lakes both indicate a sustained presence of liquid  
106 water and commonly preserve biosignatures on Earth (Farmer and DesMarais, 1999; Summons  
107 et al., 2011). However, determining the size of sediment grains in rocks is challenging on Mars  
108 due to the resolution limits of rover image data. Whether or not grains larger than mud in size (>  
109 62.5  $\mu\text{m}$ ) can be resolved in an image depends on a variety of factors, such as camera  
110 specifications, the distance of the camera from the rock, illumination, grain color contrast, and  
111 dust cover. Except for coarse-grained rocks and several well studied sites of importance (e.g.,  
112 Williams et al., 2013; Edgar et al., 2017; Stack et al., 2016; Banham et al., 2018), the grain sizes  
113 of most rocks observed in *Curiosity* rover images are unknown.

114 To supplement the grain sizes determined using image-based analyses, Mangold et al.  
115 (2017) proposed that point-to-point chemical variabilities in ChemCam Laser Induced  
116 Breakdown Spectroscopy (LIBS) data could be used as a proxy to infer the size of grains or  
117 crystals in sedimentary and igneous rocks. This grain size proxy was validated for a few rocks  
118 sampled by the ChemCam LIBS instrument (Mangold et al., 2017), and it can be applied  
119 extensively to diverse suites of rocks once a standard procedure for its application is developed  
120 and tested.

121 In this paper, we evaluate the robustness and reproducibility of using ChemCam LIBS  
122 data to infer the size of grains in sedimentary rocks along the *Curiosity* traverse in two areas,  
123 informally called Yellowknife Bay (YKB) and the Pahrump Hills (PH; Fig. 1). Both regions  
124 have mudstones and sandstones (Grotzinger et al., 2014; 2015), providing an opportunity to  
125 compare and contrast the point-to-point chemical variability of rocks with similar grain size  
126 distributions. Results from this comparison were then used to develop a scale to infer the grain  
127 size distribution of rocks with known and unknown grain size. The standard procedure presented  
128 here can be applied to additional ChemCam LIBS data and to results from future Mars missions  
129 with LIBS, such as the Mars 2020 rover (Wiens et al., 2017).

## 130 2. Methods

### 131 2.1. ChemCam LIBS data

132 The ChemCam instrument suite consists of a LIBS and the Remote Micro-Imager (RMI)  
133 that collects co-located context images (Maurice et al., 2012a; Wiens et al., 2012). The LIBS  
134 uses a pulsed laser to ablate small spots (~0.4-0.6 mm in diameter), commonly in lines of 5 to 10  
135 points or matrixes of 3x3 points, on targets 2 to 7 m from the rover (Maurice et al., 2012a; Wiens  
136 et al., 2012). The measurement points are typically spaced by 2 mrad, which represents a  
137 sampling every 6 mm for a target at 3 m. The laser spot size increases with distance from the  
138 rover, affecting the amount of material that is ablated by the laser and thus analyzed by  
139 ChemCam (Maurice et al., 2012b). The light from the ablated plasma at each spot is passed  
140

141 through three spectrometers to acquire an atomic emission spectrum in the wavelength ranges:  
142 240-342 nm, 382-469 nm, and 474-906 nm (Maurice et al., 2012a; Wiens et al., 2012). When the  
143 plasma cools, atoms in the plasma can recombine and molecular emission lines can also be  
144 observed (e.g., Cremers and Radziemski, 2013; Anderson et al., 2017). For each laser pulse, a  
145 LIBS spectrum is acquired. Commonly, there are 30 laser shots per point, with the first laser  
146 shots removing surface dust and coatings (Wiens et al., 2013). For each point, the weight percent  
147 (wt.%) of eight major-element oxides ( $\text{SiO}_2$ ,  $\text{TiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{FeO}_T$ ,  $\text{MgO}$ ,  $\text{CaO}$ ,  $\text{Na}_2\text{O}$ , and  $\text{K}_2\text{O}$ ) are  
148 derived from multivariate analysis comparing the elemental emission lines of martian targets to  
149 Earth standards (Clegg et al., 2009; 2017; Cousin et al., 2011; Wiens et al., 2013).  $\text{FeO}_T$  refers to  
150 total iron, including both  $\text{FeO}$  and  $\text{Fe}_2\text{O}_3$ , as these are not distinguishable using ChemCam. The  
151 first five shots per LIBS point are removed to exclude the influence of dust (Lasue et al., 2016)  
152 and the other 25 shots are used to calculate the mean, root mean square error (RMSE), and  
153 standard deviation of every major-element oxide (Wiens et al., 2013; Clegg et al., 2017). If the  
154 modeled spectrum can be reproduced with all eight major-element oxides, then the total sum of  
155 their weight percent equals 100% with error in accuracy of less than 10% (Clegg et al., 2017).  
156 When the major-element oxide total is less than 100%, there is a missing component in the  
157 model, for example from an abundance of Cl, H, F, Li, Sr, Ba, Rb, Mn, P, S, Ni, Zn, or Cr. Thus,  
158 the sum of the major-element oxides is not normalized to 100%. An abundance of sulfate  
159 minerals from diagenesis (Nachon et al., 2014; 2017) is the most common cause for oxide totals  
160 of less than 100%. Only the major-element oxides are discussed here.

161 To verify the quality of the LIBS data, calibration targets onboard *Curiosity* are regularly  
162 analyzed (Fabre et al., 2011; Vaniman et al., 2012). Precision for the ChemCam LIBS data can  
163 be determined by quantifying the variability between repeated measurements either on the  
164 calibration targets, or on relatively homogeneous and fine-grained rock exposures on Mars (i.e.,  
165 mudstones; Mangold et al., 2015; Clegg et al., 2017). The accuracy of analyses is predicted using  
166 a RMSE for a representative test set of rock calibration targets, chosen such that each major-  
167 element oxide has distributions similar to the full set of rock standards used to generate the  
168 regression models in the multivariate analysis (Clegg et al., 2017). Typically for the ChemCam  
169 LIBS, accuracy is higher in magnitude than precision. For example, for the first 801 sols of the  
170 MSL mission, the total RMSEs (accuracy) for the Shergottite calibration target are (in wt.%):  
171  $\text{SiO}_2=5.04$ ,  $\text{TiO}_2=0.14$ ,  $\text{Al}_2\text{O}_3=4.02$ ,  $\text{FeO}_T=5.28$ ,  $\text{MgO}=1.39$ ,  $\text{CaO}=2.56$ ,  $\text{Na}_2\text{O}=1.09$ , and  
172  $\text{K}_2\text{O}=0.67$  (Clegg et al., 2017). Whereas, precision values calculated from sols 271, 352, and  
173 357, for the same major elements as above are 1.53, 0.14, 0.57, 1.83, 0.49, 0.42, 0.49, and 0.14  
174 wt.% respectively (Blaney et al., 2014). As this study depends on point-to-point LIBS  
175 variabilities, precision error is propagated in our calculations (see Section 2.2 and the Appendix).  
176

## 177 **2.2. ChemCam LIBS data as proxy for grain size**

178 The size of grains in a rock can be constrained using LIBS data because the diameter of  
179 each point vaporized by the ChemCam laser is known (medium to coarse sand in size, with  
180 increasing distance to the target). If a rock has grains considerably smaller than the laser spot  
181 diameter, the bulk composition of the rock is analyzed at each point (Anderson et al., 2011;  
182 McCanta et al., 2013; 2017) and little to no point-to-point chemical variability is observed (Fig.  
183 2; Sivakumar et al., 2014). However, if a rock has grains similar in size or larger than the laser  
184 spot diameter, each point can potentially have a different composition reflecting individual grain  
185 compositions (McCanta et al., 2013; 2017). A large number of spots would have to be analyzed  
186 to get a bulk composition for the rock (Fig. 2; Anderson et al., 2011). If a rock has grains much

187 larger than the distance between the LIBS points (typically ~6 mm; pebble or larger), the laser  
188 may hit the same grain more than once, reducing variability, though these grains should be  
189 resolved in images. If a rock has grains of uniform composition, regardless of grain size, the  
190 LIBS data will exhibit little to no point-to-point chemical variability and grain size cannot be  
191 constrained by this method.

192 For coarse-grained rocks with non-uniform compositions and grains smaller than fine  
193 gravel in size, the presence of grains may be inferred by looking at heterogeneities in the LIBS  
194 data (Sivakumar et al., 2014; Mangold et al., 2017). Mangold et al. (2017) proposed that these  
195 heterogeneities can be quantified and used as a proxy for inferring grain size by implementing a  
196 statistical measurement called the Gini index ( $G$ ). The Gini index is commonly used to  
197 characterize variations across a population, such as inequalities in wealth (Gini, 1921). The Gini  
198 index varies from 0 to 1, where  $G=0$  expresses complete equality or uniformity and  $G=1$   
199 complete inequality. Applied to grain size, a  $G=0$  suggests the rock has grains much smaller than  
200 the laser spot size (mud) or are of a uniform composition, and if  $G=1$ , then the laser hit distinct  
201 compositions at each point and the grain size is likely larger than the spot size. To use the Gini  
202 index as a proxy for grain size, we developed the Gini index mean score (GIMS), with a slightly  
203 different implementation from Mangold et al. (2017). The values reported here are averages,  
204  $G_{\text{MEAN}}$ , calculated by applying an arithmetic mean to the Gini indices of each major-element  
205 oxide,  $G_i$ , except  $\text{TiO}_2$ . For all of the rock targets, the weight percent of each oxide was  
206 normalized based on the range of values in all samples before calculating  $G_i$  and  $G_{\text{MEAN}}$  (details in  
207 *Appendix*). Thus,  $G_i$  and  $G_{\text{MEAN}}$  depend on the details of the normalization, and different rocks  
208 can only be compared to each other if they are similarly normalized. When different  
209 normalizations are used, Gini index values may be different for the same grain size and thus,  
210 Gini index values reported in Mangold et al. (2017) or in other publications may not correspond  
211 to the same grain sizes presented here using the GIMS. Two different standard deviation metrics  
212 were calculated for each  $G_{\text{MEAN}}$ ,  $\text{STDr}$  and  $\text{STDc}$ , both derived from standard deviations of every  
213 major-element oxide per LIBS point. For  $\text{STDr}$ , the standard deviations are based on variations  
214 in the shot-to-shot LIBS measurements on the rock targets used in the GIMS analysis  
215 (Supplemental Table 1), whereas for  $\text{STDc}$ , the values are scaled standard deviations derived  
216 from the Shergottite calibration target on *Curiosity* (see Blaney et al., 2014). The standard  
217 deviations for each LIBS point and major-element oxide, were propagated to errors on  $G_{\text{MEAN}}$   
218 using an iterated bootstrap with 1000 iterations.  $\text{STDr}$  and  $\text{STDc}$  are 0.00-0.02 and 0.00-0.01,  
219 respectively, which is much lower than the variations due to rock composition ( $G_{\text{MEAN}}=0.02$ -  
220 0.29; Table 2).  $\text{STDr}$  is used in the text and figures due to its slightly larger variation.  
221 Interestingly, the lowest  $G_{\text{MEAN}}$  value gives another precision estimate because mudstones are  
222 predicted to have a  $G_{\text{MEAN}}=0$  (completely homogenous). The lowest calculated  $G_{\text{MEAN}}$  was 0.02  
223 on YKB mudstones, consistent with the  $1\sigma$   $\text{STDr}$  of 0.00-0.01 on those samples (Table 2),  
224 suggesting that instrumental precision error is smaller than typical variations in LIBS spot  
225 compositions.

226  $G_{\text{MEAN}}$  scores need to be calibrated to grain size using samples with both LIBS data and  
227 appropriate corresponding images, with the approximate compositions and mineral assemblages  
228 present in the unknown rocks. By calculating the  $G_{\text{MEAN}}$  of rocks with known grain size,  $G_{\text{MEAN}}$   
229 ranges can be scaled to grain sizes ranges (Section 5.2). Other rocks with unknown grain size can  
230 then be compared to this scale to constrain their grain size. By comparing rocks with known  
231 grain size that were not involved in the scaling, the robustness of the GIMS can be evaluated for  
232 a specific suite of rocks.

233 In addition to grain size, other factors can also cause point-to-point heterogeneities and  
234 homogeneities. Regardless of grain size, if a rock has grains with a uniform composition, then  
235 the rock will have little to no point-to-point chemical variability, and thus a low  $G_{\text{MEAN}}$ . This is  
236 not expected for sedimentary rocks along *Curiosity's* traverse, as these contain a diverse suite of  
237 basaltic grain compositions, including pyroxene, feldspar, olivine, other mafic minerals, and  
238 glass (Vaniman et al., 2013; Sautter et al., 2014; Mangold et al., 2016; Cousin et al., 2017;  
239 Rampe et al., 2017). Heterogeneities in composition in a rock can be caused by contamination by  
240 loose sediment (sand and dust), diagenetic features, and intergranular cements. Different steps  
241 were taken to minimize the contribution of these heterogeneities in our analyses. First, RMI  
242 images were used to identify where the laser hit a rock at each LIBS point. Those points that  
243 sampled visually resolvable diagenetic features, loose sediment, cracks, and sharp edges were  
244 excluded from the GIMS analyses. Targets with abundant diagenetic features in the scene,  
245 regardless of whether or not the laser hit the features, were also excluded. Next, points with <  
246 87% major-element oxide totals were removed (see Appendix), as low oxide totals commonly  
247 suggest the presence of a non-resolvable diagenetic contribution (e.g., Jackson, 2016), such as a  
248 cement (Newsom et al., 2017; Nellessen et al., 2018) or diagenetic feature (e.g., sulfate veins; see  
249 Section 2.1). Only rock targets with more than five points were used for the GIMS analysis, as  
250 that is the minimum number of points necessary to obtain a statistically significant Gini index  
251 result (Mangold et al., 2017). Thus, the GIMS was only applied to select rocks that passed the  
252 filtering process. Prescreening the rock targets allows a reproducible, standard procedure to be  
253 applied to all targets.

254 The filtered ChemCam LIBS data from 50 rocks at YKB and 15 at PH were used in the  
255 GIMS analysis (Supplemental Table 1). The ChemCam targets are informally named by the  
256 MSL science team and those names are used here to reference the rock targets. The Planetary  
257 Data System also includes this classification scheme. Following the format of target names in the  
258 Planetary Data System, target names containing multiple words include underscores (i.e.,  
259 rock\_name). When more than one LIBS analysis is acquired on the same rock target, underscores  
260 are used to enumerate each analysis location on the rock (i.e., rock\_name\_1, rock\_name\_2).

### 261 262 **2.3. Image Data Sets**

263 Previous work done to constrain grain sizes at YKB and PH (see Section 3.1; Blaney et  
264 al., 2014; Grotzinger et al., 2014; 2015; Mangold et al., 2015; Anderson et al., 2015; Stack et al.,  
265 2016; Edgar et al., 2017), coupled with additional textural analysis performed in this study,  
266 enables calibration of the GIMS for these units. Limitations on grain size observed in rover  
267 images depend on the camera characteristics, the target distance, illumination, and amount of  
268 dust cover. Images taken by MAHLI and RMI were used to measure grain size, whereas images  
269 taken by Mastcam were used as context for the RMI and MAHLI images and to document  
270 sedimentary textures, such as cross-bedding.

271 The RMI has an angular pixel size of 0.0195 mrad/pixel, a circular field of view of 20  
272 mrad over  $1024 \times 1024$  pixels and produces black and white images that are co-located with the  
273 LIBS analyses (Le Mouélic et al., 2015). Depending on the distance of the rover to the LIBS  
274 target, the spatial resolution of an RMI image can range from  $\sim 0.04$  to  $\sim 0.15$  mm/pixel (Le  
275 Mouélic et al., 2015). Most ChemCam LIBS measurements are made at  $\sim 2$ -3 m distance,  
276 restricting the finest grain size that can be resolved to fine to medium sand. In addition, the  
277 RMI's narrow depth of focus ( $\sim 1$  cm at 2 m distance; Le Mouélic et al., 2015) makes it difficult  
278 to resolve grains outside the best focus area in a RMI image mosaic (Anderson et al., 2014).

279 However, RMI images are the best for characterizing grain sizes for our analyses as they show  
280 the regions targeted by the ChemCam LIBS and the RMI resolution is compatible with the laser  
281 spot size.

282 The Mastcams are a two-instrument suite that provide color images (Malin et al., 2016;  
283 Bell et al., 2017). The left Mastcam (M-34) has a 34 mm focal length, 0.22 mrad/pixel, and  $18.4^\circ$   
284  $\times 15^\circ$  effective field of view over  $1600 \times 1200$  pixels (Malin et al., 2016; Bell et al., 2017). The  
285 right Mastcam (M-100) has a 100 mm focal length, 0.074 mrad/pixel, and an effective field of  
286 view of  $6.3^\circ \times 5.1^\circ$  over  $1600 \times 1200$  pixels (Malin et al., 2016; Bell et al., 2017). Most targets  
287 investigated by ChemCam have corresponding Mastcam images, typically taken by the M-100,  
288 and these can sometimes be merged with the RMI images to provide color information for the  
289 scene. The M-100 can resolve coarse sand sized features ( $\sim 500 \mu\text{m}$ ; larger than the laser spot  
290 size) at a distance of 3 m.

291 The MAHLI is mounted on the rover's arm, and it is capable of color and stereo imaging  
292 by physical offset of the arm between images (Edgett et al., 2012). It has a macrolens that can  
293 focus over a range of distances from 2.1 cm to infinity (Edgett et al., 2012). At the minimum  
294 working distance (2.1 cm), MAHLI has a 18.4 mm focal length,  $26.8^\circ \times 20.1^\circ$  effective field of  
295 view over  $1600 \times 1200$  pixels, and spatial resolution of  $\sim 14 \mu\text{m}/\text{pixel}$  (very coarse silt in size;  
296 Edgett et al., 2012). MAHLI images are commonly used for grain size and other textural  
297 analyses, as well as to provide context imaging for other contact science. It takes  $\sim 2.5$  pixels to  
298 enable a confident grain detection. In this study only rock surfaces brushed by the Dust Removal  
299 Tool are used for grain size analyses at the coarse silt to very fine sand scale because dust can  
300 mask the underlying surface and differentiating between loose dust grains and grains embedded  
301 in a rock becomes difficult. Due to the significant time and power resources needed to deploy  
302 the rover's arm, MAHLI is primarily used for high-priority science at strategically planned stops  
303 along the rover traverse (Vasavada et al., 2014; Edgett et al., 2015; Yingst et al., 2016). Thus,  
304 image data from the other cameras are necessary to provide a more continuous record of the  
305 sedimentary textures and structures of the rocks encountered along the traverse. However, the  
306 other cameras can only resolve grain sizes typically coarser than fine sand, which is insufficient  
307 for the desired environmental interpretations.

308

### 309 **3. Geologic setting**

310 The *Curiosity* rover landed in Gale crater on 6 August 2012 at a site called Bradbury  
311 Landing (Fig. 1). Gale crater is located in the equatorial region of Mars ( $137.7^\circ\text{E}$ ,  $5.44^\circ\text{S}$ ) along  
312 the crustal dichotomy between the smooth northern lowlands and the cratered southern  
313 highlands. The crater is  $\sim 154$  km in diameter, and its center it has a  $\sim 5$  km high layered mound,  
314 informally called Mount Sharp (formally named Aeolis Mons), that has hematite-, phyllosilicate-  
315 , and sulfate-bearing stratigraphic layers (Malin and Edgett, 2000; Milliken et al., 2010; Fraeman  
316 et al. 2013; 2016). Based on crater counts, Gale crater formed near the Noachian-Hesperian  
317 transition (3.6 Ga; Le Deit et al., 2013, Thomson et al., 2011), and the crater-filling material may  
318 have accumulated through to the early Hesperian (Thomson et al., 2011; Grant et al., 2014;  
319 Palucis et al., 2014; Grotzinger et al., 2015). *Curiosity* landed at the distal end of the Peace Vallis  
320 alluvial fan, which is sourced from the northern crater rim (Palucis et al., 2014). After landing,  
321 the *Curiosity* rover performed analyses at various waypoints on its journey up Mount Sharp,  
322 including the outcrops of the YKB and PH member of the Murray formation (Fig. 1b). The YKB  
323 and PH regions include important facies characterized by the rover, and a number of rocks were



324 co-investigated by ChemCam, Mastcam, and MAHLI at these locations. Such targets provide  
325 good controls for connecting  $G_{\text{MEAN}}$  and grain sizes determined from images.  
326

### 327 **3.1. Sedimentology and stratigraphy**

328 *Yellowknife Bay formation:* On sol 125, 445 m east and 14 m below Bradbury Landing,  
329 *Curiosity* encountered the first outcrops of the ~5.2 m thick YKB formation (Fig. 3). The rover  
330 investigated the region in detail until sol 324. In ascending order in elevation, the three members  
331 of the YKB formation are informally named the Sheepbed, Gillespie Lake, and Glenelg members  
332 (Grotzinger et al., 2014; 2015). The mean dip of the rocks at YKB and PH regions is  
333 approximately horizontal, which allows elevation to be used as a proxy for stratigraphic height  
334 (Grotzinger et al., 2015). The Sheepbed member consists of mudstones interpreted to be  
335 lacustrine in origin (Grotzinger et al., 2014; 2015). The Gillespie Lake member consists  
336 primarily of well-cemented medium to coarse sandstones with occasional pebbly sandstones. It  
337 generally has a massive appearance, though poorly defined cross-bedding was observed  
338 (Grotzinger et al., 2014; Mangold et al., 2015). The sediments of the Gillespie Lake sandstones  
339 are interpreted as deposited in a distal fluvial environment (Grotzinger et al., 2014). The Glenelg  
340 member is exposed at four main outcrops: Point Lake, Shaler, Rocknest, and Bathurst Inlet.  
341 Rocks from Bathurst Inlet were not considered in our analyses. The rocks of the Point Lake  
342 outcrop exhibit pitted textures with voids ranging from millimeters to several centimeters in  
343 diameter (Grotzinger et al., 2014; 2015). They are interpreted to be coarse to very coarse  
344 sandstones based on the identification of a few grains that were 0.5-2.0 mm in size (Mangold et  
345 al., 2015). The Shaler outcrop consists of well-exposed cross-stratified coarse sandstones with  
346 granules interpreted to be fluvial in origin (Grotzinger et al., 2014; 2015; Anderson et al., 2015;  
347 Edgar et al., 2017). The rocks of the Rocknest outcrop have either massive or finely laminated  
348 textures, including low angle cross-lamination, and some exhibit circular to ovoid pits several  
349 millimeters in diameter (Blaney et al., 2014; Grotzinger et al., 2014; Mangold et al., 2015). They  
350 are interpreted as either siltstones or sandstones.  
351

352 *Pahrump Hills member, Murray formation:* On sol 750, *Curiosity* encountered the first  
353 exposures of lower Mount Sharp stratigraphy at an area informally called Pahrump Hills, the  
354 lowest member of the Murray formation examined by the rover (Grotzinger et al., 2015). The  
355 rover investigated the region in detail until sol 940, performing three successive traverses of the  
356 outcrop. The PH section is ~13 m thick and had 9 areas studied in detail, in ascending order in  
357 elevation: Shoemaker, Confidence Hills, Pink Cliffs, Book Cliffs, Alexander Hills, Chinle,  
358 Telegraph Peak, Whale Rock, and Salsberry Peak (Fig. 4; Stack et al., 2016). Rocks from:  
359 Shoemaker, Pink Cliffs, Telegraph Peak, and Salsberry Peak were not included in our analyses.  
360 The lower PH section, from Shoemaker to Telegraph Peak, consists of laminated mudstones  
361 and/or siltstones with grains smaller than the limit of resolution of MAHLI images (coarse  
362 silt/very fine sand; Grotzinger et al., 2015; Stack et al., 2016). The section includes the cross-  
363 laminated sandstone facies of Whale Rock (Grotzinger et al., 2015; Stack et al., 2016). The  
364 association of facies at PH is consistent with a coarsening upward trend and is interpreted to be a  
365 fluvio-lacustrine depositional environment, with the mudstones representing lacustrine  
366 deposition and sandstones fluvial or subaqueous deposition on the foreslope of a delta  
367 (Grotzinger et al., 2015).  
368

### 369 **3.2. Overview of the chemistry and diagenetic history of Yellowknife Bay and Pahrump Hills**

370 While the rocks of both YKB and PH have bulk basaltic compositions (Grotzinger et al.,  
371 2015), they are chemically distinct from each other with varying diagenetic histories (McLennan  
372 et al., 2014; Ming et al., 2014; Stack et al., 2014; Vaniman et al., 2013; Mangold et al., 2015;  
373 Nachon et al., 2017). Compared to PH, the rocks of YKB generally have mean compositions  
374 more similar to average martian crust (McLennan, 2012), with lower SiO<sub>2</sub>, and higher MgO,  
375 FeO, and CaO (see Supplemental Table 1; McLennan et al., 2014; Mangold et al., 2015;  
376 Mangold et al., 2016; Nachon et al., 2017). There are compositional variations within both  
377 formations as well (see Supplemental Table 1; Mangold et al., 2015; Rampe et al., 2017). For  
378 example, the Whale Rock outcrop in PH has relatively high CaO, up to 15.2 wt %, compared to  
379 the mean of YKB and PH, 5.3 and 4.97 wt%, respectively (see Supplemental Table 1).  
380 Variations in composition between members and formations are likely due to variations in  
381 provenance, mechanical sorting, post-depositional interaction with different diagenetic fluids,  
382 and intergranular cements (e.g., Mangold et al., 2017; Siebach et al., 2017). The GIMS  
383 calibration includes the normalization of all the LIBS data to account for regional variations of  
384 major-element oxide weight fractions (see Appendix).

385 Both YKB and PH contain diverse diagenetic features (Vaniman et al., 2013; Grotzinger  
386 et al., 2014; 2015; McLennan et al., 2014; Stack et al., 2014; Nachon et al., 2014; 2017; Mangold  
387 et al., 2015). At YKB, diagenetic features included solid nodules, hollow nodules, raised ridges,  
388 sulfate-filled fractures and nodules, vugs, and sedimentary dykes (Grotzinger et al., 2014; Stack  
389 et al., 2014; Siebach et al., 2014; Nachon et al., 2014). Nodules were sub-mm to mm concretions  
390 and densely clustered in some locations of the Sheepbed member; however, the compositions of  
391 concretion-rich rocks were not distinct from the overall composition of other Sheepbed rocks  
392 (Stack et al., 2014). The Sheepbed member is also cross-cut by pervasive light toned Ca-sulfate  
393 veins, <2cm in width, that were less frequent in other YKB members (Nachon et al., 2014).  
394 Raised ridges at YKB were curvilinear, narrow (<1-6 mm in width and several centimeters in  
395 length) and differentially weathered relative to the surrounding rock, creating the raised relief  
396 (Siebach et al., 2014; Leveille et al., 2014). These ridges are primarily located in the Sheepbed  
397 member and have elevated Mg and Li, with some increases in Fe, Si, Cl, and Br also observed,  
398 based on ChemCam and APXS analyses (Grotzinger et al., 2014; Leveille et al., 2014).

399 At PH, diagenetic features included dendritic aggregates, enhanced relief features with  
400 various geometries, dark raised-ridges, and light-toned sulfate veins (Nachon et al., 2017).  
401 Enhanced relief features were enriched in MgO, and depleted in SiO and FeO compared to the  
402 host rock (Kah et al., 2015; Nachon et al., 2017). Dark raised ridges were enhanced with MgO  
403 and CaO and did not have any obvious systematic depletions in SiO or FeO (Nachon et al.,  
404 2017). Light-toned Ca-sulfate veins were a few millimeters wide and a centimeter to tens of  
405 centimeters in length. They either cross-cut the host rock or occurred aligned within coarse-  
406 grained sedimentary layers (Kronyak et al., 2015; Nachon et al., 2017). The lower outcrops of  
407 PH have evidence for diagenetic mineral precipitation, including lenticular crystal pseudomorphs  
408 (Kah et al., 2015), preferentially cemented laminae, and late-diagenetic crystal clusters. Silt to  
409 medium sand-sized dark features were pervasive in rocks from Pink Cliffs, Shoemaker, Book  
410 Cliffs, and Telegraph Peak, but it is unclear whether they are clastic grains or micro-diagenetic  
411 concretions (Stack et al., 2016). Rocks from YKB and PH with observable or suspected  
412 diagenetic features were excluded from the GIMS analysis because diagenesis can obscure the  
413 relationship between chemical variability and grain size.

414

#### 415 **4. Constraining grain size from images**

416 Rocks with grain sizes ranging from mud to coarse sand at YKB and PH serve as  
417 standards for the GIMS calibration (Table 1). Of the calibration rocks, 13 are in the Sheepbed  
418 member, 2 in the Gillespie Lake member, 1 in the Shaler outcrop of the Glenelg member, 3 in  
419 Confidence Hills, 2 in Book Cliffs, 3 in Chinle, and 3 in Whale Rock (Table 1). In addition to the  
420 rocks used as calibration standards from YKB, grain sizes are also reported for 11 other rocks,  
421 including some with rare resolvable grains in RMI images (Table 2).

422 Three main criteria were used to identify grains in images: they had to 1) be embedded in  
423 the rock, 2) have positive relief, and 3) be discrete and approximately spherical (to differentiate  
424 them from scratches). The diameters of grains were measured as their longest axes using the  
425 image-processing program ImageJ (<http://imagej.nih.gov/ij/>). Grain sizes were classified using  
426 the Wentworth scale (Wentworth, 1992), where very fine sand=62.5-125  $\mu\text{m}$ , fine sand=125-250  
427  $\mu\text{m}$ , medium sand=0.25-0.5 mm, coarse sand=0.5-1 mm, very coarse sand=1-2 mm, very fine  
428 gravel (granule)=2-4 mm, and fine gravel (pebble)=4-8 mm. Clay and silt were grouped together  
429 as mud (62.5  $\mu\text{m}$  and smaller), where coarse silt=20-62.5  $\mu\text{m}$ . The finest grain size observable in  
430 RMI images is fine sand due to resolution limits. Mud was inferred for those targets that did not  
431 have resolvable grains in MAHLI images nor evidence of cross stratification in any images. Due  
432 to limited resolution of the images and to dust mantling the rocks, the median grain size of a rock  
433 might be smaller than the finest resolvable grain in an image.

434

## 435 **5. Results**

### 436 **5.1. Grain size of calibration rocks using image data**

437 All ChemCam analyses on the Sheepbed member of YKB reported here are used as  
438 standards (Table 1). The other members in YKB are characterized by having a wide range of  
439 grain sizes, and specific rock targets were picked to represent each grain size range. For rocks  
440 with fine to medium sand, the target Nanok from the Gillespie Lake member is used as a  
441 standard (Fig. 5a). Nanok has a rough texture and consists of moderately to moderately well  
442 sorted fine to medium sand with sparse coarse sand, though the coarse sand was not near the  
443 laser pits (Fig. 5a). Only <5-10 % distinct grains are resolvable in the rock with most grains too  
444 small or showing too little color contrast to resolve. Dust mantled the area near the laser pits,  
445 and it was difficult to ascertain the proportions of fine to medium sand in the RMI image. For  
446 rocks with medium to very coarse sand, the targets Gillespie\_Lake\_1 and Wakham\_Bay are used  
447 as standards. Gillespie\_Lake\_1 is very poorly sorted and has fine sand to granule size sediment,  
448 though the LIBS laser hit an area dominated by medium to coarse sand with sparse very coarse  
449 sand (Fig. 5b). Wakham\_Bay is moderately to poorly sorted and has medium to very coarse  
450 sand with sparse granules; at least one of the points hit a very coarse sand size grain, and another  
451 hit a granule (Fig. 5c).

452 Unlike the upper members of YKB, grains could not be resolved in the RMI images for  
453 the majority of the rocks in PH due to their fine grain sizes. Because of this, grain size estimates  
454 were primarily based on observations made using MAHLI images, supplemented by the RMI  
455 images when applicable. Thus, grain size estimates for most PH rocks do not correspond to the  
456 specific ChemCam LIBS scene used for  $G_{\text{MEAN}}$ . The rocks of Confidence Hills are composed  
457 predominantly of grains below the resolution of MAHLI and RMI, but occasional sand-sized  
458 grains were discernible in MAHLI images. They represent mud-dominated rocks at PH. The  
459 rocks at Alexander Hills are mudstones and have silt to very fine sand sized voids; rare voids  
460 were more than a millimeter in diameter (Fig. 6a). The rocks of Book Cliffs and the upper  
461 section of Chinle, appear coarser-grained than mudstones observed elsewhere at PH, with

462 abundant discernible silt- and very fine sand-sized grains observed in MAHLI images. The rocks  
463 of the upper Chinle are cross-laminated supporting the presence of sand (Fig. 6b). Medium sand-  
464 sized protrusions are also present in the rocks at Book Cliffs, though they may be diagenetic  
465 features. These targets are used to define the lower and upper limits of rocks with silt to very fine  
466 sand, and the lower limit of rocks with fine sand. The three rock targets at the Whale Rock  
467 outcrop are the coarsest rocks analyzed by ChemCam at PH, and grains can be resolved in the  
468 RMI images. All Whale Rock targets have very poorly sorted medium to coarse sand with  
469 sparse very coarse sand in a matrix of finer sediment not resolved in the RMI or MAHLI images  
470 (Fig. 6c). These rocks help define  $G_{\text{MEAN}}$  values for rocks composed of medium to very coarse  
471 sand. The Whale rock outcrop has climbing-ripple cross-stratification supporting the presence of  
472 sand (Fig. 6d).

473

## 474 **5.2. Calibration of the GIMS to grain size**

475 The GIMS values of rock targets with grain sizes constrained by images were used to  
476 calibrate  $G_{\text{MEAN}}$  to grain size (Table 1). For these targets, the magnitude of  $G_{\text{MEAN}}$  increased  
477 consistently with grain size in both YKB and PH. Mudstones from the Sheepbed member have  
478 the lowest mean  $G_{\text{MEAN}}$ , at  $0.04 \pm 0.01$ , with a minimum  $G_{\text{MEAN}}$  value of  $0.02 \pm 0.00$  and maximum  
479 of  $0.06 \pm 0.00$ . Whereas the medium to coarse sandstones of the Whale Rock outcrop have the  
480 highest mean  $G_{\text{MEAN}}$  at  $0.23 \pm 0.05$ , with a minimum  $G_{\text{MEAN}}$  value of  $0.18 \pm 0.01$  and maximum of  
481  $0.29 \pm 0.01$ . Rocks with intermediate grain sizes have intermediate  $G_{\text{MEAN}}$  values. Also, rocks  
482 with the same grain size have similar  $G_{\text{MEAN}}$  in both YKB and PH. For example, the target  
483 Gillespie\_Lake\_1 from YKB and Vasquez from PH, have medium to coarse sand sized grains,  
484 and have  $G_{\text{MEAN}}$  values of  $0.16 \pm 0.00$  and  $0.18 \pm 0.01$ , respectively. Thus,  $G_{\text{MEAN}}$  values correlate  
485 well with grain sizes constrained from images.

486 Based on the  $G_{\text{MEAN}}$  of the standards, four grain size regimes in  $G_{\text{MEAN}}$  space (GSR1-4)  
487 were defined using a similar classification scheme as the Wentworth scale (Wentworth, 1992):  
488 clay to silt ( $G_{\text{MEAN}}=0.00-0.07$ ; GSR1), silt to fine sand ( $G_{\text{MEAN}}=0.08-0.11$ ; GSR2), fine to  
489 medium sand ( $G_{\text{MEAN}}=0.11-0.14$ ; GSR3), and medium to very coarse sand ( $G_{\text{MEAN}}=0.15-0.29$ ;  
490 GSR4). The GSR1, GSR2, and GSR4 bins have multiple calibration standards, and thus their  
491  $G_{\text{MEAN}}$  ranges are well constrained. The upper and lower  $G_{\text{MEAN}}$  bounds of these GSRs were  
492 defined by using the minimum and maximum  $G_{\text{MEAN}}$  of the calibration rocks in each bin (Table  
493 1). The lower bound of GSR1 was extended to 0.00 as that is the theoretical minimum for a  
494 homogenous rock (see Section 2.2). GSR3, unlike the other bins, is not well constrained because  
495 it has only one calibration rock (Nanok;  $G_{\text{MEAN}}=0.11$ ). Because the  $G_{\text{MEAN}}$  of Nanok overlaps  
496 with the upper bound of the pre-defined GSR2 bin, its value was chosen to be the lower bound of  
497 GSR3. Rocks with  $G_{\text{MEAN}}=0.11$  are exactly at the boundary and are reported as GSR2/GSR3.  
498 The upper bound of GSR3 was chosen to be the mid-point distance between the lower bound of  
499 GSR3 and that of GSR4. GSR3 is illustrated differently in figures 3, 4, and 8 to highlight its poor  
500 calibration.

501

## 502 **5.3. Inferred grain sizes using the GIMS**

503  $G_{\text{MEAN}}$  was calculated for 50 rocks from YKB and 15 from PH (Table 2). Of the 50 rocks  
504 in YKB, 16 are calibration standards, an additional 11 have known grain sizes from image data,  
505 and 23 have unknown grain sizes (Table 2). Of the 15 rocks from PH, 11 are calibration  
506 standards and 4 have unknown grain sizes (Table 2). All rocks were pre-screened to exclude

507 significant diagenetic features (see Section 2.2). Grain size ranges for all rocks were then  
508 assigned using the regimes from the calibration exercise (see Section 5.2).

509 At YKB, only the rocks from the Sheepbed member are in GSR1 (Table 2; Fig. 3). Rocks  
510 from Gillespie Lake and the Glenelg members are primarily in GSR3 and GSR4 (Table 2; Fig.  
511 3). Thus, there is a discontinuity in the predicted grain sizes of rocks between the Sheepbed  
512 mudstone and rest of YKB.

513 At PH, the grain size regime increases overall with increasing elevation from GSR1 at  
514 Confidence Hills to GSR4 at Whale Rock. This increasing upward trend deviates at Chinle  
515 where the  $G_{MEAN}$  is GSR1 at the base of the outcrop and increases to GSR2/GSR3 at the top.  
516 Most GSR4 rocks are in Whale Rock; the exception is Aztec\_2 from Alexander Hills.

517 In general, the Sheepbed mudstones have lower  $G_{MEAN}$  than those from Confidence Hills.  
518 Apart from one rock target, YKB also appears to be devoid of rocks in GSR2; although three are  
519 transitional between GSR2 and GSR3. Thus, at YKB, rocks are either in GSR1 or in GSR3 and  
520 4. The  $G_{MEAN}$  of rocks in GSR4 in PH is greater than those in GSR4 at YKB.

521 Some of the rocks at YKB have multiple LIBS analyses allowing an opportunity to test  
522 the reproducibility of the GIMS (Table 2). For example, at Gillespie Lake, the same rock was  
523 targeted twice using 3x3 rasters; Gillespie\_Lake\_1 was taken on the vertical face of the bedrock  
524 whereas Gillespie\_Lake\_2 was taken on the exposed top face. Both are in GSR4 (Fig. 7a). In the  
525 Point Lake outcrop, Balboa\_Dismal\_Lakes and Balboa2, are 3x3 rasters that overlap each other  
526 on the vertical face of the same rock and are in GSR3 and GSR2/GSR3, respectively. At the  
527 Rocknest outcrop, three rocks were targeted multiple times by the LIBS (Table 2). An example is  
528 a float rock that was targeted three times, twice by 1x10 vertical rasters, Rocknest\_3a and  
529 Rocknest\_3b, and once by a 1x5 diagonal raster, Rocknest3 (Fig. 7b). The Rocknest3a and  
530 Rocknest3b rasters formed a vertical transect from the bottom to top of the float rock, and are  
531 both in GSR4, whereas Rocknest3 was several millimeters to the left and below the previous  
532 rasters and is GSR3 (Fig. 7b).

533

#### 534 **5.4. Validation of GIMS results to grain size from images**

535 For all rocks with known grain size, their  $G_{MEAN}$  values were compared to the grain sizes  
536 measured from images (Fig. 8). Overall,  $G_{MEAN}$  increases with increasing grain size for rocks  
537 with mud to medium sand from 0.00-0.13 (Fig. 8). For rocks with medium sand to very fine  
538 gravel,  $G_{MEAN}$  varies between 0.15-0.29, and there are no apparent trends with grain size with  
539 increasing  $G_{MEAN}$  (Fig. 8).

540 The predicted GSRs of rocks are consistent with their grain size constrained from images  
541 (Fig. 8). All rocks composed of mud to very fine sand based on image data are calibration  
542 standards and are in GSR1 and GSR2 bins. Three rocks with known grain size are in GSR3,  
543 including the GSR3 calibration standard. The other two rocks in GSR3 have fine to medium  
544 sand, although one also has at least some coarse sand. All three GSR3 rocks have <5-10%  
545 resolvable grains in the RMI images, making visual grain size estimates biased toward coarser  
546 grains or those with more contrast. For the GSR4 bin, in addition to the four standards, there are  
547 ten other rocks with resolvable grains in images, and these all have grains that are coarser than  
548 medium sand. Out of the ten, three have <5-10% resolvable grains in the RMI images. Most of  
549 the GSR4 rocks are poorly to very poorly sorted based on visual grain size estimations.

550

## 551 **6. Discussion**

### 552 **6.1. GIMS as a proxy for grain size**

553 The GIMS is a robust technique for inferring grain size differences in the rocks from  
554 YKB and PH across regional and compositional differences. At both locations, mudstones have  
555 the smallest  $G_{\text{MEAN}}$  and medium to coarse sandstones have the largest  $G_{\text{MEAN}}$  (Fig. 3, Fig. 4, Fig.  
556 8; Table 2). GSRs were calibrated to a select number of rock standards and validated by applying  
557 the GIMS to other rocks with known grain size (Fig. 8). The predicted GSRs of rocks not used in  
558 the calibration are consistent with their grain size constrained from images (Fig. 8).

559 Overall, the GIMS successfully predicts the grain size of rocks of known grain size (Fig.  
560 3, Fig. 4, Fig. 8). In our dataset, the GIMS is particularly sensitive when rocks have grains  
561 smaller than medium sand and  $G_{\text{MEAN}} < 0.15$  (Fig. 8), because  $G_{\text{MEAN}}$  increases with increasing  
562 grain size. However, when rocks have grains that are medium sand to granule in size, the  $G_{\text{MEAN}}$   
563 rolls off instead of continuing to increase with grain size because medium sand to granule sized  
564 grains are all larger than the LIBS spot size; each LIBS spot samples only one or two grains (Fig.  
565 2; Section 2.2). Theoretically, rocks with grains up to the size of the LIBS spot spacing, typically  
566 5 mm, would produce similar  $G_{\text{MEAN}}$  values. Thus, rocks with grains ranging from medium sand  
567 to fine gravel in size could cause the same magnitude of variability from point-to-point and thus  
568 be part of GSR4. This insensitivity of  $G_{\text{MEAN}}$  to grain size explains the large bin size for GSR4  
569 compared to the other bins.

570 The ChemCam LIBS targeted some rocks more than once, allowing an opportunity to  
571 investigate the reproducibility of the GIMS (e.g., Fig. 7; Table 2). Commonly, the  $G_{\text{MEAN}}$  of the  
572 analyses are similar, with values falling within 0.03 of each other, placing them in the same GSR  
573 (Table 2). Results are particularly consistent for the rocks at PH and the Sheepbed member of  
574 YKB. However, GSRs predicted by the GIMS are variable for rocks in the Gillespie Lake and  
575 Glenelg members at YKB. Rocks from both members are predominantly poorly sorted and some  
576 are matrix supported (Grotzinger et al., 2014; 2015; Mangold et al., 2015; Anderson et al., 2015;  
577 Edgar et al., 2017). Since the GIMS is a statistical technique, if the LIBS analysis preferentially  
578 samples a subset of grain sizes, the analysis will not represent the rock or unit as a whole. This  
579 was the case for Gillespie\_Lake\_1, which is in a rock that has fine sand to medium gravel sized  
580 grains (Grotzinger et al., 2014; Mangold et al., 2015), but medium to coarse sand near the laser  
581 pits of the LIBS analysis (Fig. 5b). The GSR prediction of GSR4 for Gillespie\_Lake\_1 is  
582 consistent with the grain sizes sampled by LIBS, but not the rock as a whole. The variability in  
583  $G_{\text{MEAN}}$  between analyses on the same rock can be used to suggest poor sorting, or the data can be  
584 evaluated to see how many analyses are necessary to obtain a rigorous grain size prediction.  
585 Thus, to obtain a meaningful GRS prediction, significantly more data are required for poorly  
586 sorted or heterogeneous rocks than for ones with grains of uniform size. It is possible that the  
587 variability between some GSRs for the same rock is due to the poor calibration of the GSR3 bin.  
588 One of the Rocknest float rocks (Fig. 7b) has two analyses in GSR4 with  $G_{\text{MEAN}} = 0.15$  and 0.16,  
589 and one in GSR3 with a  $G_{\text{MEAN}} = 0.13$  (Table 2). Grain sizes were not resolved in this rock from  
590 image data. The variable GSR prediction may reflect either grain size variations or the poor  
591 calibration of the GSR3 bin. It is possible that the GSR3 bin spans a narrower range of  $G_{\text{MEAN}}$ ,  
592 and that the GSR4 bin extends to lower values, as there are no rocks with  $G_{\text{MEAN}}$  from 0.13-0.15  
593 with known grain size in our dataset.

594 One outcrop shows an inconsistency between the GSR inferred and image analysis. At  
595 PH, the Alexander Hills outcrop has a higher inferred GSR than is interpreted from image data.  
596 Clastic grains could not be resolved in the MAHLI images of rocks in this outcrop, suggesting  
597 that it is dominated by mud, but the  $G_{\text{MEAN}}$  results suggest a grain size of fine to medium sand.  
598 Voids ranging in size from silt/very fine sand to very coarse sand were resolved in images (Fig.

599 6a). These observations suggest that this outcrop either is a sandstone, or that there are  
600 unresolved diagenetic features present in the rock that increased compositional heterogeneity and  
601 thus led to a higher predicted GSR.

602 Variability in  $G_{\text{MEAN}}$  can arise from factors in addition to grain size and sorting. Rocks  
603 with grains that have a small range in composition will have low point-to-point variability and a  
604 low  $G_{\text{MEAN}}$  irrespective of grain size, whereas rocks with grains of diverse compositions will  
605 have high point-to-point variability and higher  $G_{\text{MEAN}}$  for a given grain size. Martian  
606 sedimentary rocks are generally expected to have a diverse suite of grain compositions, because  
607 Mars, unlike Earth, is mostly basaltic, with sediment grains consisting of pyroxene, feldspar,  
608 olivine, other mafic minerals, and glass. Basaltic minerals are the most abundant minerals  
609 present at YKB and PH based on ChemCam and the CheMin analyses (Sautter et al., 2014;  
610 Vaniman et al., 2013; Mangold et al., 2016; Cousin et al., 2017; Rampe et al., 2017). Even  
611 aeolian dunes, which are normally dominated by quartz on Earth, are composed of basaltic  
612 minerals on Mars (Lapotre et al., 2017). Because the GIMS is very sensitive to grain  
613 compositions, calibration relative to sampled compositions is critical for GSR predictions to be  
614 meaningful. To account for compositional variability between rocks, the major-element oxides  
615 for all rocks must be normalized together before calculating  $G_{\text{MEAN}}$  (see Appendix). This  
616 calibration takes into account that some oxides, such as CaO and MgO, vary more in magnitude  
617 between samples than other oxides, such as SiO<sub>2</sub> (see Section 3.2). The element variability  
618 normalization ensures that the contribution from each oxide to the overall variability of a rock is  
619 weighted to make the GIMS sensitive to grain size variations. If the LIBS data were not  
620 normalized, the  $G_{\text{MEAN}}$  of rocks would depend on both compositional variations and grain size,  
621 and the use of the GIMS would predict inaccurate grain sizes. Because of this need for  
622 compositional calibration, the calibration of  $G_{\text{MEAN}}$  to GSR presented in this manuscript is not  
623 directly comparable to results of past studies that have used the Gini index (e.g., Mangold et al.,  
624 2017) or future studies that use the GIMS without renormalization of the compositions.

625 Due to the sensitivity of the GIMS to compositional variations,  $G_{\text{MEAN}}$  can also be biased  
626 on rocks with a high proportion of diagenetic phases. LIBS spots might include variable  
627 proportions of diagenetic phases, making the heterogeneity higher or lower than that produced by  
628 grain size alone. This would be the case for coarse rocks with intra-granular cements, as the laser  
629 may hit different proportions of cement and grain at each spot. Since coarser grained rocks have  
630 more pore space to accommodate cements, contributions from cements may increase with  
631 increasing grain size. For rocks with grains smaller than the laser spot size (medium sand), this  
632 would provide a component with a consistent composition from point to point, and would not  
633 affect  $G_{\text{MEAN}}$ . For rocks with grains about the size or coarser than the laser spot size, the LIBS  
634 may sample cement in some raster points at grain boundaries, providing a variable contribution  
635 that could potentially increase  $G_{\text{MEAN}}$ . Thus, rocks with medium sand may end up in GSR4 rather  
636 than GSR3, whereas rocks with grains coarser than medium sand would remain in GSR4, due to  
637 the roll off in  $G_{\text{MEAN}}$  with increasing grain size (see above). Rocks that are poorly sorted would  
638 accommodate less cements than those that are better sorted and would be less affected by the  
639 cement contribution. Regardless of grain size and sorting, if the composition of the cement  
640 includes elements that are not one of the major-element oxides detected by ChemCam (e.g., SO<sub>4</sub>  
641 cement), then the filtering process would remove points with high cement contributions based on  
642 their sum of oxides. In contrast, the filtering process would not remove points with cement  
643 contributions that have compositions similar to the host rock.

644 Similarly, diagenetic features such as concretions can cause variability. Sand sized dark  
645 features, possibly micro-diagenetic concretions, are pervasive in rocks from Pink Cliffs,  
646 Shoemaker, Book Cliffs, and Telegraph Peak, and may be present at Alexander Hills. As these  
647 features are of the same scale as the LIBS spot, they would have caused variations consistent  
648 with sand. Differentiating between heterogeneities due to diagenetic contributions and grain size  
649 may be difficult when both are not resolvable or distinguishable in image data. However, more  
650 detailed analyses can improve interpretations. If variability is due to grain size, most major-  
651 element oxides should vary, whereas only certain oxides will vary for diagenetic contributions  
652 (e.g., Nachon et al., 2017). For example, high CaO is associated with CaSO<sub>4</sub> cement; when  
653 CaSO<sub>4</sub> is sampled, CaO is high with all other oxides showing a reduced contribution (e.g.,  
654 Nachon et al., 2017).

655 Where grains and diagenetic features cannot be distinguished, interpretations can be  
656 challenging. It is possible that chemical variability in rocks inferred to be in GSR2 could be due  
657 to diagenetic contributions and not grain size. Such an interpretation is difficult to test when  
658 sand or coarse silt cannot be verified in images and sedimentary structures associated with sand,  
659 such as cross-bedding, are not present. Silt to fine sand grain sizes are at or below the boundary  
660 of resolution for all of the cameras on the rover, and it is difficult to confirm visually the  
661 presence of grains in this size range. This grain size is also below the minimum LIBS laser spot  
662 size. However, for this study, the only rocks in GSR2 with known grain size are in the upper  
663 Chinle outcrop, which shows cross-bedding (Fig. 4b; see Section 6.2). The sedimentary  
664 structures support the GSR interpretation that there is sand present in these rocks. Several rocks  
665 from YKB with unknown grain size have  $G_{\text{MEAN}}$  that straddle the GSR2 and GSR3 bins. Due to  
666 their unknown grain size, it is uncertain whether their  $G_{\text{MEAN}}$  reflect grain size or the poor  
667 calibration of the boundaries for GSR3. The transition between GSR1 and GSR2 is important, as  
668 it may represent a change in flow regime for the sediments. GSR1 grain sizes are associated with  
669 suspended load transport and those of GSR2 with bedload (traction) transport.

670 Due to the statistical nature of the GIMS,  $G_{\text{MEAN}}$  also depends on the size of LIBS  
671 analysis spots and the spacing between these spots. The size of LIBS spots varies with rock  
672 hardness and distance between ChemCam and the rock (e.g., Maurice et al., 2012b), producing  
673 different analysis areas between observations. Each LIBS point covers a smaller area and thus  
674 fewer grains for short distance analyses versus long distance ones. Thus, it is predicted that  
675 sandstones with grain sizes on the order of LIBS spot sizes or smaller could show greater  
676 heterogeneity at shorter distances than longer ones. In contrast, rocks with mud-sized grains will  
677 consistently have little to no point-to-point heterogeneities because LIBS spots are always large  
678 enough to analyze a statistically significant number of grains. LIBS spot size and shape is also  
679 affected by rock hardness (Arvidson et al., 2014), which may produce minor variations in GIMS  
680 results. For rocks in our dataset with known grain size, distances from the rover varied  
681 (Supplemental Table 1) but no effects from grain size were identified. Based on the calibration  
682 data and evaluation, it appears that variations in the laser spot size are smaller than the breadth of  
683 the defined GSR and can be accounted for with detailed analysis of spot sizes if called for in  
684 future analyses.

685 In this study, the GSRs inferred from  $G_{\text{MEAN}}$  generally correlate very well with the grain  
686 sizes observed in images where data is available. Thus,  $G_{\text{MEAN}}$  provides an excellent estimate of  
687 grain size ranges for rocks lacking visible diagenetic features. For future studies using the GIMS,  
688 the accuracy of the predicted GSRs can be increased if more rocks with known grain size are  
689 calibrated to  $G_{\text{MEAN}}$ . This requires complimentary targeting of rocks with both MAHLI and



690 ChemCam in multiple regions along *Curiosity's* traverse, to be able to tie together detailed grain  
691 size information with point-to-point variabilities in ChemCam LIBS data. Rocks with fine to  
692 medium sand grains are particularly important to characterize as these are needed to constrain the  
693 extent of the GSR3 bin.

## 694 695 **6.2. New insights into depositional environments of Yellowknife Bay and Pahrump Hills**

### 696 *Vertical Trends in Stratigraphy*

697 The grain sizes at YKB are heterogeneous within the stratigraphic column (Fig. 3, Fig. 8).  
698 The discontinuity in grain size between the Sheepbed mudstone and rest of YKB sedimentary  
699 rocks demonstrate that flow characteristics shifted abruptly between accumulation of the  
700 Sheepbed mudstone and deposition of the overlying poorly sorted sandstones. The grain size of  
701 rocks at Point Lake and Rocknest outcrops previously were not well constrained from image  
702 data. For Point Lake, the mean  $G_{MEAN}$  is at the upper end of the fine to medium sand size range,  
703 with the majority of the rocks in GSR3 and GSR4. The variability of the  $G_{MEAN}$  between targets  
704 suggests that Point Lake rocks are poorly sorted. For Rocknest, the mean  $G_{MEAN}$  is at the lower  
705 end of the medium to very coarse sand size range, with the majority of the rocks in GSR4. Thus,  
706 the Rocknest outcrop is likely dominated by grains of medium sand and coarser. The lack of  
707 variability between the analyses suggests that the rocks are better sorted than those of Point  
708 Lake. Overall, the grain size variations at YKB are heterogeneous within the stratigraphic  
709 column (Fig. 3, Fig. 8), and the lack of rocks in GSR2 suggests bimodal flow conditions in the  
710 depositional environment. These results were consistent with deposition in an alluvial  
711 environment with a lake (Grotzinger et al., 2014).

712 At PH, there is a progressive increase in  $G_{MEAN}$  from mud to coarse sand through time  
713 (Fig. 4, Fig. 8), which supports prior interpretations that the PH sequence records progradation  
714 within a lacustrine depositional setting (Grotzinger et al., 2015; Stack et al., 2016). The overall  
715 trend is interrupted at Chinle, where grain sizes varied on the decimeter scale. The Chinle  
716 outcrop coarsens upward, and  $G_{MEAN}$  increases stratigraphically from mud to very fine to fine  
717 sand. From image data, the distribution and grain size of sand were difficult to constrain, but the  
718 presence of sand was inferred from cross-stratification in the outcrop (Fig. 6b). The  $G_{MEAN}$   
719 results show that the outcrop coarsens upward, which is consistent with increasing average flow  
720 speed through time. The accumulation of sediment, however, required that the instantaneous  
721 flow was slowing down at this location, which is consistent with deposition in a near-shore  
722 environment with a proximal fluvial influx of sediment. This interpretation fits the overall  
723 interpretation that the PH region accumulated as part of a delta.

### 724 725 *Mudstone Variations*

726 The mudstones at YKB and PH have similar mean  $G_{MEAN}$  values, but the ranges of  
727  $G_{MEAN}$  values are different. The mudstones of the Sheepbed (YKB) have much lower minimum  
728 values than those from Confidence Hills (PH), and Confidence Hills has one rock at the upper  
729 end of GSR1. The high  $G_{MEAN}$  values suggest that the rocks at Confidence Hills may include  
730 coarse silt and potentially dispersed sand grains. In contrast, the finer grain sizes of the Sheepbed  
731 member may be due to: 1) alteration and formation of clay minerals, as observed in drill analyses  
732 (Vaniman et al., 2013), which homogenized the chemical composition, or 2) a depositional  
733 environment with the accumulation of only the finest grain sizes.

## 734 735 **7. Concluding Remarks**

736 Overall, the GIMS provides a rigorous method for estimating grain size from chemical  
737 heterogeneities in ChemCam LIBS data. Its application requires a careful evaluation of the  
738 distribution and characteristics of available LIBS data that is tested against calibration images of  
739 rocks with known grain size. The calibration also includes the element variability normalization  
740 of all the LIBS data, to account for regional variations of major-element oxide weight fractions  
741 (see Appendix). All of the calibration steps are proposed as a standard procedure to use the  
742 GIMS, and they were validated by applying the GIMS to sedimentary rocks of various grain  
743 sizes from YKB and PH in Gale crater (Table 2; Fig. 8).

744 By providing grain size predictions, the GIMS expands the current use of the ChemCam  
745 LIBS instrument on the *Curiosity* rover. If used appropriately, the GIMS may be used to re-  
746 evaluate image-based grain size measurements and provide grain size constraints for regions in  
747 Gale crater that have incomplete textural information. The grain sizes inferred from the GIMS  
748 are complimentary to those determined from image data and together both techniques can be  
749 used to improve interpretations of the depositional environments of rocks analyzed by *Curiosity*  
750 and future Mars missions with LIBS, such as the Mars 2020 rover (Maurice et al., 2015; Wiens  
751 et al., 2017). Constraining the grain size of martian sedimentary rocks is crucial for interpreting  
752 ancient depositional environments and habitability of early Mars.

753

## 754 **8. Appendix: Calculating Gini index mean scores from ChemCam LIBS data**

755 The variability of each major-element oxide that ChemCam can detect goes into the  
756 GIMS calculation. The compositional ranges of major-element oxides need to be normalized to  
757 properly capture grain-size related variability. For example, oxide variations in quartzites are  
758 very different than those in basaltic sandstones. By normalizing oxide variations based on the  
759 total variability in the suite of rocks analyzed, the Gini mean score will be sensitive to changes in  
760 grain size. However, if rocks from regions with two distinctly different compositions are  
761 normalized together, the high variability across the sample suite can skew the variability caused  
762 by grain size. Thus, to calculate a meaningful Gini mean score, the weight percent range of each  
763 oxide within the sample suite must be known *a priori*, compositions should have a moderately  
764 narrow range, and the full range of oxides should be used in the normalization. The weight  
765 percent of each major-element oxide is normalized from 0 to 1 using *Equation 1*:

766

$$767 \quad z_{i,j} = \frac{x_{i,j} - \min(x_i)}{\max(x_i) - \min(x_i)}, \quad (1)$$

768

769 where  $z_{i,j}$  is the normalized weight percent for point  $j$ , and oxide  $i$ , and  $x_{i,j}$  is the oxide weight  
770 percent calculated from LIBS spectra. For any given target, the normalized weight percent values  
771 for each oxide are then binned using the same bin size. For this study, the bin size was set by  
772 averaging together the mean of 1/5 of the standard deviation for each oxide. The percentage of  
773 points per bin relative to the total number of points,  $P$ , is calculated. The sums of  $z_{i,j}$  in each bin  
774 and for the entire sample set are calculated, and the percent oxide sum for each bin,  $S$ , is  
775 tabulated. For each target and oxide,  $P$  is plotted in the x-axis and  $S$  in the y-axis to determine the  
776 Lorenz curve of the data set. The Gini index for each oxide is,

777

$$G_i = 1 - 2B_i, \quad (2)$$

778

779 where  $B_i$  is the area under the Lorenz curve for oxide  $i$ . The area can be calculated using a  
trapezoidal approximation. To obtain an overall homogeneity parameter, 7 major oxides were

780 averaged using an arithmetic mean to calculate an average  $G_{ave}$ . The maximum value that  $G_{ave}$   
781 can have depends on the number of points,  $N$ , per target, where

$$782 \quad G_N = \frac{(N-1)}{N} \quad (3)$$

783 (Mangold et al., 2017). To compensate for this variability,  $G_N$  for each target is used to  
784 normalize  $G_{ave}$ ,  
785

$$786 \quad G_{MEAN} = \frac{G_{ave}}{G_N} \cdot (3)$$

787  
788  $G_{MEAN}$ , the Gini mean score, can then be used to calibrate grain size ranges for the specific suite  
789 of samples being studied.

790 Discretion is needed when applying the GIMS, including filtering of data to use, choice  
791 of oxides to include, and bin size. In this study, some rock targets were excluded for the GIMS  
792 analysis primarily due to the presence or suspected presence of diagenetic features. Contributions  
793 from diagenetic features were suspected when the LIBS sum of oxides value was low, suggesting  
794 the presence of sulfates, chlorides, or other non-oxide minerals. Analysis of the sum of oxides  
795 versus the weight percent of CaO and MgO (oxides commonly associated with diagenesis) of  
796 different rock targets from YKB and PH suggested a conservative minimum threshold value of  
797 87% for the sum of oxides, and this value was picked for the GIMS analysis presented here. This  
798 threshold value should be evaluated for each suite of rocks that are being analyzed with the  
799 GIMS. Similarly, different sample suites may require the inclusion or exclusion of specific  
800 oxides. For this study,  $TiO_2$  was not included as a major oxide because it has low to no  
801 variability in most of the rock targets. However, when it does vary, the magnitude of variation is  
802 really high, skewing the  $G_{MEAN}$  values for some targets. The exclusion of  $TiO_2$  led to a  
803 significantly better grain size calibration. Finally, the bin size chosen for calculating  $G_i$  should  
804 depend on the purpose of the study. The bin size used in this study was chosen to provide good  
805 coverage of the observed grain sizes, which spanned from mud to gravel (see above). However,  
806 the appropriate bin size depends on the question being asked. For example, characterizing  
807 variability within mudstones and siltstones may require a finer bin size than used in this study  
808 (e.g., more bins), to pick out subtle point-to-point chemical variabilities.  
809

## 810 **9. Acknowledgements**

811 This research was funded by the Mars Science Laboratory Project through the NASA  
812 Mars Exploration Program and the Centre National d'Etudes Spatiales, France. Rivera-  
813 Hernandez was funded by the Chateaubriand STEM Fellowship sponsored by the Embassy of  
814 France in the United States. We are grateful to the MAHLI, Mastcam, and ChemCam teams for  
815 providing outstanding data on which to base this research. In particular, Rivera-Hernandez  
816 would like to thank the ChemCam science team for welcoming her to the team and providing  
817 constructive conversations on how to use, interpret, and access the ChemCam data.  
818

## 819 **10. References**

- 820 Anderson, R.B. & Bell III, J. F. (2010). Geologic mapping and characterization of Gale Crater  
821 and implications for its potential as a Mars Science Laboratory landing site. *Mars* 5, pp.  
822 76-128.  
823 Anderson, R.B., Morris, R.V., Clegg, S.M., Bell, J.F., Wiens, R.C., Humphries, S.D.,  
824 Mertzman, S.A., Graff, T.G. and McInroy, R. (2011). The influence of multivariate

825 analysis methods and target grain size on the accuracy of remote quantitative chemical  
826 analysis of rocks using laser induced breakdown spectroscopy. *Icarus*, 215(2), pp.608-  
827 627.

828 Anderson, R., Bridges, J.C., Williams, A., Edgar, L., Ollila, A., Williams, J., Nachon, M.,  
829 Mangold, N., Fisk, M., Schieber, J. and Gupta, S. (2015). ChemCam results from the  
830 Shaler outcrop in Gale crater, Mars. *Icarus*, 249, 2-21.

831 Anderson, D.E., Ehlmann, B.L., Forni, O., Clegg, S.M., Cousin, A., Thomas, N.H., Lasue, J.,  
832 Delapp, D.M., McInroy, R.E., Gasnault, O. and Dyar, M.D. (2017). Characterization of  
833 Laser-Induced Breakdown Spectroscopy (LIBS) emission lines for the identification of  
834 chlorides, carbonates, and sulfates in salt/basalt mixtures for the application to MSL  
835 ChemCam data. *Journal of Geophysical Research: Planets*.

836 Arvidson, R. E., Bellutta, P., Calef, F., Fraeman, A. A., Garvin, J. B., Gasnault, O., Grant, J. A.,  
837 Grotzinger, J. P., Hamilton, V. E., Heverly, M., Iagnemma, K. A., Johnson, J. R., Lanza,  
838 N., LeMouélic, S., Mangold, N., Ming, D. W., Mehta, M., Morris, R. V., Newsom, H. E.,  
839 Rennó, N., Rubin, D., Schieber, J., Sletten, R., Stein, N. T., Thuillier, F., Vasavada, A.  
840 R., Vizcaino, J., and Wiens, R. C. (2014). Terrain physical properties derived from  
841 orbital data and the first 360 sols of Mars Science Laboratory Curiosity rover  
842 observations in Gale Crater. *Journal of Geophysical Research: Planets*, 119(6), pp.1322-  
843 1344.

844 Banham, S. G., Gupta, S., Rubin, D.M., Watkins, J.A., Sumner, D.Y., Edgett, K.S., Grotzinger,  
845 J.P., Lewis, K.W., Edgar, L.A., Stack-Morgan, K.M., Barnes, R., Bell III, J.F., Day,  
846 M.D., Ewing, R.C., Lapotre, M.P.A, Stein, N.T., Rivera-Hernández, F., Vasavada, A.R.  
847 (2018) Sedimentology of an Ancient Aeolian Sandstone in the Lower Slope of Aeolis  
848 Mons, Stimson Formation, Gale Crater, Mars, *Sedimentology*.

849 Bell, J.F., Godber, A., McNair, S., Caplinger, M.A., Maki, J.N., Lemmon, M.T., Van Beek, J.,  
850 Malin, M.C., Wellington, D., Kinch, K.M. and Madsen, M.B. (2017). The Mars Science  
851 Laboratory Curiosity Rover Mast Camera (Mastcam) Instruments: Pre-Flight and  
852 In-Flight Calibration, Validation, and Data Archiving. *Earth and Space Science*.

853 Blaney, D.L., Wiens, R.C., Maurice, S., Clegg, S.M., Anderson, R.B., Kah, L.C., Le Mouélic,  
854 S., Ollila, A., Bridges, N., Tokar, R. and Berger, G. (2014). Chemistry and texture of the  
855 rocks at Rocknest, Gale Crater: Evidence for sedimentary origin and diagenetic  
856 alteration. *Journal of Geophysical Research: Planets*, 119(9), pp.2109-2131.

857 Clegg, S. M., Sklute, E., Dyar, M. D., Barefield, J. E., & Wiens, R. C. (2009). Multivariate  
858 analysis of remote laser-induced breakdown spectroscopy spectra using partial least  
859 squares, principal component analysis, and related techniques. *Spectrochimica Acta Part*  
860 *B: Atomic Spectroscopy*, 64(1), 79-88.

861 Clegg, S.M., Wiens, R.C., Anderson, R., Forni, O., Frydenvang, J., Lasue, J., Cousin, A., Payré,  
862 V., Boucher, T., Dyar, M.D. and McLennan, S.M. (2017). Recalibration of the Mars  
863 Science Laboratory ChemCam instrument with an expanded geochemical  
864 database. *Spectrochimica Acta Part B: Atomic Spectroscopy*, 129, pp.64-85.

865 Cousin, A., Forni, O., Maurice, S., Gasnault, O., Fabre, C., Sautter, V., Wiens, R.C. and  
866 Mazoyer, J. (2011). Laser induced breakdown spectroscopy library for the Martian  
867 environment. *Spectrochimica Acta Part B: Atomic Spectroscopy*, 66(11), pp.805-814.

868 Cousin, A., Sautter, V., Payré, V., Forni, O., Mangold, N., Gasnault, O., Le Deit, L., Johnson,  
869 J., Maurice, S., Salvatore, M. and Wiens, R.C. (2017). Classification of igneous rocks  
870 analyzed by ChemCam at Gale crater, Mars. *Icarus*, 288, pp.265-283.

871 Cremers D.A. and Radziemski L.J. (2013) Handbook of Laser-Induced Breakdown  
872 Spectroscopy, 2<sup>nd</sup> Edition, Wiley, doi: 10.1002/9781118567371.

873 Edgar, L.A., Gupta, S., Rubin, D.M., Lewis, K.W., Kocurek, G.A., Anderson, R.B., Bell, J.F.,  
874 Dromart, G., Edgett, K.S., Grotzinger, J.P. and Hardgrove, C. (2017). Shaler: In situ  
875 analysis of a fluvial sedimentary deposit on Mars. *Sedimentology*.

876 Edgett, K.S., Yingst, R.A., Ravine, M.A., Caplinger, M.A., Maki, J.N., Ghaemi, F.T.,  
877 Schaffner, J.A., Bell, J.F., Edwards, L.J., Herkenhoff, K.E. and Heydari, E. (2012).  
878 Curiosity's Mars hand lens imager (MAHLI) investigation. *Space science reviews*, 170(1-  
879 4), pp.259-317.

880 Edgett, K.S., Caplinger, M.A., Maki, J.N., Ravine, M.A., Ghaemi, F.T., McNair, S.,  
881 Herkenhoff, K.E., Duston, B.M., Willson, R.G., Yingst, R.A. and Kennedy, M.R.  
882 (2015). *Curiosity's robotic arm-mounted Mars Hand Lens Imager (MAHLI):*  
883 *Characterization and calibration status* (Vol. 1, No. 19, p. 2). MSL MAHLI Technical  
884 Report.

885 Fabre, C., S. Maurice, A. Cousin, R. C. Wiens, O. Forni, V. Sautter, and D. Guillaume.  
886 "Onboard calibration igneous targets for the Mars Science Laboratory Curiosity rover and  
887 the Chemistry Camera laser induced breakdown spectroscopy  
888 instrument." *Spectrochimica Acta Part B: Atomic Spectroscopy* 66, no. 3 (2011): 280-  
889 289.

890 Farmer, J. D., & Des Marais, D. J. (1999). Exploring for a record of ancient Martian  
891 life. *Journal of Geophysical Research: Planets*, 104(E11), 26977-26995.

892 Fraeman, A.A., Arvidson, R.E., Catalano, J.G., Grotzinger, J.P., Morris, R.V., Murchie, S.L.,  
893 Stack, K.M., Humm, D.C., McGovern, J.A., Seelos, F.P. and Seelos, K.D. (2013). A  
894 hematite-bearing layer in Gale Crater, Mars: Mapping and implications for past aqueous  
895 conditions. *Geology*, 41(10), pp.1103-1106.

896 Fraeman, A.A., Ehlmann, B.L., Arvidson, R.E., Edwards, C.S., Grotzinger, J.P., Milliken, R.E.,  
897 Quinn, D.P. and Rice, M.S. (2016). The stratigraphy and evolution of lower Mount Sharp  
898 from spectral, morphological, and thermophysical orbital data sets. *Journal of*  
899 *Geophysical Research: Planets*, 121(9), pp.1713-1736.

900 Jackson, R. S. (2016). Investigation of Aqueous Processes in the Valle Grande Paleo-Lake,  
901 Valles Caldera as a Martian Analog; Chemcam Investigation of the John Klein and  
902 Cumberland Drill Holes and Tailings, Gale Crater, Mars (master's thesis). University of  
903 New Mexico, Albuquerque, New Mexico.

904 Kah, L.C., Kronyak, R., Van Beek, J., Nachon, M., Mangold, N., Thompson, L., Wiens, R.,  
905 Grotzinger, J., Farmer, J., Minitti, M. and Schieber, J. (2015). Diagenetic crystal clusters  
906 and dendrites, lower Mount Sharp, Gale Crater.

907 Kronyak, R.E., Kah, L.C., Nachon, M., Mangold, N., Weins, R.C., Williams, R., Schieber, J.  
908 and Grotzinger, J. (2015, March). Distribution of mineralized veins from Yellowknife  
909 Bay to Mount Sharp, Gale Crater, Mars: Insight from textural and compositional  
910 variation. In *Abstract 1903 presented at Lunar and Planet. Sci. Conf* (Vol. 46).

911 Gini, C. (1921). Measurement of inequality of incomes. *The Economic Journal*, 31(121),  
912 pp.124-126.

913 Grant, J.A., Wilson, S.A., Mangold, N., Calef, F. and Grotzinger, J.P. (2014). The timing of  
914 alluvial activity in Gale crater, Mars. *Geophysical Research Letters*, 41(4), pp.1142-1149.

915 Grotzinger, J. P., Crisp, J., Vasavada, A. R., Anderson, R. C., et al. (2012). Mars Science  
916 Laboratory mission and science investigation. *Space science reviews*, 170(1-4), 5-56.

917 Grotzinger, J.P., Sumner, D.Y., Kah, L.C., Stack, K., Gupta, S., Edgar, L., Rubin, D., Lewis, K.,  
918 Schieber, J., Mangold, N. and Milliken, R. (2014). A habitable fluvio-lacustrine  
919 environment at Yellowknife Bay, Gale Crater, Mars. *Science*, 343(6169), p.1242777.

920 Grotzinger, J. P., Gupta, S., Malin, M. C., Rubin, D. M., Schieber, J., Siebach, K., et al. (2015).  
921 Deposition, exhumation, and paleoclimate of an ancient lake deposit, Gale crater, Mars.  
922 *Science*, 350(6257), aac7575.

923 Lasue, J., Mangold, N., Cousin, A., Meslin, P.Y., Wiens, R., Gasnault, O., Rapin, W., Schroder,  
924 S., Ollila, A., Fabre, C. and Berger, G. (2016, October). ChemCam analysis of martian  
925 fine dust. In *AAS/Division for Planetary Sciences Meeting Abstracts* (Vol. 48).

926 Le Deit, L., Hauber, E., Fueten, F., Pondrelli, M., Rossi, A.P. and Jaumann, R. (2013).  
927 Sequence of infilling events in Gale Crater, Mars: Results from morphology, stratigraphy,  
928 and mineralogy. *Journal of Geophysical Research: Planets*, 118(12), pp.2439-2473.

929 Le Mouélic, S., Gasnault, O., Herkenhoff, K.E., Bridges, N.T., Langevin, Y., Mangold, N.,  
930 Maurice, S., Wiens, R.C., Pinet, P., Newsom, H.E. and Deen, R.G. (2015). The  
931 ChemCam Remote Micro-Imager at Gale crater: Review of the first year of operations on  
932 Mars. *Icarus*, 249, pp.93-107.

933 Leveille, R.J., Bridges, J., Wiens, R.C., Mangold, N., Cousin, A., Lanza, N., Forni, O., Ollila,  
934 A., Grotzinger, J., Clegg, S. and Siebach, K. (2014). Chemistry of fracture-filling raised  
935 ridges in Yellowknife Bay, Gale Crater: Window into past aqueous activity and  
936 habitability on Mars. *Journal of Geophysical Research: Planets*, 119(11), pp.2398-2415.

937 Malin, M.C. and Edgett, K.S., 2000. Sedimentary rocks of early Mars. *Science*, 290(5498),  
938 pp.1927-1937.

939 Malin, M.C., Ravine, M.A., Caplinger, M.A., Ghaemi, F.T., Schaffner, J.A., Maki, J.N., Bell,  
940 J.F., Cameron, J.F., Dietrich, W.E., Edgett, K.S. and Edwards, L.J. (2016). The Mars  
941 Science Laboratory (MSL) Mast cameras and Descent imager: I. Investigation and  
942 instrument descriptions. *Earth and Space Science*.

943 Mangold, N., Forni, O., Dromart, G., Stack, K., Wiens, R.C., Gasnault, O., Sumner, D.Y.,  
944 Nachon, M., Meslin, P.Y., Anderson, R.B. and Barraclough, B. (2015). Chemical  
945 variations in Yellowknife Bay formation sedimentary rocks analyzed by ChemCam on  
946 board the Curiosity rover on Mars. *Journal of Geophysical Research: Planets*, 120(3),  
947 pp.452-482.

948 Mangold, N., Thompson, L.M., Forni, O., Williams, A.J., Fabre, C., Le Deit, L., Wiens, R.C.,  
949 Williams, R., Anderson, R.B., Blaney, D.L. and Calef, F. (2016). Composition of  
950 conglomerates analyzed by the Curiosity rover: Implications for Gale Crater crust and  
951 sediment sources. *Journal of Geophysical Research: Planets*, 121(3), pp.353-387.

952 Mangold, N., Schmidt, M. E., Fisk, M. R., Forni, O., McLennan, S. M., Ming, D. W., Sautter,  
953 V., Sumner, D., et al. (2017). Classification scheme for sedimentary and igneous rocks in  
954 Gale crater, Mars. *Icarus*, 284, 1-17.

955 Maurice, S., Wiens, R. C., Saccoccio, M., Barraclough, B., Gasnault, O., Forni, O., et al.  
956 (2012a). The ChemCam instrument suite on the Mars Science Laboratory (MSL) rover:  
957 Science objectives and mast unit description. *Space science reviews*, 170(1-4), 95-166.

958 Maurice, S., Cousin, A., Wiens, R.C., Gasnault, O., Parès, L., Forni, O., Meslin, P.Y., Clegg, S.  
959 and Team, C. (2012b), March. Laser Induced Breakdown Spectroscopy (LIBS) spot size  
960 at Stand-off distances with ChemCam. In *Lunar and Planetary Science Conference* (Vol.  
961 43).

962 Maurice, S., Wiens, R.C., Anderson, R., Beyssac, O., Bonal, L., Clegg, S., DeFlores, L.,  
963 Dromart, G., Fischer, W., Forni, O. and Gasnault, O. (2015, March). Science objectives  
964 of the SuperCam instrument for the Mars2020 rover. In *Lunar and Planetary Science*  
965 *Conference* (Vol. 46, p. 2818).

966 McCanta, M. C., Dobosh, P. A., Dyar, M. D., & Newsom, H. E. (2013). Testing the veracity of  
967 LIBS analyses on Mars using the LIBSSIM program. *Planetary and Space Science*, 81,  
968 48-54.

969 McCanta, M.C., Dyar, M.D., Dobosh, P.A., Davidson, G.R., Hill, C.A., Wolgemuth, K.,  
970 Romanowicz, B., Hirschmann, M., Kellogg, L., Manga, M. and Mukhopadhyay, S.  
971 (2017). Extracting Bulk Rock Properties from Microscale Measurements: Subsampling  
972 and Analytical Guidelines. *GSA Today*, 27(7).

973 McLennan, S. M. (2012). Geochemistry of sedimentary processes on Mars. *Sedimentary*  
974 *Geology of Mars*, 102, 119-138.

975 McLennan, S.M., Anderson, R.B., Bell, J.F., Bridges, J.C., Calef, F., Campbell, J.L., Clark,  
976 B.C., Clegg, S., Conrad, P., Cousin, A. and Des Marais, D.J. (2014). Elemental  
977 geochemistry of sedimentary rocks at Yellowknife Bay, Gale crater,  
978 Mars. *Science*, 343(6169), p.1244734.

979 Milliken, R.E., Grotzinger, J.P. and Thomson, B.J. (2010). Paleoclimate of Mars as captured by  
980 the stratigraphic record in Gale Crater. *Geophysical Research Letters*, 37(4).

981 Ming, D.W., Archer, P.D., Glavin, D.P., Eigenbrode, J.L., Franz, H.B., Sutter, B., Brunner,  
982 A.E., Stern, J.C., Freissinet, C., McAdam, A.C. and Mahaffy, P.R. (2014). Volatile and  
983 organic compositions of sedimentary rocks in Yellowknife Bay, Gale Crater,  
984 Mars. *Science*, 343(6169), p.1245267.

985 Nachon, M., Clegg, S.M., Mangold, N., Schröder, S., Kah, L.C., Dromart, G., Ollila, A.,  
986 Johnson, J.R., Oehler, D.Z., Bridges, J.C. and Le Mouélic, S. (2014). Calcium sulfate  
987 veins characterized by ChemCam/Curiosity at Gale crater, Mars. *Journal of Geophysical*  
988 *Research: Planets*, 119(9), pp.1991-2016.

989 Nachon, M., Mangold, N., Forni, O., Kah, L.C., Cousin, A., Wiens, R.C., Anderson, R., Blaney,  
990 D., Blank, J.G., Calef, F. and Clegg, S.M. (2017). Chemistry of diagenetic features  
991 analyzed by ChemCam at Pahrump Hills, Gale crater, Mars. *Icarus*, 281, pp.121-136.

992 Nellessen, M.A., Baker, A.M., Newsom, H.E., Jackson, R.S., Nachon, M., Rivera-Hernandez,  
993 F., Williams, J., Wiens, R.C., Frydenvang, J., Gasda, P., Lanza, N., Ollila, A., Clegg, S.,  
994 Gasnault, O., Maurice, S., Meslin, P.Y., Cousin, A., Rapin, W., Lasue, J., Forni, O.,  
995 L'Haridon, J, Blaney, D., Payré, V., Mangold, N., LeDeit, L., Edgett, K., Anderson, R.  
996 Distribution and Analysis of Calcium Sulfate-Cemented Sandstones along the MSL  
997 Traverse, Gale Crater, Mars. (March, 2018). In *Abstract 2858 presented at Lunar and*  
998 *Planet. Sci. Conf* (Vol. 49).

999 Newsom, H., Edgett, K., Wiens, R., Mangold, N., Schieber, J., Stack, K., Rapin, W., Stein, N.,  
1000 Rivera-Hernandez, F. (October, 2017). Imaging and chemical signatures of sandstone  
1001 cemented by calcium sulfate, in the Stimson and Murray formation rocks of Gale Crater,  
1002 Mars, International Meeting of Sedimentology 33, Toulouse, France.

1003 Palucis, M.C., Dietrich, W.E., Hayes, A.G., Williams, R.M., Gupta, S., Mangold, N., Newsom,  
1004 H., Hardgrove, C., Calef, F. and Sumner, D.Y. (2014). The origin and evolution of the  
1005 Peace Vallis fan system that drains to the Curiosity landing area, Gale Crater,  
1006 Mars. *Journal of Geophysical Research: Planets*, 119(4), pp.705-728.

1007 Rampe, E.B., Ming, D.W., Blake, D.F., Bristow, T.F., Chipera, S.J., Grotzinger, J.P., Morris,  
1008 R.V., Morrison, S.M., Vaniman, D.T., Yen, A.S. and Achilles, C.N. (2017). Mineralogy  
1009 of an ancient lacustrine mudstone succession from the Murray formation, Gale crater,  
1010 Mars. *Earth and Planetary Science Letters*.

1011 Sautter, V., Fabre, C., Forni, O., Toplis, M.J., Cousin, A., Ollila, A.M., Meslin, P.Y., Maurice,  
1012 S., Wiens, R.C., Baratoux, D. and Mangold, N. (2014). Igneous mineralogy at Bradbury  
1013 Rise: The first ChemCam campaign at Gale crater. *Journal of Geophysical Research:*  
1014 *Planets*, 119(1), pp.30-46.

1015 Siebach, K.L., Grotzinger, J.P., Kah, L.C., Stack, K.M., Malin, M., L veill , R. and Sumner,  
1016 D.Y. (2014). Subaqueous shrinkage cracks in the Sheepbed mudstone: Implications for  
1017 early fluid diagenesis, Gale Crater, Mars. *Journal of Geophysical Research:*  
1018 *Planets*, 119(7), pp.1597-1613.

1019 Siebach, K.L., Baker, M.B., Grotzinger, J.P., McLennan, S.M., Gellert, R., Thompson, L.M.  
1020 and Hurowitz, J.A. (2017). Sorting out compositional trends in sedimentary rocks of the  
1021 Bradbury group (Aeolis Palus), Gale crater, Mars. *Journal of Geophysical Research:*  
1022 *Planets*, 122(2), pp.295-328.

1023 Sivakumar, P., Taleh, L., Markushin, Y., & Melikechi, N. (2014). Packing density effects on the  
1024 fluctuations of the emission lines in laser-induced breakdown  
1025 spectroscopy. *Spectrochimica Acta Part B: Atomic Spectroscopy*, 92, 84-89.

1026 Stack, K.M., Grotzinger, J.P., Kah, L.C., Schmidt, M.E., Mangold, N., Edgett, K.S., Sumner,  
1027 D.Y., Siebach, K.L., Nachon, M., Lee, R. and Blaney, D.L. (2014). Diagenetic origin of  
1028 nodules in the Sheepbed member, Yellowknife Bay formation, Gale crater, Mars. *Journal*  
1029 *of Geophysical Research: Planets*, 119(7), pp.1637-1664.

1030 Stack, K.M., Grotzinger, J.P., Edgett, K.S., Gupta, S., Kah, L.C., Lamb, M.P, Lewis, K.W.,  
1031 Rubin, D.M., Schieber, J., Sumner, D.Y. (2016, September) Facies analysis and  
1032 stratigraphic context of the Pahrump Hills outcrop, type locality of the basal Murray formation,  
1033 Gale Crater, Mars. Geological Society of America Abstract, Vol. 48, No. 7.

1034 Summons, R.E., Amend, J.P., Bish, D., Buick, R., Cody, G.D., Des Marais, D.J., Dromart, G.,  
1035 Eigenbrode, J.L., Knoll, A.H. and Sumner, D.Y. (2011). Preservation of martian organic  
1036 and environmental records: final report of the Mars Biosignature Working  
1037 Group. *Astrobiology*, 11(2), pp.157-181.

1038 Thomson, B.J., Bridges, N.T., Milliken, R., Baldrige, A., Hook, S.J., Crowley, J.K., Marion,  
1039 G.M., de Souza Filho, C.R., Brown, A.J. and Weitz, C.M. (2011). Constraints on the  
1040 origin and evolution of the layered mound in Gale Crater, Mars using Mars  
1041 Reconnaissance Orbiter data. *Icarus*, 214(2), pp.413-432.

1042 Vaniman, D., Dyar, M.D., Wiens, R., Ollila, A., Lanza, N., Lasue, J., Rhodes, J.M., Clegg, S.  
1043 and Newsom, H. (2012). Ceramic ChemCam calibration targets on Mars science  
1044 laboratory. *Space science reviews*, 170(1-4), pp.229-255.

1045 Vaniman, D.T., Bish, D.L., Ming, D.W., Bristow, T.F., Morris, R.V., Blake, D.F., Chipera, S.J.,  
1046 Morrison, S.M., Treiman, A.H., Rampe, E.B. and Rice, M. (2013). Mineralogy of a  
1047 mudstone at Yellowknife Bay, Gale crater, Mars. *Science*, p.1243480.

1048 Vasavada, A.R., Grotzinger, J.P., Arvidson, R.E., Calef, F.J., Crisp, J.A., Gupta, S., Hurowitz,  
1049 J., Mangold, N., Maurice, S., Schmidt, M.E. and Wiens, R.C. (2014). Overview of the  
1050 Mars Science Laboratory mission: Bradbury landing to Yellowknife Bay and  
1051 beyond. *Journal of Geophysical Research: Planets*, 119(6), pp.1134-1161.

1052 Wentworth, C.K. (1922). A scale of grade and class terms for clastic sediments. *The Journal of*  
1053 *Geology*, 30(5), pp.377-392.



1054 Wiens, R.C., Maurice, S., Barraclough, B., Saccoccio, M., Barkley, W.C., Bell, J.F., Bender, S.,  
1055 Bernardin, J., Blaney, D., Blank, J. and Bouyé, M., et al. (2012). The ChemCam  
1056 instrument suite on the Mars Science Laboratory (MSL) rover: Body unit and combined  
1057 system tests. *Space Science Reviews*, 170(1-4), pp.167-227.

1058 Wiens, R. C., Maurice, S., Lasue, J., Forni, O., Anderson, R. B., Clegg, S., et al. (2013). Pre-  
1059 flight calibration and initial data processing for the ChemCam laser-induced breakdown  
1060 spectroscopy instrument on the Mars Science Laboratory rover. *Spectrochimica Acta Part*  
1061 *B: Atomic Spectroscopy*, 82, 1-27.

1062 Wiens R.C., Maurice S., and Rull Perez F. (2017) The SuperCam remote sensing instrument  
1063 suite for the Mars 2020 rover mission: A preview. *Spectroscopy* 32(5), 50-55.

1064 Williams, R.M., Grotzinger, J.P., Dietrich, W.E., Gupta, S., Sumner, D.Y., Wiens, R.C.,  
1065 Mangold, N., Malin, M.C., Edgett, K.S., Maurice, S. and Forni, O. (2013). Martian  
1066 fluvial conglomerates at Gale crater. *science*, 340(6136), pp.1068-1072.

1067 Yingst, R. A., Edgett, K. S., Kennedy, M. R., Krezoski, G. M., McBride, M. J., Minitti, M. E.,  
1068 Ravine, M. A., and Williams, R. M. E. (2016). MAHLI on Mars: lessons learned  
1069 operating a geoscience camera on a landed payload robotic arm. *Geoscientific*  
1070 *Instrumentation, Methods and Data Systems*, 5, 205-217.

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## 1100 11. Figure Captions

1101

1102 **Figure 1.** a) A mosaic of Gale Crater using images taken by the Mars Reconnaissance Orbiter  
1103 Context Camera. A list of the images used in this mosaic is provided by Anderson and Bell  
1104 (2010). The black box highlights the area shown in Figure 1b. b) A mosaic of the *Curiosity* field  
1105 area using images taken by the High Resolution Imaging Science Experiment camera on the  
1106 Mars Reconnaissance Orbiter. The white line shows the rover traverse path between landing and  
1107 sol 1850. The yellow star marks the landing site of *Curiosity*, the orange star marks the location  
1108 of Yellowknife Bay, and the red star that of Pahrump Hills. The green highlighted area is the  
1109 mapped aerial extent of the Murray formation from Fraeman et al. (2016).

1110

1111 **Figure 2.** Schematic showing the relationship between the LIBS laser spot diameter and grain  
1112 size. Rocks with grains smaller than the laser spot size yield the bulk rock compositions at each  
1113 LIBS spot, whereas those with grains about the size or larger can have chemical contributions  
1114 from individual grains at each spot, producing non-uniform compositions.

1115

1116 **Figure 3.**  $G_{\text{MEAN}}$  for the rocks used in the GIMS analysis plotted with the stratigraphic column of  
1117 the Yellowknife Bay formation constructed using outcrop elevation (Grotzinger et al., 2014).  
1118 The four GSRs were defined during the calibration procedure (see Section 5.2). GSR3 has a  
1119 hashed pattern because its bounds are not well constrained.

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1121 **Figure 4.**  $G_{\text{MEAN}}$  for the rocks from Pahrump Hills used in the GIMS analysis plotted with the  
1122 stratigraphic column of the Pahrump Hills member of the Murray formation constructed using  
1123 outcrop elevation (Stack et al., 2016). The four GSRs were defined during the calibration  
1124 procedure (see Section 5.2). GSR3 has a hashed pattern because its bounds are not well  
1125 constrained.

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1127 **Figure 5.** Cropped RMI image mosaics of the ChemCam targets Nanok (a), Gillespie\_Lake\_1  
1128 (b), and Wakham\_Bay (c). The red crosshairs mark the locations of LIBS laser spots on the rock  
1129 targets. The low depth of field of RMI images results in some parts of the mosaics being out of  
1130 focus. For Wakham\_Bay (c), the laser hit a very coarse sand sized grain at point 12 and a granule  
1131 at point 15.

1132

1133 **Figure 6.** a) Cropped RMI image of the Aztec\_2 ChemCam target. Orange arrows highlight sand  
1134 sized voids. b) Oblique MAHLI image of the upper Chinle outcrop, showing low angle cross-  
1135 stratification, marked by the white arrows. MAHLI product 0828MH0004520020301655C00. c)  
1136 Cropped MAHLI image of the Whale Rock outcrop. Sand grains can clearly be resolved in the  
1137 image. MAHLI product 0860MH0004580000302120R00. d) Mastcam (M-100) mosaic of the  
1138 Whale Rock outcrop. White arrows highlight climbing-ripple cross-stratification. Mastcam  
1139 products 0796MR0034760210500167E01 and 0796MR0034760220500168E01.

1140

1141 **Figure 7.** a) Cropped Mastcam (M-34) of the rock with the Gillespie\_Lake\_1 and  
1142 Gillespie\_Lake\_2 analyses. Mastcam product 0132ML0008020000103972E01. b) Cropped  
1143 Mastcam (M-100) of the rock with the Rocknest\_3, Rocknest\_3a, and Rocknest\_3b analyses.  
1144 Mastcam product 0086MR0003750000104151E0. For both (a) and (b) the approximate locations  
1145 of the LIBS analyses are highlighted by white rectangles.

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**Figure 8.** Grain size from image-data versus  $G_{MEAN}$  for the rocks of the Yellowknife Bay formation and Pahrump Hills member. The four GSRs were defined during the calibration procedure (see Section 5.2). GSR3 has a hashed pattern because its bounds are not well constrained. Each data point includes  $1\sigma$  STD<sub>r</sub> horizontal error bars and a grey vertical box showing the range of observed grain sizes in image data. Most samples may include finer grain sizes that could not be resolved in the images.

1192 **12. Supplemental tables**

1193

1194 **Supplemental Table 1.** The ChemCam LIBS data used in the GIMS analysis. Only data that  
1195 passed through the filtering procedure described in Section 2.2 are included. The accuracy (oxide  
1196 RMSEP) and precision (oxide\_shots\_stdev) are included for every major-element oxide per  
1197 LIBS point.

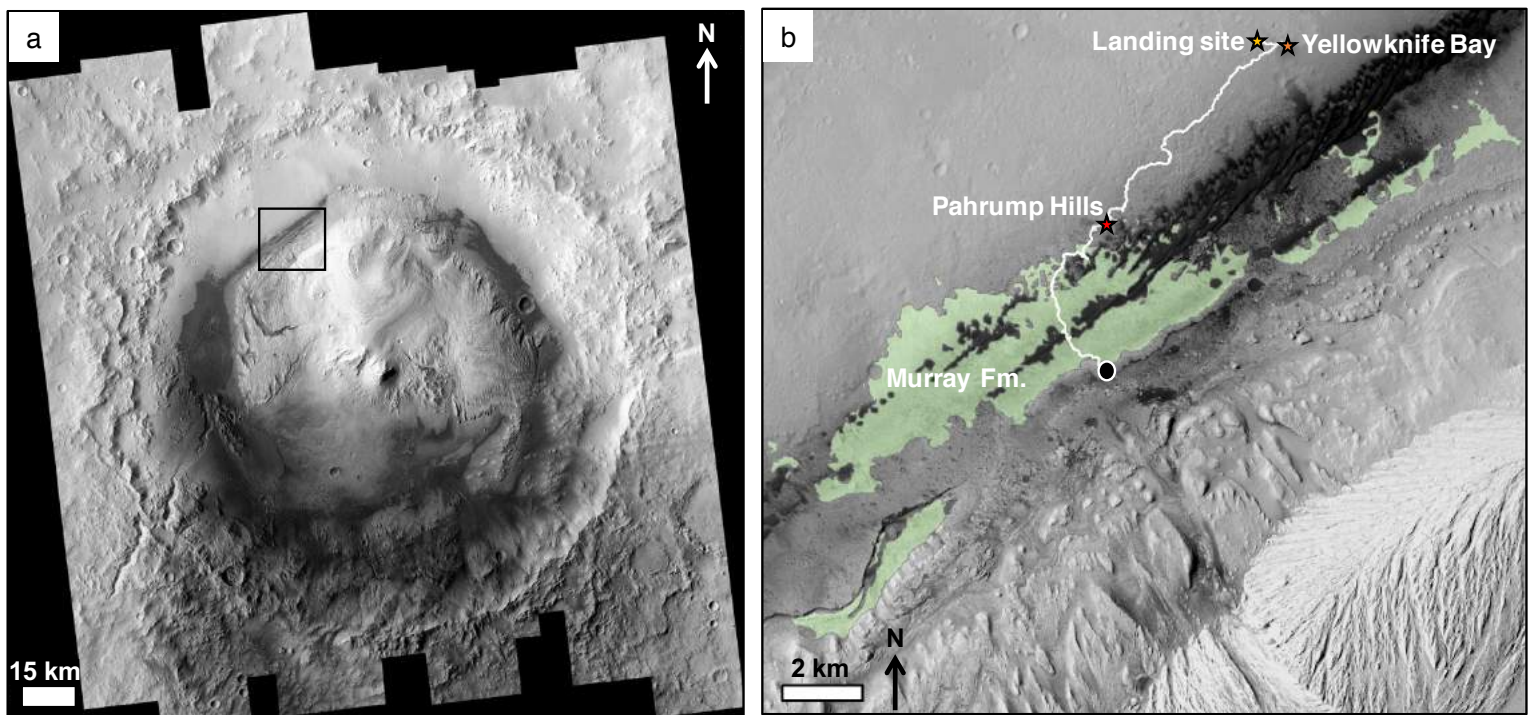


Figure 1.

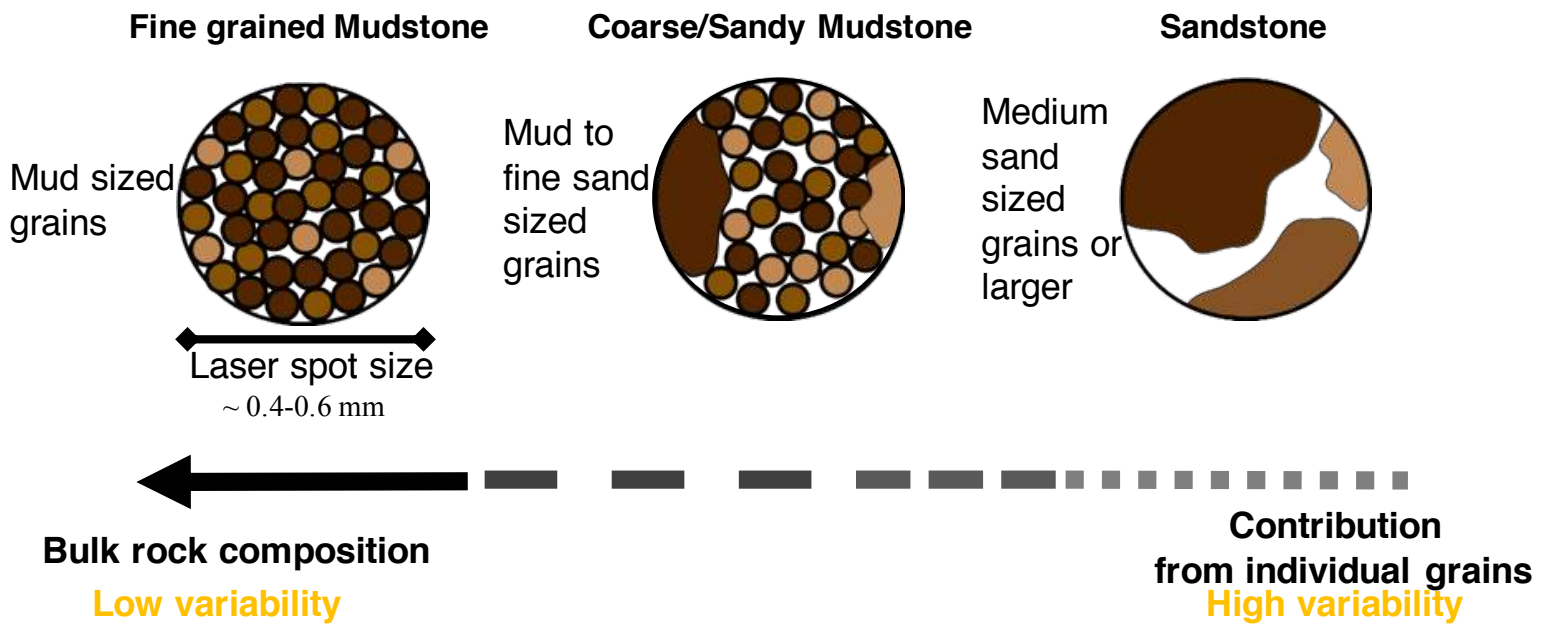
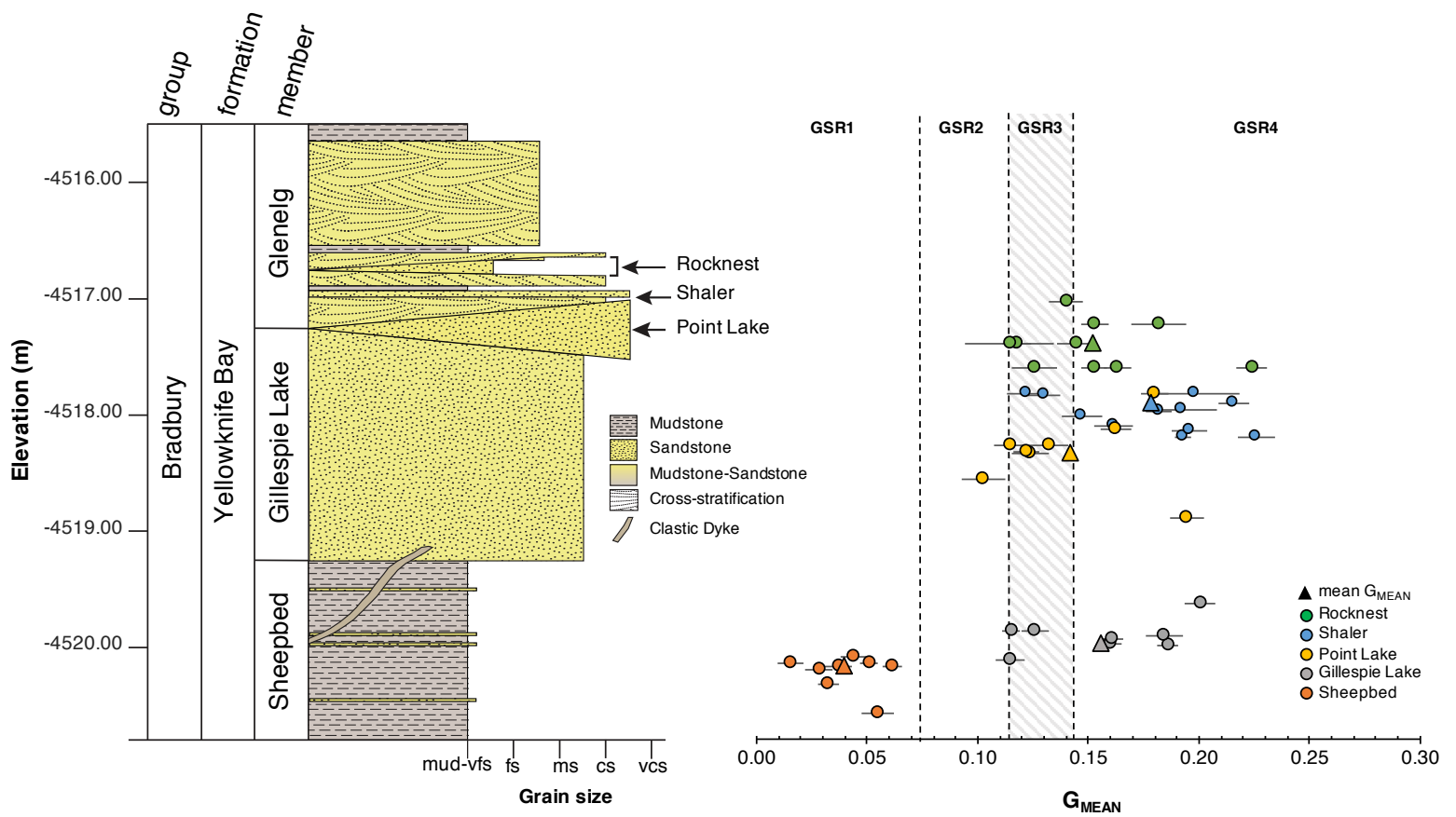
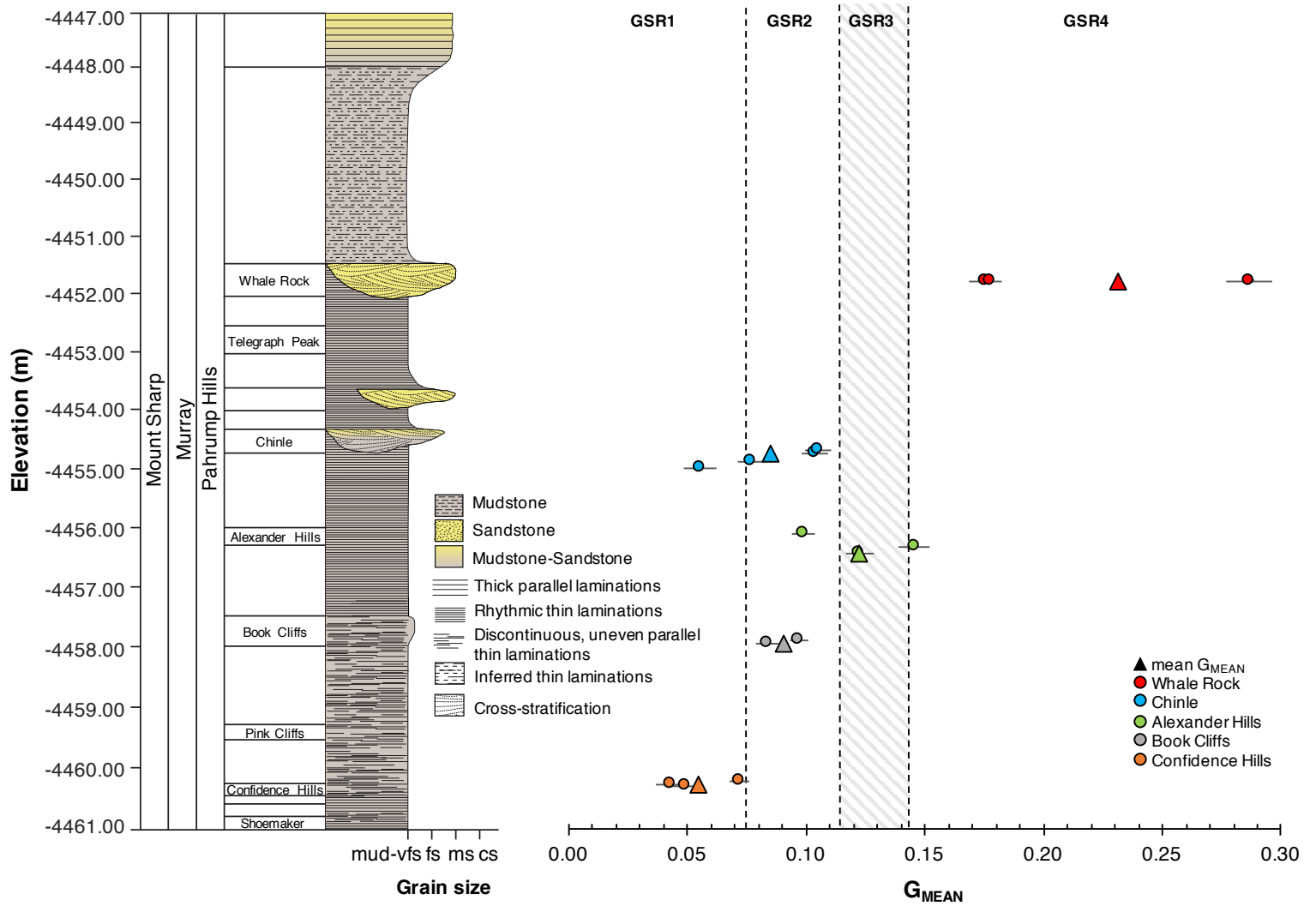


Figure 2.



**Figure 3.**



**Figure 4.**



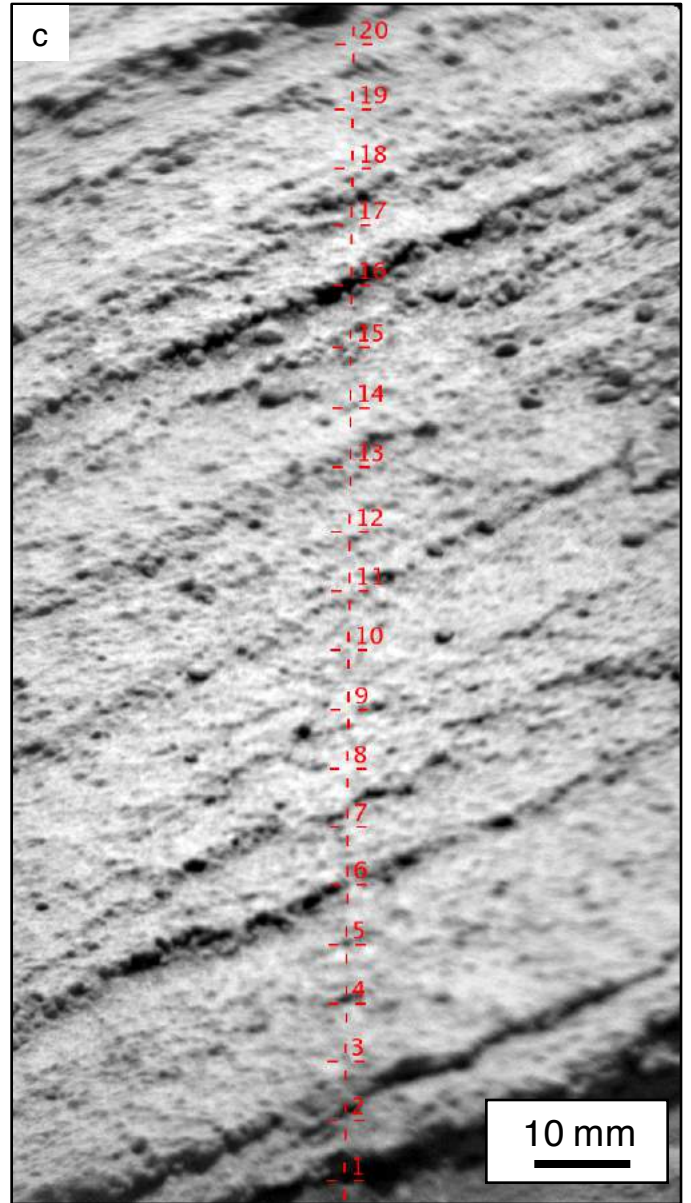
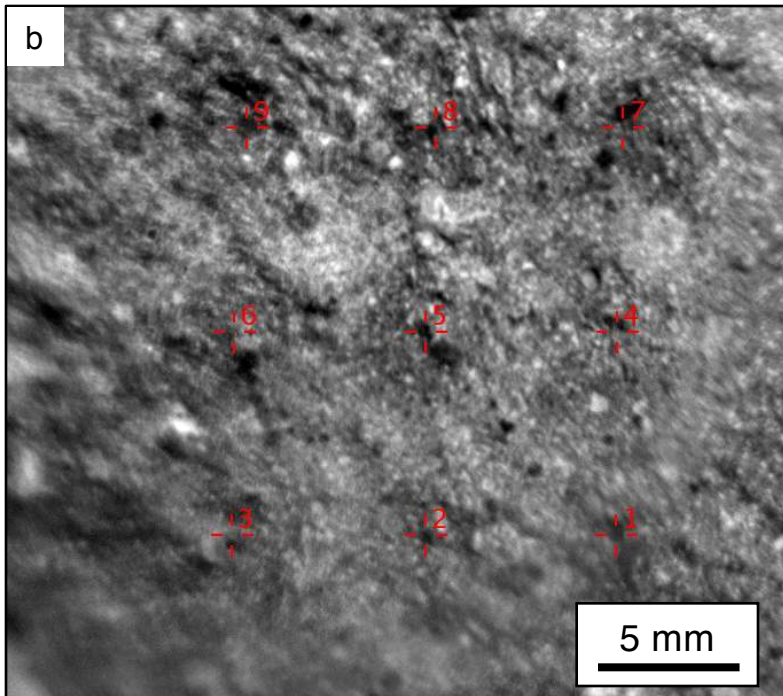
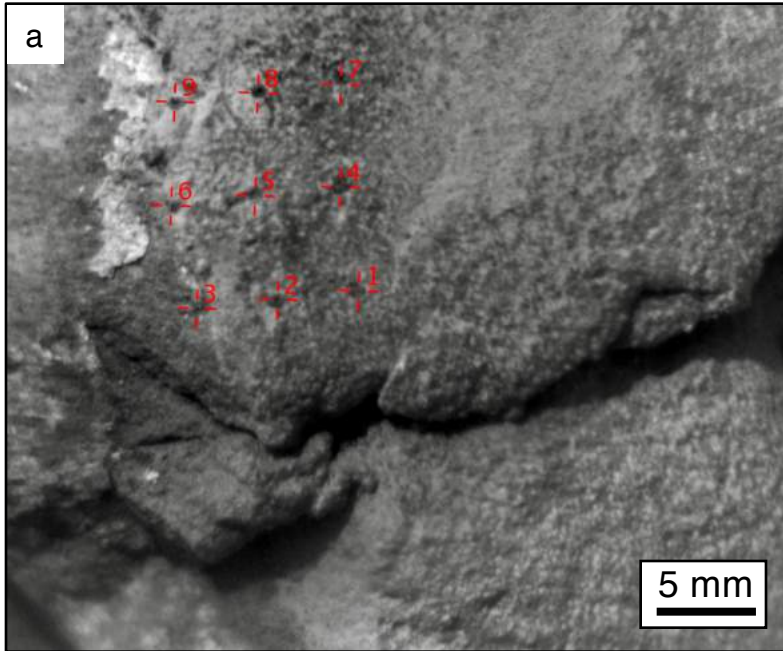


Figure 5.

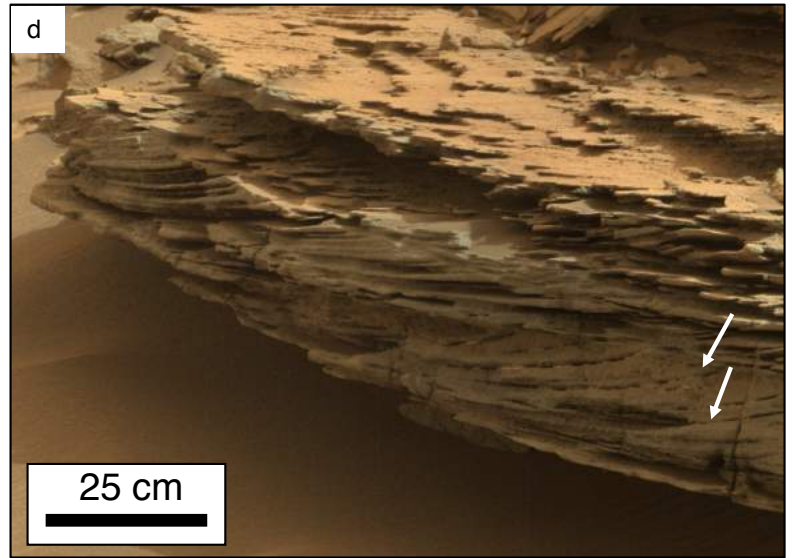
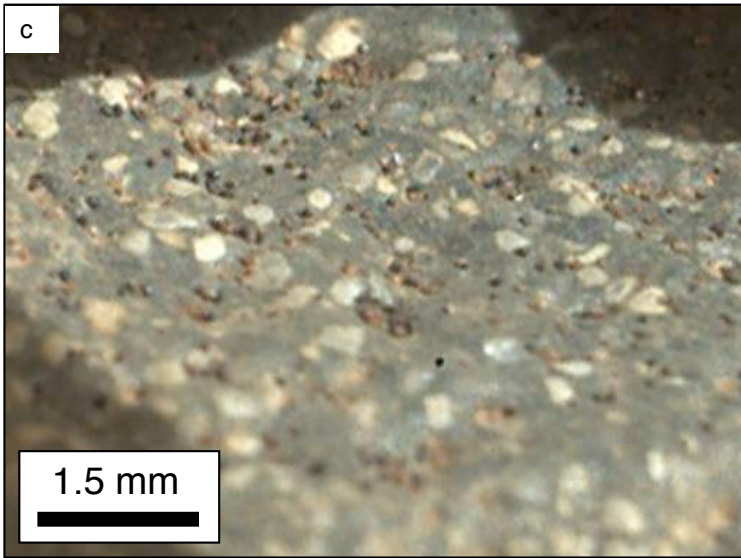
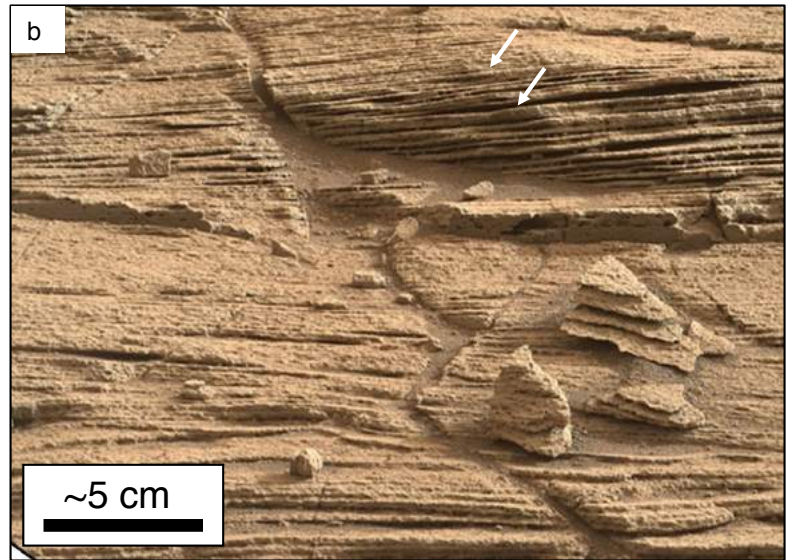
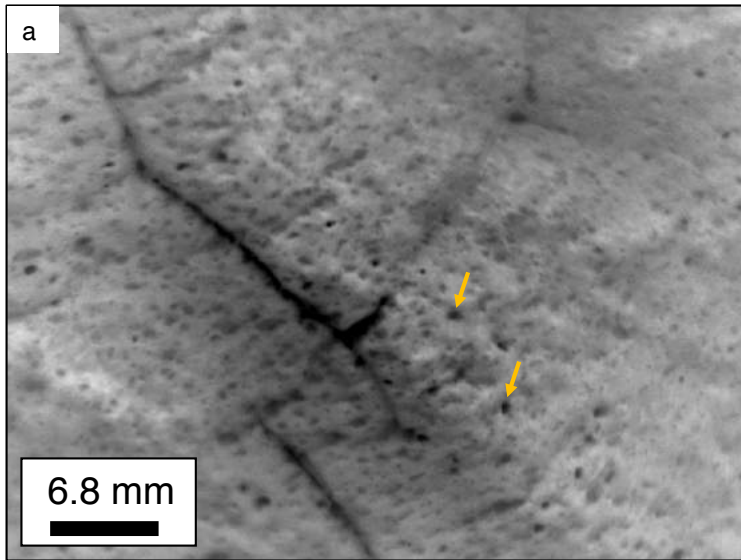
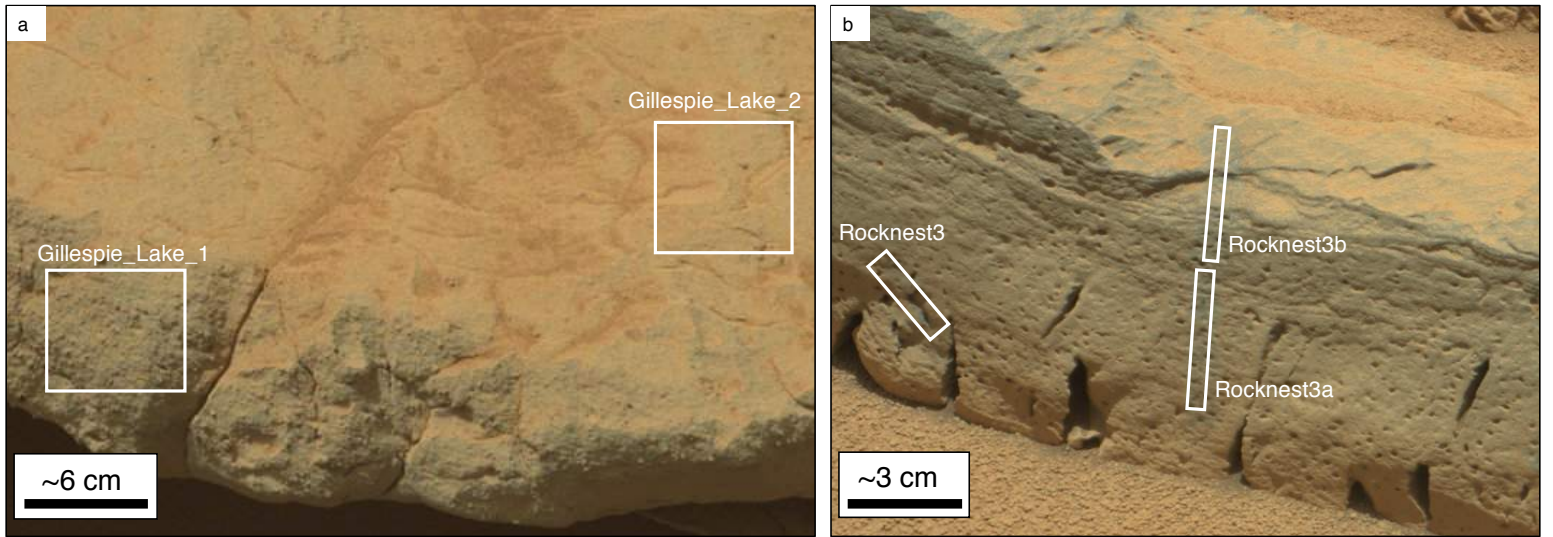


Figure 6.





**Figure 7.**

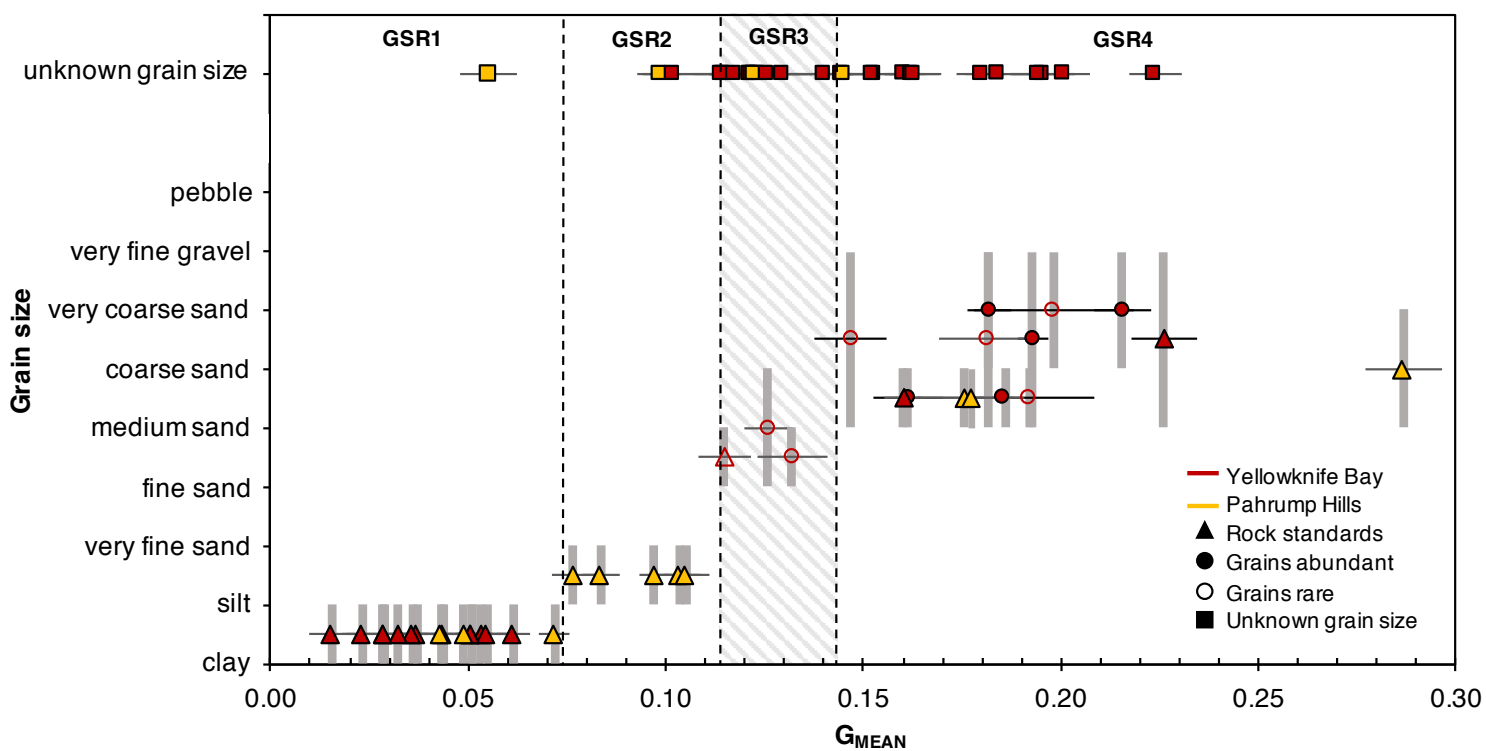


Figure 8.

## 11. Tables

**Table 1.** Rocks that were used as calibration standards for the GIMS reported in each grain size range considered. The GSRs were defined based on  $G_{MEAN}$  values. Grain size estimates for the rock standards at Yellowknife Bay were based on previous studies or made directly using RMI images, whereas for the rock standards at Pahrump Hills, grain size estimates were made by using MAHLI images of nearby regions or directly using RMI images. RMI image grain size estimates are for grains near the laser pits. Numerical values for grain size are reported for the bulk of the grains.

Target Names	Locality and Member/Outcrop	Image-based grain size estimate and instrument used for measurements	Sorting Estimate	Min and max $G_{MEAN}$	Grain Size Regime
Sheepbed Beachrock Belcher Flaherty Richardson Flaherty_2 Barn_2 Rackla Haig Hudson_Bay Nastapoka Wernecke_1 Rae2	Sheepbed, YKB	Mud, <62.5 $\mu\text{m}$ (Grotzinger et al., 2014; 2015)	Well sorted	0.01, 0.06	GSR1
The_Maze Crowley Hanaupah	Confidence Hills, PH	Mud to silty/sandy mud, <125 $\mu\text{m}$ (MAHLI)	Moderately well sorted	0.04, 0.07	GSR1
Goblin_Valley, Deadman_Pass, Sespe, Aguereberry_Point, Soledad_Pass	Book Cliffs, and Chinle, PH	Silt to very fine sand, 62.5-125.0 $\mu\text{m}$ (MAHLI)	Moderately well sorted	0.08, 0.11	GSR2
Nanok	Gillespie Lake Member, YKB	Fine to medium sand sized grains with sparse coarse sand, 0.125-0.500 mm (RMI)	Moderately well sorted	0.11	GSR3
Gillespie_Lake_1	Gillespie Lake Member, YKB	Medium sand to coarse sand with sparse very coarse sand, 0.5-1.0 mm (RMI)	Poorly sorted	0.16	GSR4
Wakham_Bay	Glenelg Member, Shaler, YKB	Medium to very coarse sand size sediment with sparse granules, 0.5-2.0 mm (RMI)	Moderately to poorly sorted	0.23	GRS4
Orocopia Vasquez, Wild Horse Mesa	Whale Rock, PH	Medium to coarse sand with sparse very coarse sand in a matrix of finer sediment, 0.5-1.0 mm (MAHLI & RMI)	Poorly to very poorly sorted	0.18, 0.29	GSR4

**Table 2.** All targets used in the GIMS analysis, with summary information, including independent grain size estimates if known and  $G_{MEAN}$ . Two standard deviation metrics are reported for each  $G_{MEAN}$ ,  $STDr$  and  $STDc$ , derived from standard deviations for each LIBS point and oxide on rock ( $STDr$ ) or the Shergottite calibration target ( $STDc$ ). Grain size estimates for rocks at Yellowknife Bay are for grains near the laser pits in RMI images. For rocks at Pahrump Hills, grain size estimates were indirectly made by using MAHLI images of regions near the ChemCam analyses. Rocks with  $G_{MEAN}=0.11$  are reported as GSR2/GSR3. Target names are merged in the same cell for those analyses that were taken on the same rock exposure and those with asterisks are calibration standards.

Formation /Outcrop	Member /Unit	Target Name	Sequence ID	Sol	Elevation (m)	# of points used	Raster type	$G_{MEAN}$	$STDr$ ( $1\sigma$ )	$STDc$ ( $1\sigma$ )	mean $G_{MEAN}$	Grain size estimate	GSR
Pahrump Hills	Confidence Hills	The_Maze*	ccam02767	766	-4460.28	8	3x3	0.05	0.01	0.00	0.05	Mud with silt to very fine sand	GSR1
		Crowley*	ccam04771	770	-4460.26	9	1x10	0.04	0.01	0.00		Mud with silt to very fine sand	GSR1
		Hanaupah*	ccam03779	778	-4460.21	9	3x3	0.07	0.00	0.00		Mud with silt to very fine sand	GSR1
	Book Cliffs	Goblin_Valley*	ccam01787	786	-4457.83	10	1x10	0.10	0.00	0.00	0.09	Silt to very fine sand	GSR2
		Deadman_Pass*	ccam02787	786	-4457.88	8	1x10	0.08	0.00	0.00		Silt to very fine sand	GSR2
	Alexander Hills	Cajon	ccam01792	791	-4456.01	10	1x10	0.10	0.00	0.00	0.12	Voids and dark features that are silt/very fine sand to very coarse sand in size	GSR2
		Agate_Hill	ccam02792	791	-4456.36	10	1x10	0.12	0.01	0.00		Voids and dark features that are silt/very fine sand to very coarse sand in size	GSR3

		Aztec_2	ccam03792	791	-4456.24	10	1x10	0.15	0.01	0.00		Voids and dark features that are silt/very fine sand to very coarse sand in size	GSR4
	Chinle	Cima	ccam01794	793	-4454.92	5	1x10	0.06	0.01	0.00	0.09	Uncertain from RMI mosaic and no MAHLI taken nearby	GSR1
		Sespe*	ccam02794	793	-4454.78	10	1x10	0.08	0.01	0.00		Silt to very fine sand	GSR2
		Aguereberry Point*	ccam03794	793	-4454.65	5	1x10	0.10	0.01	0.00		Silt to very fine sand	GSR2
		Soledad_Pass*	ccam04794	793	-4454.60	9	1x10	0.11	0.01	0.01		Silt to very fine sand	GSR2/ GSR3
	Whale Rock	Orocopia*	ccam01796	795	-4451.73	9	1x10	0.29	0.01	0.00	0.21	Medium to very coarse sand	GSR4
		Wild_Horse_Mesa*	ccam03796	795	-4451.73	8	1x10	0.18	0.01	0.00		Medium to coarse sand in a finer grained matrix	GSR4
		Vasquez*	ccam04796	795	-4451.73	8	1x10	0.18	0.01	0.01		Medium to coarse sand in a finer grained matrix	GSR4
		Sheepbed*	ccam01126	125	-4520.33	9	3x3	0.06	0.00	0.00	0.04	Mud (Grotzinger et al., 2014; 2015)	GSR1
		Beachrock*	ccam02126	125	-4520.33	9	3x3	0.04	0.01	0.00		Mud (Grotzinger et al., 2014; 2015)	GSR1

Yellowknife Bay	Sheepbed	Belcher*	ccam01127	126	-4521.07	9	3x3	0.02	0.00	0.00	Mud (Grotzinger et al., 2014; 2015)	GSR1
		Flaherty*	ccam01129	128	-4521.33	5	1x5	0.05	0.01	0.00	Mud (Grotzinger et al., 2014; 2015)	GSR1
		Richardson*	ccam02129	128	-4521.31	5	5x1	0.03	0.01	0.00	Mud (Grotzinger et al., 2014; 2015)	GSR1
		Flaherty_2*	ccam01130	129	-4521.32	5	1x5	0.04	0.01	0.00	Mud (Grotzinger et al., 2014; 2015)	GSR1
		Barn_2*	ccam03130	129	-4521.06	5	5x1	0.05	0.01	0.00	Mud (Grotzinger et al., 2014; 2015)	GSR1
		Rackla*	ccam05135	134	-4520.76	8	3x3	0.05	0.01	0.00	Mud (Grotzinger et al., 2014; 2015)	GSR1
		Haig*	ccam01150	150	-4520.30	5	5x1	0.02	0.01	0.00	Mud (Grotzinger et al., 2014; 2015)	GSR1
		Hudson_Bay*	ccam04150	150	-4520.50	8	3x3	0.03	0.00	0.00	Mud (Grotzinger et al., 2014; 2015)	GSR1
		Nastapoka*	ccam03160	159	-4520.25	9	3x3	0.04	0.01	0.00	Mud (Grotzinger et al., 2014; 2015)	GSR1
		Wernecke_1*	ccam01172	171	-4520.36	9	3x3	0.03	0.01	0.00	Mud (Grotzinger et al., 2014; 2015)	GSR1
		Rae2*	ccam02192	191	-4520.31	9	3x3	0.05	0.00	0.00	Mud (Grotzinger et al., 2014; 2015)	GSR1



	Gillespie Lake	Gillespie_Lake_1*	ccam01132	132	-4520.13	8	3x3	0.16	0.00	0.00	0.16	Medium to coarse sand size grains with sparse very coarse sand	GSR4
		Gillespie_Lake_2	ccam02132	132	-4520.09	8	3x3	0.16	0.01	0.00		Too dusty	GSR4
		Laddie	ccam01151	150	-4520.14	7	3x3	0.19	0.00	0.00		Medium to coarse sand sized grains	GSR4
		Kipalu	ccam03157	156	-4519.76	9	3x3	0.20	0.01	0.00		Too dusty	GSR4
		Nanok*	ccam02176	175	-4520.28	9	3x3	0.11	0.01	0.00		Fine to medium sand with sparse coarse sand. Fine sand is at resolution of RMI mosaic.	GSR2/ GSR3
		Jolliffe	ccam01181	180	-4520.01	10	10x1	0.12	0.00	0.00		RMI out of focus near raster	GSR3
		Doublet	ccam01186	185	-4520.00	10	1x10	0.13	0.01	0.00		Fine to coarse sand. Fine sand is at resolution of RMI mosaic.	GSR3
		Mugford	ccam02186	185	-4520.05	5	1x10	0.18	0.01	0.00		Uncertain	GSR4
	Glenelg, Point Lake	Acasta	ccam01104	103	-4518.38	7	3x3	0.12	0.01	0.00	0.14	Too dusty	GSR3
		Amagok	ccam01111	110	-4518.16	8	3x3	0.16	0.01	0.00		RMI mosaic out of focus	GSR4
		Ingraham	ccam01116	115	-4517.84	8	3x3	0.18	0.01	0.00		Too dusty	GSR4
		Kapvik	ccam02123	122	-4518.36	9	3x3	0.12	0.01	0.00		Too dusty	GSR3





		Rocknest6a	ccam04087	87	-4517.50	9	3x3	0.14	0.01	0.00		Uncertain	GSR3
		Rocknest6b	ccam05087	87	-4517.50	5	3x3	0.11	0.02	0.01		Uncertain	GSR2/ GSR3
		Peg	ccam03071	70	-4517.00	9	3x3	0.14	0.01	0.00		Uncertain	GSR3