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1 **Title:** An interval of high salinity in ancient Gale crater lake, Mars

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12

13 **Abstract/First Parag.**

14 Precipitated minerals, including salts, are primary tracers of atmospheric conditions and water chemistry  
15 in lake basins. Ongoing *in situ* exploration by the Curiosity rover of a thick section of Hesperian (~3.3-  
16 3.7 Ga) sedimentary rocks within Gale crater has revealed clay-bearing fluvio-lacustrine deposits with  
17 sporadic sulfate occurrences primarily as late-stage diagenetic veins and concretions. Here, we report the  
18 discovery of bulk enrichments, disseminated in the bedrock, of Ca-sulfate (30-50 wt%) that occur  
19 intermittently over ~150 m of stratigraphy and hydrated Mg-sulfate (26-36 wt%) within a thinner section  
20 of strata. We use geochemical analysis, primarily from the ChemCam laser-induced breakdown  
21 spectrometer (LIBS) and combined with results from other rover instruments, to characterize the  
22 enrichments and their lithology. The deposits are consistent with early diagenetic, pre-compaction, salt

23 precipitation from brines concentrated by evaporation, including Mg-sulfate rich brines indicating extreme  
24 evaporative concentration. This saline interval represents a significant hydrologic perturbation affecting  
25 the lake basin, and possibly reflecting variations in obliquity and orbital parameters of the planet. Our  
26 findings support a stepwise change in Mars' Hesperian climate towards more arid and sulfate-dominated  
27 environments inferred from orbital observations.

28

29 Carbonate, chloride, and sulfate salts provide geochemical fingerprints of the past chemistry of terrestrial  
30 lake basins. On Mars, such chemical sediments reflect systematic differences in dissolved ions that lead  
31 to distinctive chemical divides and mineral assemblages<sup>1</sup>. For example, *in situ*, widespread Mg/Ca/Fe-  
32 sulfate enrichments within aeolian and interdune playa sediments at Meridiani Planum have been  
33 interpreted to have formed from acidic and saline early diagenetic brines<sup>2</sup>. Low CO<sub>3</sub><sup>2-</sup>, high SO<sub>4</sub><sup>2-</sup>, and  
34 low pH waters enriched in Fe(II) formed a unique assemblage of hydrated sulfate minerals, jarosite, silica,  
35 and chloride minerals<sup>3</sup>. Under less acidic conditions, iron is relatively immobile, Ca/Mg-sulfate and  
36 chloride assemblages are favored, and carbonate minerals can form under sufficiently elevated CO<sub>3</sub><sup>2-</sup>  
37 concentrations<sup>1</sup>.

38 From orbit, a diversity of sulfate, carbonate, and chloride salts is observed on Mars, reflecting fluid  
39 chemistries that varied both spatially and temporally<sup>4</sup>. Thick layered sulfate deposits are observed at a  
40 number of late Noachian to late Hesperian locations (~3.5 Ga). Their apparent absence in the older  
41 stratigraphic record is hypothesized to reflect the diminishing availability of liquid water on Mars<sup>5</sup>. Gale  
42 crater provides an exemplary sedimentary succession dated to the Hesperian<sup>6</sup> that contains clay minerals  
43 transitioning to sulfate minerals over ~300 m of stratigraphy<sup>7,8</sup>. Understanding the reason for this transition  
44 is one of the primary objectives of the MSL/Curiosity rover investigation. So far, the rover has explored  
45 clay-bearing fluvio-lacustrine sedimentary record in the lowermost stratigraphy which is dominated by

46 the formation of mineral assemblages that include authigenic or detrital clay minerals and iron oxides<sup>9-12</sup>,  
47 with a potentially punctuated transition in redox state<sup>10</sup>. Sulfates have been observed mostly as abundant  
48 late diagenetic Ca-sulfate fracture-fills<sup>13,14</sup>, and inferred from crystal pseudomorphs after gypsum that  
49 occur locally in the lowermost Murray formation<sup>15</sup>. Mg-sulfate were also observed, but primarily  
50 associated with sparse diagenetic features, such as concretions and dendrites<sup>10,16,17</sup>.

51 Here we detail the chemistry of lacustrine deposits from the upper Murray formation heterolithic facies  
52 which consists of interbedded sandstone and mudstone with exposures dominated by broken, tilted blocks  
53 with diverse resistance to erosion. This change in sedimentary facies in the Murray formation correlates  
54 with a gradual transition in the bedrock alteration index defined by major element compositions<sup>18</sup>. It also  
55 coincides with an increase in the prevalence of S, Cl and associated salts which suggest the presence of  
56 distinctively saline waters.

### 57 **Ca-sulfate bedrock enrichment**

58 Starting at the base of the Sutton Island member (Figure 1-a), enrichment of Ca-sulfate is observed within  
59 the bulk sedimentary bedrock, distinct from prior occurrences in fracture fills and merely sporadic  
60 detections (targets with <50% of points enriched). Bulk enrichments are characterized by ChemCam  
61 analyses of bedrock, where the majority of sampled points exhibit compositions that correspond to binary  
62 mixtures of typical Murray bedrock (i.e. average sulfate-free bedrock dominated by silicate minerals, with  
63 an average CaO content ~1.6 wt%) with 30 to 50 wt% of a Ca-sulfate end-member (Figure 2-a; see also  
64 Figure S1 and Table S1). For such concentrations, the S signal is clearly expressed in emission spectra by  
65 3 peaks at 543 nm, 545 nm, and 564 nm, leading to a ~0.2 nm shift in the location of peak centroids at  
66 543 nm and 545 nm, combined with an increase in the magnitude of the 564-nm peak emission (Figure  
67 S2), as is observed in laboratory experiments<sup>19</sup>. Independent measurement by the Alpha Particle X-ray

68 Spectrometer (APXS) at the centimeter-scale also record bulk CaO enrichments of the Murray bedrock  
69 that are correlated with SO<sub>3</sub> content, toward the CaSO<sub>4</sub> end-member (Figure S3).

70 Ca-sulfate corresponding to bedrock enrichment excludes contribution from mineralized veins identified  
71 from images taken at <0.5mm/pixel using the ChemCam Remote Micro Imager (RMI). In the absence of  
72 vein contribution, the repeated enrichment in the majority of observation points within both sandstone and  
73 mudstone targets indicates that the enrichment is disseminated within the bulk rock (Table S1; Figure 3).  
74 In general, the mudstone is laminated<sup>20</sup>; however, intervals with Ca-sulfate enrichment are commonly  
75 more resistant to erosion and display nodular or massive textures (Figure 3, S4 and S5). Where close-up  
76 images (< 100 μm/pixel) are available, these intervals do not show veins (Figure 3-e), although in some  
77 cases, submillimeter white grains (Figure 3-f) or elongate euhedral crystals associated with crystal molds  
78 (Figure 3-g) were observed. Bulk enrichment occurs at several discrete elevations between -4370 m and -  
79 4200 m in the stratigraphy of the upper Murray formation, contrasting with only sporadic enrichment  
80 detected elsewhere in the formation (Figure 1-b).

81 The association of calcium with carbonate, which neither ChemCam or APXS can directly detect, cannot  
82 be entirely ruled out (Figure 2-a shows that bedrock CaO content increases up to ~8 wt% while sulfur  
83 remains below detection limit which could allow for up to ~12 wt% calcite). Yet X-ray diffraction analyses  
84 on Murray bedrock drill samples confirmed the absence of carbonates (<1 wt%) and the presence of clay  
85 and Ca-sulfate<sup>12</sup>. An association between CaO and sulfate is also clearly observed (Figure S3), suggesting  
86 no significant calcite is present in the Murray formation.

### 87 **An interval with Mg-sulfates**

88 In addition to Ca-sulfate, Mg-sulfate bedrock enrichments occur in a relatively thin (<10 m) stratigraphic  
89 interval within the heterolithic Sutton Island member (Figure 1-d; purple bar). On sol 1690 (elevation, -

90 4287 m), ChemCam analysis of unusual, brown-colored bedrock fragments within the Murray mudstone  
91 showed a distinct enhancement of Mg, S, and H. The estimated composition is interpreted as a binary  
92 mixture between a hydrated Mg-sulfate end-member and typical bedrock of the Murray formation (Figure  
93 2-b,c and S1). The analyzed fragments have a bulk water content of 8.6 +/- 1.9 wt%, estimated from the  
94 hydrogen signal<sup>21</sup>, which is significantly higher than typical Murray bedrock (1.6 wt%). The lack of Ca-  
95 sulfate enrichment in these rocks is reflected in a very low CaO content (Figure S1). The magnitude of  
96 the sulfur signal is consistent with  $18.4_{-4.8}^{+4.0}$  wt% average sulfate content based on measurements of the  
97 Norwood Cove fragment (Figure S6) with 1-sigma uncertainty, corresponding to a bulk content of 33 wt%  
98 hydrated Mg-sulfate (1-sigma lower bound is 26 wt% ; Table S2).

99 Mg-sulfate enriched lithologies occur in planar exposures of relatively erosion-resistant rocks. The  
100 outcrop is poorly preserved, and consists of broken, meter-scale blocks separated by unconsolidated fines.  
101 Relative to surrounding lithologies, strata with Mg-sulfate enrichment have a reddish-brown color, are  
102 variably fractured, show a typical nodular to massive texture (Figure S7), and weather into centimeter-  
103 sized angular fragments (such as the Norwood Cove and Beach Cliff targets, see Figure S6). The grain  
104 size of these Mg-sulfate enriched rocks is silt-sized or smaller, on the basis of compositional homogeneity  
105 of the ChemCam data (Figure S8), and is similar to grain sizes estimated for surrounding Murray  
106 bedrock<sup>22</sup>. At several locations, this lithology is exposed in large contiguous blocks and confined to cm-  
107 thick planar exposures that are evenly interbedded with finely-laminated Murray mudstone (Figure 3-a).  
108 The lateral extent of Mg-sulfate-enriched exposures is uncertain because of the lack of high-resolution  
109 image coverage and the scattered nature of bedrock exposures (Figure S7). The maximum lateral distance  
110 observed in images is 4 m. Sporadic Mg-sulfate detections were also observed over 30 meters of thickness  
111 of heterolithic facies above (Figure 1-d).

112 The Mg-sulfate enriched rocks were not examined by the Alpha Particle X-ray Spectrometer (APXS), but  
113 high epithermal neutron count rates, recorded by the Dynamic Albedo of Neutrons (DAN) instrument in  
114 the same area (Figure S9), support the presence of elevated bedrock hydration within this stratigraphic  
115 interval. A signature consistent with a hydrated sulfate phase is also recorded on the site from orbit:  
116 hydration and hydrated sulfate spectral indices are enhanced in the portion of the Murray formation where  
117 Mg-sulfate enriched lithologies were observed *in situ*, and the signature was less clear or absent in bedrock  
118 exposures below that interval (Figure S11). The hydrated sulfate signature also extends laterally at similar  
119 elevation to either side of the rover traverse.

#### 120 **Timing of enrichment and formation scenario**

121 Understanding the timing of bedrock sulfate enrichment is crucial to deciphering its environmental  
122 implication. Enrichments have not been observed in the form of large continuous beds of nearly pure salt,  
123 as could be expected for evaporite minerals formed as cumulates or as lake bottom-growth deposits. In  
124 available images of bedrock, no features were observed that clearly indicate primary salts, their  
125 dissolution, or the presence of disruptive fabrics common to terrestrial evaporitic settings<sup>23</sup>.

126 Although a late diagenetic origin for Ca and Mg-sulfate enrichments cannot be uniquely discounted, we  
127 believe that such an origin is unlikely. Local bedrock does not appear to show a greater abundance of  
128 mineralized fractures (Figure 3, S4 and S5), and sulfate enrichments are disseminated in the host rock and  
129 do not correlate with the late-diagenetic fractures mineralized by Ca-sulfate<sup>13</sup> (Figure S5). Additionally,  
130 the presence of mineralized veins within typically sulfate-poor Murray bedrock suggests that bedrock  
131 porosity was sufficiently reduced that later diagenetic fluids were confined to fracture systems.

132 Early diagenetic precipitation from saline brines, followed by recrystallization during burial and  
133 exhumation, is in line with the observed geochemistry and textures. In particular, the disseminated sulfate

134 enrichments are restricted to a specific stratigraphic interval, where they occur as repetitive discrete  
135 horizons (Figure 3). In this scenario, episodic saturation of sulfate-rich brines resulted in the early  
136 diagenetic precipitation of sulfate salts within shallow sediments (Figure 4). Such subsurface  
137 crystallization can occur either during subaerial exposure resulting in evaporative enrichment in the  
138 capillary fringe, or subaqueously as dense supersaturated brines diffuse downward into the sediment pore  
139 space<sup>24</sup>. The estimated 30-50 wt% of Ca-sulfate enrichment is consistent with intrasediment crystallization  
140 in poorly compacted sediment<sup>25</sup>. Similar deposition could have occurred with Mg-sulfates where brines  
141 became much more concentrated by evaporation (Figure 4). The estimated proportion of 26-36 wt% Mg-  
142 sulfate initially represents 38-50% by volume, assuming precipitation of epsomite ( $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ ) in pores  
143 of siliciclastic sediment with grains of approximately basalt density ( $\sim 3.0 \text{ g}\cdot\text{cm}^{-3}$ ), again implying growth  
144 in poorly compacted sediments.

145 Sulfate enrichment within discrete beds of limited lateral extent in the heterolithic mudstone-sandstone  
146 facies indicate that salts were likely deposited along shallow lake margins, possibly within multiple ponds  
147 fed by distributive channel systems and groundwaters (Figure 4). Mg-sulfate enrichment, which is  
148 confined to relatively small stratigraphic intervals, occurs in horizons discretely separate from Ca-sulfate  
149 suggesting that Mg-sulfate rich brines were physically fractionated from the fluid that formed Ca-sulfates  
150 as the brine concentrated downstream. Changes in seasonal temperature and humidity cycling during post-  
151 depositional exposure of such hydrated sulfate salts would have exposed these materials to repeated  
152 changes in hydration state and dissolution. In the absence of complete dissolution, volumetric changes  
153 associated with changes in hydration state are expected to disrupt initial depositional textures<sup>26,27</sup>.

154 After deposition, the Murray formation was likely buried to  $\sim 1 \text{ km}^9$ , which itself may have triggered a  
155 change in Ca-sulfate hydration state<sup>28,29</sup>. Following compaction and lithification, late-stage fluids  
156 deposited Ca-sulfate within fractures. Horizontal and subhorizontal veins occur specifically in the



157 stratigraphic interval with the enrichments (Figure 1-c), a setting similar to terrestrial satin spar veins  
158 associated to primary Ca-sulfate deposits<sup>30</sup>. These late Ca-sulfate fluids were likely to have partially  
159 mobilized highly soluble salts within the bedrock such as halite. Indeed enhanced Na-chloride detections  
160 occur at specific stratigraphic intervals (Figure 1-e, Figure S10), and their sporadic distribution in bedrock  
161 and occasionally in association with fracture-filling Ca-sulfates indicate remobilization<sup>31</sup>. Locally, lateral  
162 variation and possible cross-cutting of Mg-sulfate enrichment occurs at a cm-dm scale (Figure S7), as well  
163 as sporadic detections of Mg-sulfate nearby the stratigraphic succession (Figure 1-d), may also reflect  
164 localized dissolution and reprecipitation of initial deposits. (Figure 1-d). Yet, while Ca-sulfate-enriched  
165 fluids would easily mobilize halite, Mg-sulfate-enriched bedrock could persist even though Mg-sulfate is  
166 soluble, because  $\text{SO}_4^{2-}$  ions released during dissolution would immediately trigger the formation of less-  
167 soluble Ca-sulfate, which could shield Mg-sulfates from further dissolution.

168 Upon exhumation and exposure of the Murray formation to temperatures and relative humidity of modern  
169 Mars, hydrated Mg-sulfate minerals turn to amorphous phases of various hydration states. The calculated  
170 21 wt% water associated with the Mg-sulfate phase (Figure 2-c; Table S2) corresponds to a hydration state  
171 of 1.8  $\text{H}_2\text{O}$  per  $\text{MgSO}_4$ , and is consistent with the predicted amorphous desiccation products of epsomite  
172 or hexahydrate<sup>26,32</sup>. The intensely fractured and broken nature of Mg-sulfate enriched rocks may reflect  
173 diurnal and seasonal cycles of dehydration-rehydration<sup>33</sup>.

174 Overall, geochemical and textural data are consistent with sulfate enrichment in this interval by early  
175 diagenetic precipitation from saline waters although unambiguous sedimentary structures (e.g., nodular  
176 displacive fabrics) are not observed. This may result from the scarcity of high-resolution images acquired  
177 in this interval or to the post-depositional alteration of primary depositional textures. Importantly,  
178 however, features other than sulfate enrichments indicate lowering of lake levels and intermittent  
179 evaporative conditions in this part of the Murray formation. First, desiccation cracks are identified in the

180 stratigraphy at Old Soaker<sup>34</sup> and possibly at several other locations (Figure 11) in close proximity to the  
181 strata containing sulfate enrichment (Figure 1-d). At Old Soaker, Ca-sulfate enrichment occurs in  
182 sandstone adjacent to the mudcracks (Figure 3-d). Second, elevated concentration of Na-chloride in the  
183 bedrock is also observed, relative to stratigraphically lower strata<sup>31</sup>. Na-chloride occurrences in specific  
184 intervals, close to sulfate enrichments suggests remobilization from layers potentially enriched in halite  
185 (Figure 1-e). Third, the first occurrence of Ca-sulfate enrichment coincides with a previously reported  
186 change in clay mineralogy, consistent with episodic wetting and drying of lake environments<sup>12</sup> and  
187 increased chemical weathering of the bedrock detrital component<sup>18</sup> (Figure 1). In Gale's closed-basin  
188 hydrology, leached ions would accumulate in waters and upon evaporation create brines that precipitate  
189 salts, such as Ca and Mg-sulfates.

#### 190 **Climatic variability**

191 The observed Ca-/Mg-sulfate and chloride assemblages are predicted to result from alteration of Martian  
192 basalts to produce sulfate- and chloride-rich but iron-poor brines<sup>1</sup>. The relatively low solubility of Ca-  
193 sulfate minerals results in their widespread production during evaporation, while less common Mg-sulfate  
194 and chloride minerals represent terminal evaporation. Iron oxides are observed in visible/near-infrared  
195 spectral data and x-ray diffraction of Murray bedrock samples<sup>28,35</sup>, yet there is no evidence for Fe-sulfate  
196 enrichments in this interval, likely because Fe was not mobile in solution, indicating that saline waters  
197 were either oxidizing or not acidic. While Ca-sulfate enrichment is observed, calcite is absent from the  
198 stratigraphy, i.e. not detected by CheMin at its threshold of 1 wt.%. This could result from either low  
199 alkalinity or low atmospheric CO<sub>2</sub>, or an environment where precipitation of calcite was otherwise  
200 inhibited<sup>36</sup>. Besides, the presence of Ca-sulfate means prior CaCO<sub>3</sub> formation (if any) was insufficient to  
201 deplete Ca from solution. Our findings do not compromise the search for life in Gale crater; terrestrial Mg

202 sulfate-rich hypersaline lakes are known to accommodate halotolerant biota<sup>37-39</sup> and crystallization of  
203 sulfate salts may also aid preservation of biosignatures<sup>40</sup>.

204 Previous observations suggest different lake depths were recorded in the lower Murray formation<sup>10</sup>. In the  
205 upper part of the formation, where clear rock exposures are more scarce, sedimentary structures suggest  
206 deposition in lake-margin environment with potential episodes of desiccation<sup>34,41,42</sup>. Our discovery of  
207 sulfate deposits is consistent with episodic dessication. The limited lateral extent of sulfate-enriched beds  
208 also suggests the potential for segmentation of the Gale lake into discrete ponds, including those where  
209 extremely evapo-concentrated brines might form (Figure 4). Changes in lake level are an anticipated  
210 response within closed basin lakes to variation in the regional hydrological budget. In addition, over time  
211 intervals for lacustrine deposition of ~1 Ma or less, based on plausible sedimentary deposition rates<sup>9</sup>, Mars  
212 experiences significant changes in obliquity and orbital eccentricity driving cyclical climate fluctuations<sup>43</sup>.  
213 Alternatively, a drier lake Gale might be a sign of long-term, secular global drying of Mars, posited based  
214 on orbital observations<sup>5</sup>.

215 The interval discussed here preserves the first occurrence of significant sulfate-enriched bedrock (>30  
216 wt% sulfate with siliciclastics) in Gale crater's sedimentary record. As the rover continues to traverse  
217 upsection toward successively younger rocks, key questions include the prevalence of similar bedrock  
218 sulfate enrichments. If thin stratigraphic intervals with salt enrichments or sedimentological evidence for  
219 aridity continue to occur, cyclical or episodic processes are a plausible underlying cause. If, on the other  
220 hand, sulfate enriched intervals increase in frequency and thickness, perhaps incorporating observable  
221 displacive fabrics and pure sulfate beds, the enrichments investigated here may be a harbinger of the long  
222 term, secular drying of Mars. This has been hypothesized for the Mt. Sharp sulfate unit observed from  
223 orbit further upsection<sup>8</sup> and deposits elsewhere on Mars<sup>5</sup>. As the rover continues its traverse towards this  
224 sulfate unit, it will be therefore crucial to recognize sulfate-enriched bedrock in reconnaissance

225 observations, so as to obtain accompanying high resolution stereo images of textures and drill samples for  
226 x-ray diffraction, evolved gas, and isotopic analyses.

227

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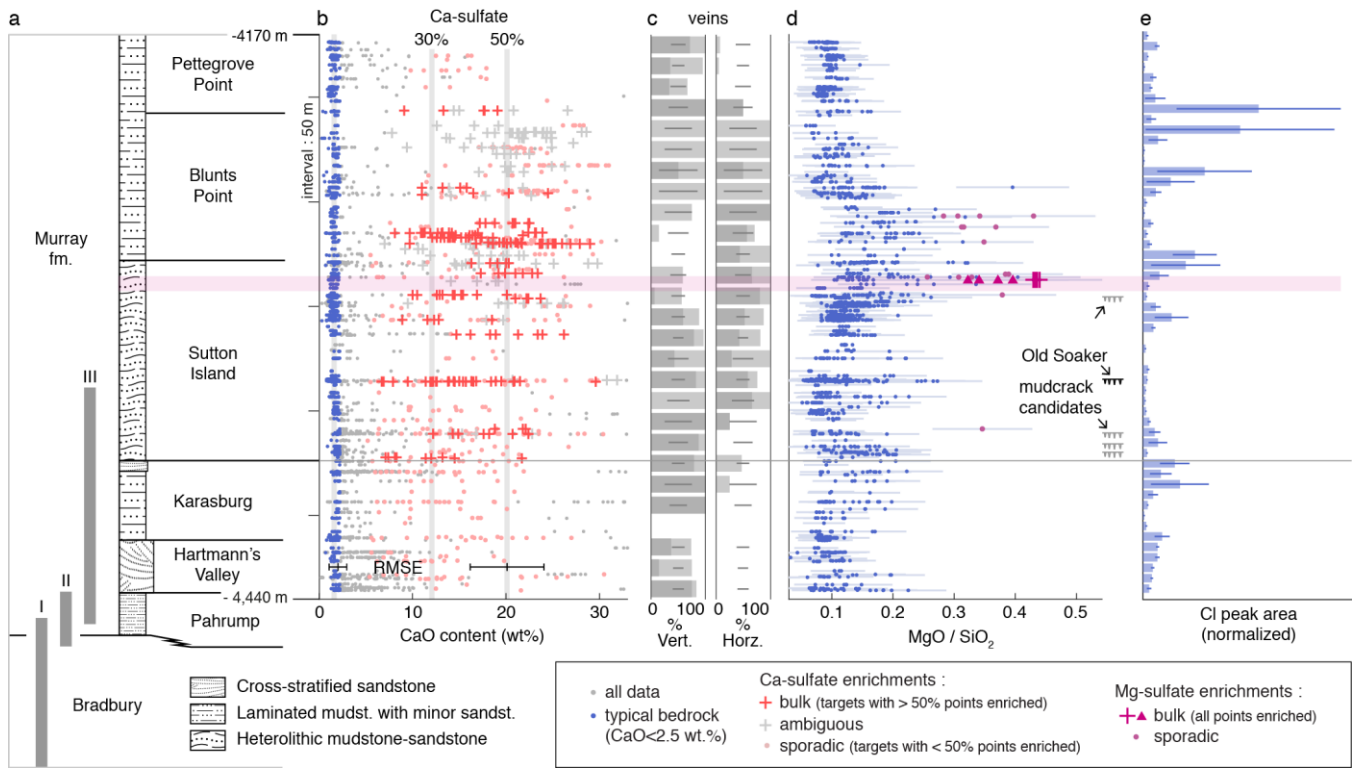
321

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330

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332 conceived and revised the manuscript; J.S., W.W.F., B.C.C., L.C.K., N.M., R.C.W. and A.R.V.,  
333 contributed to the interpretation of the data and revisions of the manuscript; N.H.T. provided Cl peak  
334 analysis; V.K.F. analyzed CRISM signatures of hydrated sulfates; N.T.S. and F.R.H. provided grain size  
335 estimates from MAHLI images of Murray bedrock; M.N. and H.A.M. identified and mapped sulfate veins  
336 occurrences; L.T. analyzed APXS data of Murray bedrock; T.S.J.G and C.H. provided DAN data analysis.

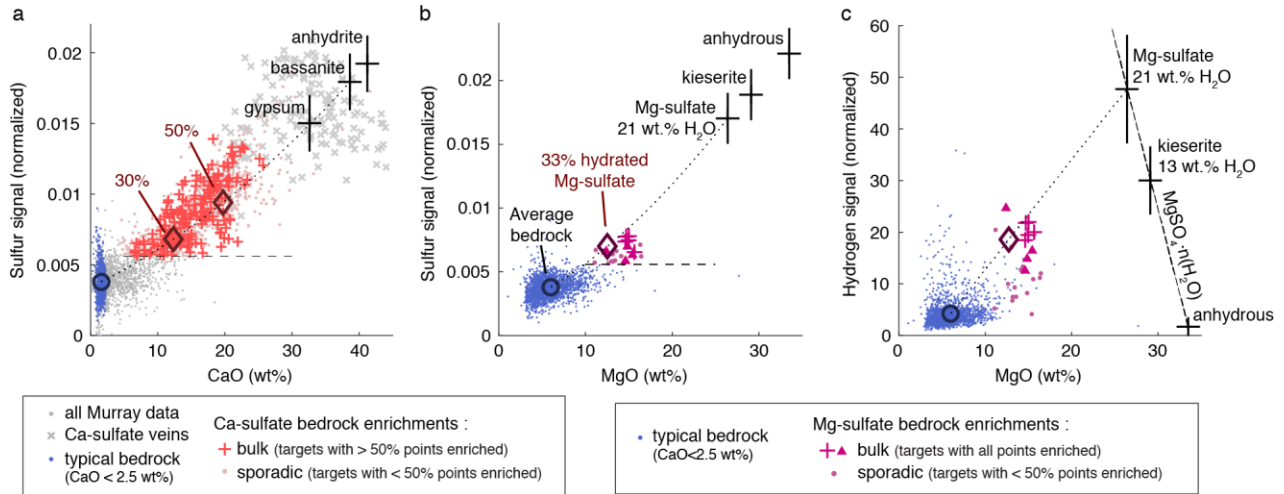




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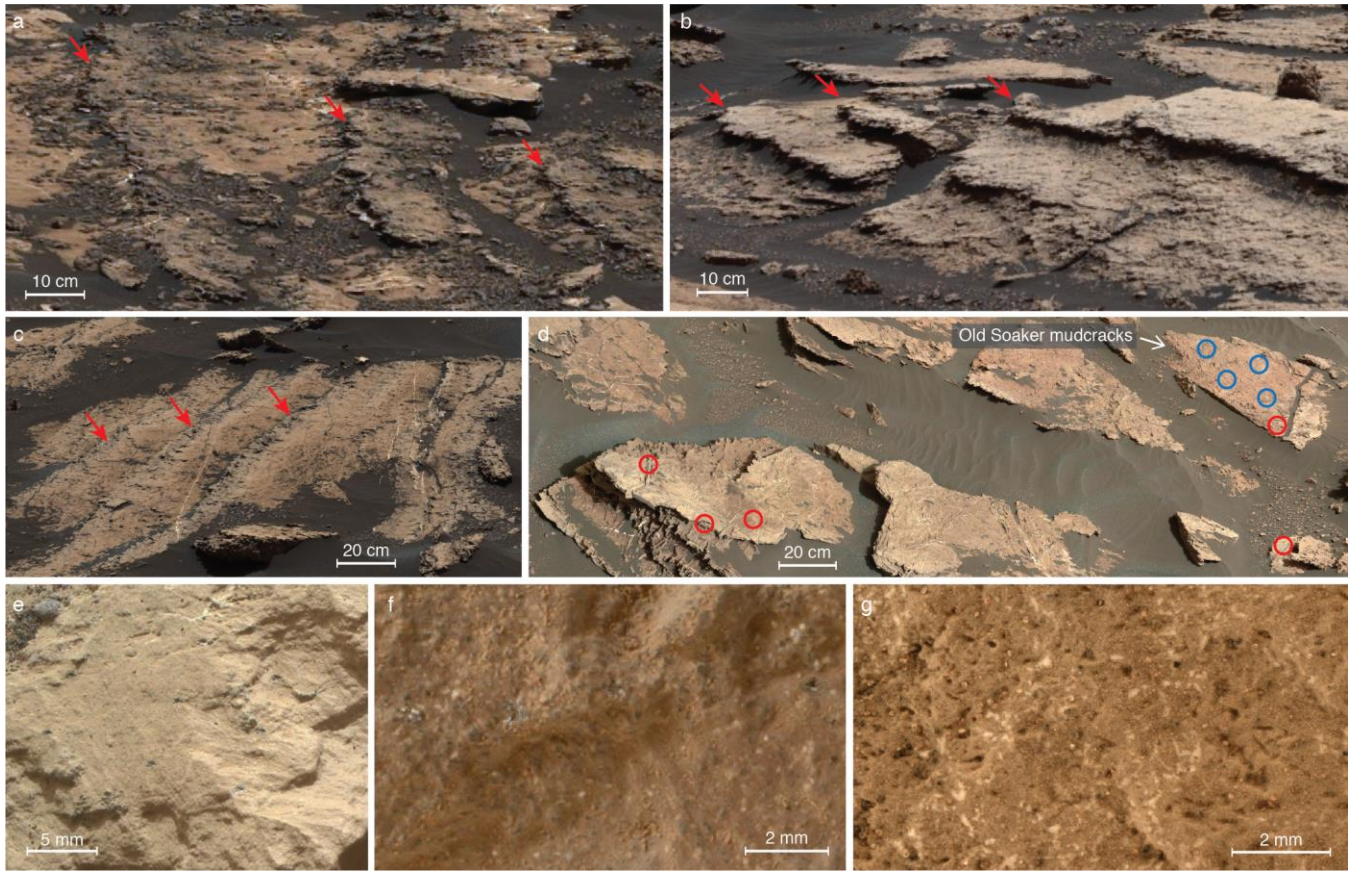
338 **Figure 1: Stratigraphic context on bedrock composition.** **a**, Overview of the ~350 m of stratigraphy  
 339 explored *in situ* at Gale crater including dominant facies observed<sup>41</sup> and sections highlighting evidence  
 340 for: I, fluvio-lacustrine environment<sup>9</sup>; II, lake redox stratification<sup>10</sup>; III, increase of bedrock chemical  
 341 weathering upsection due to leaching of Ca-bearing mafic minerals<sup>18</sup>. **b**, Bedrock CaO content with bulk  
 342 (crosses), or sporadic (dots) enrichments. Grey crosses represent bulk enriched targets but which  
 343 distinction to fracture-fill veins remains ambiguous from available images. Vertical grey lines highlight  
 344 average bedrock composition (1.6 wt% CaO), with 30% and 50% Ca-sulfate (bassanite) enrichments. **c**,  
 345 Occurrence of fracture-fill veins for vertical and horizontal orientations as percent of the number of  
 346 workspace images with bedrock (Figure S12). Light-grey indicates at least a vein is visible and dark grey  
 347 correspond to higher density of veins; horizontal bars represent the uncertainty of the average. **d**, Bedrock  
 348 MgO/SiO<sub>2</sub> content ratio (same data as Figure 2-b,c). Error bars represent RMSE for the ratio of MgO and  
 349 SiO<sub>2</sub> content. The purple horizontal bar represents the elevation range where Mg-sulfate enriched

350 lithology is observed (Figure S7). Location of Old Soaker<sup>34</sup> and other candidate desiccation cracks (Figure  
351 S13). **e**, Average normalized Cl peak area associated with halite detections (Figure S10)<sup>31</sup>, error bar  
352 represents the uncertainty of the average.



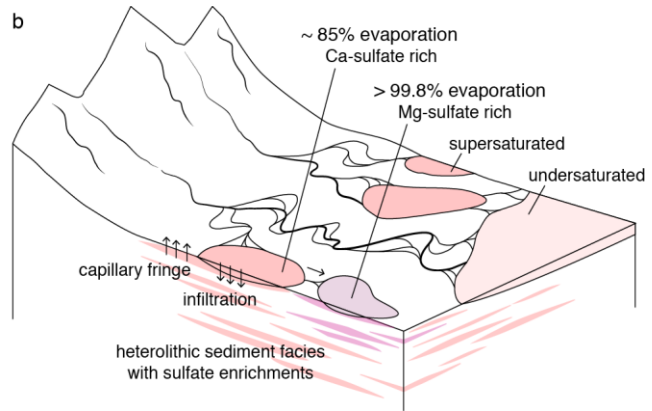
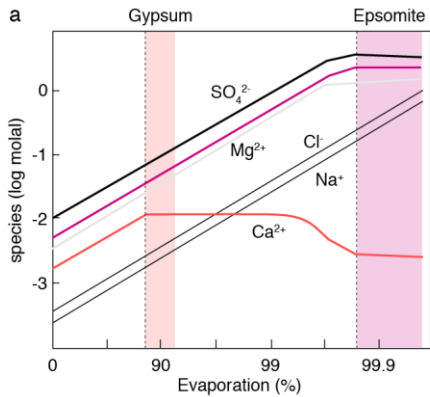
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354 **Figure 2: ChemCam oxide and elemental data of Ca- and Mg-sulfate enrichments.** Sulfur signal as a  
 355 function of CaO **(a)** and MgO content **(b)**, and hydrogen signal as a function of MgO content, for typical  
 356 Murray bedrock with CaO < 2.5 wt% (blue) and for sulfate enrichments. **a**, For Ca-sulfates (red), sulfur  
 357 signal above the limit of detection (dashed line, SO<sub>3</sub> > 10.6 wt%) and CaO > 5 wt%. Mixtures at 30% and  
 358 50% (diamonds) of Ca-sulfate with average typical bedrock (circle) are shown with the binary mixing  
 359 trend (dotted line) and end-members (black crosses). **b-c**, Mg-sulfate enrichments (purple), defined by  
 360 sulfur signal above the limit of detection and MgO > 10 wt%, found disseminated in the Norwood Cove  
 361 (crosses) and Beach Cliff (triangle) bedrock fragments (Figure S6), and compared to 33% hydrated Mg-  
 362 sulfate mixture (diamond) with average bedrock (Table S2). **c**, Dashed line represents Mg-sulfate with  
 363 varying water content. Vertical error bars on end-members represent sulfate **(a,b)** and water content **(c)**  
 364 calibration uncertainties.



365

366 **Figure 3: Lithology of the sulfate enrichments. a,** Mg-sulfate enriched lithology within multiple parallel  
 367 horizons (red arrows) on a large block of bedrock (mcam08715). **b,** Ca-sulfate enrichments as erosion-  
 368 resistant horizons (red arrows) in mudstone (mcam08669; Figure S4-a). **c,** Multiple horizons (red arrows)  
 369 where euhedral white crystals (Figure S4-g) and heterogeneous Ca-sulfate and Mg-sulfate enrichments  
 370 were observed (mcam08733). **d,** Exposed blocks at Old Soaker with desiccation cracks<sup>34</sup> (Figure S13-a)  
 371 (mcam07752); circles represent the locations of ChemCam targets with Ca-sulfate enrichments (red) and  
 372 without (blue). Close-up MAHLI images of a mudstone (**e**, Lookout Point, Figure S5-a) and a sandstone  
 373 enriched in Ca-sulfate (**f**, 1710MH0001220010604335C00; White Ledge, Figure S5-c). **g,** Close-up  
 374 MAHLI image of bedrock with white euhedral crystals along with empty casts/molds  
 375 (1679MH0001930000603555R00; Figure S4-d).



376

377 **Figure 4: Evaporation of surface brines and early diagenetic deposition. Left,** Evaporation of basaltic  
 378 weathering-derived fluids leads to major precipitation of Ca-sulfates first like gypsum (red) and then Mg-  
 379 sulfates like epsomite (purple) within the shaded ranges of evaporation, after Tosca et al. (2005)<sup>3</sup>. The  
 380 evolution of Fe species, which are dependent on pH, does not significantly change the result on Ca- and  
 381 Mg-sulfate precipitation<sup>3</sup> and are not shown here because Fe is relatively immobile (found in iron oxides,  
 382 clays, and amorphous phases). **Right,** Deposition scenario in the margin of the Gale lake basin, possibly  
 383 forming several over-filled or evaporative ponds fed by distributive fluvial systems consistent with the  
 384 heterolithic mudstone-sandstone facies observed in the upper Murray formation<sup>41</sup>. Brine crystallizes in the  
 385 shallow sediment pore space either by subaqueous infiltration, or within capillary fringes where occasional  
 386 subaerial exposure occurs.

387

388 **Methods**

389 **Rover-based geochemical datasets and data processing.** MSL has 5 instruments that can measure  
390 geochemistry and mineralogy<sup>44</sup>. Because of the rover traverse path and rate at the time, APXS, CheMin,  
391 and SAM data were not acquired of the Mg-S-H enriched strata so ChemCam is the primary source of  
392 chemical data used along with hydration of the upper ~0.5 m of the surface probed by the DAN instrument.  
393 The latter is a neutron spectrometer, sensitive to thermal and epithermal neutrons, which is regularly  
394 operated in active and passive modes throughout the traverse. The count rate of thermal neutrons in passive  
395 mode (data shown in Figure S9) can be elevated either due to the presence of neutron scatterers  
396 (predominantly H) or the paucity of neutron absorbers (predominantly Cl and Fe)<sup>45</sup>. On the other end,  
397 ChemCam is a laser-induced breakdown spectrometer (LIBS) that provides chemical analyses at a  
398 submillimeter scale and detailed images with the Remote Micro Imager (RMI)<sup>46,47</sup>. Major-element  
399 contents were obtained using the current calibration model<sup>48</sup>. Water and sulfate contents were estimated  
400 using dedicated calibration models, see Rapin et al. (2017)<sup>21</sup> for water content quantification using the  
401 hydrogen signal, and see below a description of the sulfur signal calibration used in this study.

402 The ChemCam dataset used here includes bedrock targets of the Murray formation. A subset of bedrock  
403 observations was selected from all ChemCam observations based on: 1) image classification to select only  
404 bedrock analyses and remove potential contribution from soils or diagenetic features such as light-toned  
405 fracture-fill veins, 2) signal strength to keep points above a minimum total spectral intensity (more than  
406  $1.6 \cdot 10^{14}$  photons) for which noise level is acceptable for sulfur peak analyses. An additional selection is  
407 made based on major-element oxide contents to separate points that indicate contributions from Ca-  
408 sulfate-, silica- or iron-rich features in order to represent typical bedrock composition (Figure 2, Figure  
409 S1). The threshold for rejection due to Ca-sulfate enrichment was defined at 2.5 wt% CaO from the  
410 distribution of bedrock observations which shows that measurements group tightly around 1.6 wt% CaO

411 with sporadic enrichments above 2.5 wt% (Figure S3) related to increased S signal (Figure S2). The APXS  
412 instrument independently measures centimeter-scale bulk compositions and demonstrates the association  
413 of calcium and sulfur in Murray bedrock observations. The CaO content measured by APXS ranges from  
414 2 wt% to 25 wt% and is correlated with SO<sub>3</sub> content along the 1:1 stoichiometric ratio of Ca-sulfate (Figure  
415 S3). Given the correlation observed on APXS data down to 2 wt% CaO, the slightly lower CaO abundance  
416 of the cluster defined by ChemCam data is most likely related to its ability to examine bedrock free of  
417 dust and without Ca-sulfate enrichments with its 0.3-0.5 mm analytical footprint compared to the 17 mm  
418 footprint of APXS.

419 **Estimation of sulfate content from ChemCam data.** SO<sub>3</sub> content is estimated from the sulfur signal  
420 observed in ChemCam spectra. The challenge with the measurement of the signal is that the sulfur  
421 emission peaks present in ChemCam spectra are relatively weak in comparison to nearby interfering iron  
422 emission peaks<sup>13</sup>, necessitating a distinct method for their quantification compared to the major oxides.  
423 Others have investigated the calibration of S with LIBS<sup>49-51</sup>, here we used a distinct approach dedicated  
424 to data collected *in situ* on sulfates within the Murray bedrock. The approach is first to show from  
425 laboratory data that spectral parameters can be defined to identify and differentiate the sulfur signal from  
426 other interfering spectral features, including identification of the threshold of S wt. % at which it becomes  
427 apparent. Second, the sulfur signal identified on Mars is calibrated *in situ* using a large set of Murray  
428 bedrock measurements containing Ca-sulfate enrichments.

429 *Collection of laboratory reference spectra.* The ChemCam testbed at Los Alamos National Laboratory  
430 was used to collect data from pelletized mixtures of Ca-sulfates and Mg-sulfate with basalt as described  
431 in Anderson et al. (2017)<sup>19</sup>. Additional Mg-sulfate pressed pellets were also prepared to resolve sulfur  
432 signal variation closer to the limit of detection and compositional range observed in the enrichments at  
433 Gale crater. The same basalt as Anderson et al. (2017)<sup>19</sup> was used –K1919 basalt sieved powder (< 45



434  $\mu\text{m}$ )– but it was doped with 7 wt% sieved hematite powder ( $< 45 \mu\text{m}$ ) so that the mixture had a total iron  
435 content of 19 wt%, closer to the bedrock composition of the Murray formation at Gale crater. The Mg-  
436 sulfate powder was synthetic reagent, the same as used in <sup>19</sup> from Macron Chemicals (CAS 7487-88-9),  
437 and was mixed in various proportions with the basalt-hematite powder (Table S3). The powder mixture  
438 pellets were each analyzed with 8 observation points of 30 laser shots each.

439 *Processing and measurement of S emission peaks.* Iron peaks interfere significantly with the sulfur  
440 emission peaks at  $\sim 543 \text{ nm}$  (S II at 543.02 nm and S II at 543.43 nm forming a single peak in ChemCam  
441 spectra) and 545.53 nm, but the sulfur still produces specific spectral peak features when present. The  
442 sulfur signal can be distinguished using the position of the spectral peak centroid computed from,  
443 respectively, 10 and 11 spectral bins on the two specific locations after baseline correction (Figure S14).  
444 The sulfur emission peak area at  $\sim 564 \text{ nm}$  (multiple S II peaks forming a single peak in ChemCam spectra),  
445 with minor Si and Fe emission peaks interferences, is then used to evaluate sulfur signal intensity. It is  
446 normalized to the oxygen emission peak area at  $\sim 778 \text{ nm}$  (O I at 777.41 nm, 777.63 nm and 777.75 nm  
447 forming a single peak). Sulfur and oxygen peak areas are obtained by integrating the spectral bins after  
448 baseline correction. As shown on figure 2 the increase of sulfate abundance and associated decrease of  
449 emission of other peaks from breakdown of the basalt matrix produces a specific shift to longer  
450 wavelengths for the spectral centroids at  $\sim 543 \text{ nm}$  and  $\sim 545 \text{ nm}$ , while simultaneously the normalized  
451 peak area at  $\sim 564 \text{ nm}$  increases. Using this approach, laboratory data suggest a threshold of sulfate  
452 detectability around 10 wt% sulfate with the possibility of obtaining a still lower threshold by more refined  
453 fitting and/or multivariate spectral fitting procedures.

454 *Calibration of the S signal.* The S abundance estimates based on 564 nm peak area are calibrated with *in*  
455 *situ* sulfate data collected on Mars. Indeed, bedrock of the upper Murray formation has a rather low bulk  
456 CaO content, but a large number of CaO enrichments were observed at the LIBS scale, related to Ca-



457 sulfates. The S association with Ca is observed as the data from Murray bedrock, after removal of other  
458 Fe and Mg-rich diagenetic features, follows the same trend of peak area increase at ~564 nm and centroid  
459 shifts observed in the laboratory. Additionally, measurements from the APXS instrument independently  
460 confirm the correlation of CaO with SO<sub>3</sub> content in the upper Murray formation (Figure S3). The SO<sub>3</sub>  
461 content calibration curve for the normalized peak area at ~564 nm (Figure 2-f) is obtained by attributing  
462 CaO enrichments from average bedrock above 2.5 wt% CaO to Ca-sulfate.

463 This *in situ* calibration curve obtained using Ca-sulfate data can then be used to predict sulfate content  
464 related to Mg-sulfates because tests in the laboratory have shown that for both Mg-sulfates and Ca-sulfates  
465 the normalized peak area increases along the same calibration curve and up to the same normalized peak  
466 area value for the pure sulfate pellets (Figure 2-c). The limit of detection on sulfate content using this  
467 approach is calculated from the 95% confidence bound of the calibration curve. Its intercept has a  
468 normalized peak area which corresponds to a predicted 10.6 wt% SO<sub>3</sub> content. Data from upper Murray  
469 shows that MgO enrichments are correlated with the normalized S peak area at ~ 564 nm, as well as  
470 centroid shifts (Figure 2), which confirm the presence of sulfate from the signal. The current calibration  
471 model to estimate major oxide contents<sup>48</sup> is not optimized yet to include Mg-sulfate bearing basaltic  
472 mixtures, and consequently, the total amount of oxides predicted exceeds 100% after water and sulfate  
473 contents are included. This was also found to be the case when laboratory Mg-sulfate bearing basaltic  
474 mixture major elements were calculated using the current calibration model. Consequently, assuming the  
475 proportions predicted for the major oxides are correct, the oxide amounts are then renormalized to account  
476 for water and sulfate content, so that totals equal 100%.

477 **Identification of the Mg-sulfate enriched lithology.** A correlation was identified between the occurrence  
478 of Mg sulfates in LIBS data on sol 1680 (Figure 5) and the specific lithology observed on Murray bedrock  
479 in color image data (Figure S7). MastCam images of bedrock or blocks of bedrock were analyzed to

480 identify the Mg-sulfate enriched lithology with characteristics defined by its 1) texture: nodular or  
481 massive, erosion resistant, forming centimeter-thick layers in some cases, 2) weathering style: cracking in  
482 poorly rounded centimeter-sized chunks which remain more resistant to erosion than typical Murray  
483 bedrock, 3) color: brown relative to typical Murray bedrock. The specific color was identified and mapped  
484 on MastCam images to verify the same color was observed on the lithology at the different visited sites.

485 *Color identification.* RGB pixel values of the radiometrically corrected MastCam images were converted  
486 into CIE L\*a\*b\* color space, with L\* lightness, and a\* and b\* color parameters (Figure S15). This  
487 determined the color and lightness parameters interval for the fragments of bedrock where the Mg-sulfate  
488 enrichments were analyzed with LIBS. The interval then was used as a guide to map areas of the bedrock  
489 not measured by LIBS, with color and lightness parameters characteristic of the Mg-sulfate enrichments.  
490 The lithology is also identified from texture, morphology and weathering style in addition to this color-  
491 mapping technique.

492 **Orbital data analysis.** The Compact Reconnaissance Imaging Spectrometer for Mars (CRISM) onboard  
493 the Mars Reconnaissance Orbiter collects targeted hyperspectral images of the surface with spatial  
494 resolution between 12 and 18 m/pixel. Absorption features due to atmospheric carbon dioxide are removed  
495 using the Volcano scan method<sup>52</sup>. The Discrete Ordinate Radiative Transfer model<sup>53</sup> is used to account  
496 for effects due to temporally and spatially variable atmospheric dust and ice aerosols. The observations  
497 shown on Figure S11 are map-projected and spectrally smoothed using an iterative log likelihood  
498 algorithm to incorporate the instrument's spatial and spectral transfer functions to retrieve the best estimate  
499 of the surface albedo in the presence of Poisson-distributed noise and spatial blur<sup>54</sup>. A Spectral absorption  
500 at 1.9  $\mu\text{m}$  is indicative of H<sub>2</sub>O within a hydrated mineral and convexity at 2.29  $\mu\text{m}$  measures the dual  
501 presence of the broad hydration absorption and the S-O bending or mineral H<sub>2</sub>O overtone commonly  
502 observed in hydrated sulfates<sup>55</sup>. Spectra are extracted from spatial regions of interest as determined by

503 the spatial parameterization, and spectral features are compared to those of laboratory standards to confirm  
504 mineral identifications.

505 **Mapping of vein distribution.** The presence of veins was mapped along the traverse using MastCam  
506 end-of-drive imaging of the surface in front of the rover. The area covered by the images is divided into  
507 typically 4 areas covering ~4 m<sup>2</sup> of Martian surface. In each area covering bedrock, the presence of veins  
508 is reported in vertical and horizontal categories depending on their apparent orientation being either close  
509 to vertical or horizontal. In addition, reference cases of dense vein networks are used to report qualitatively  
510 the presence of higher density for veins either vertical or horizontal (Figure S12).

511

512 **Data availability:** All *in situ* and orbital data used in this study are available in the NASA Planetary Data  
513 System ([pds-geosciences.wustl.edu](https://pds-geosciences.wustl.edu)). All other supplementary data that support the findings of this study  
514 are available from the corresponding author upon request.

515

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