



HAL
open science

Earth-like Habitable Environments in the Subsurface of Mars

J.D. D Tarnas, J.F. F Mustard, B. Sherwood Lollar, V. Stamenković, K.M. M Cannon, J.-P. Lorand, T.C. C Onstott, J.R. R Michalski, O. Warr, A.M. M Palumbo, et al.

► **To cite this version:**

J.D. D Tarnas, J.F. F Mustard, B. Sherwood Lollar, V. Stamenković, K.M. M Cannon, et al.. Earth-like Habitable Environments in the Subsurface of Mars. *Astrobiology*, 2021, 21 (6), pp.741-756. 10.1089/ast.2020.2386 . hal-03358460

HAL Id: hal-03358460

<https://hal.science/hal-03358460v1>

Submitted on 29 Sep 2021

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

1 Earth-like habitable environments in the subsurface of Mars

2

3 J.D. Tarnas¹, J.F. Mustard¹, B. Sherwood Lollar², V. Stamenković³, K.M. Cannon^{4,5}, J.-P.

4 Lorand⁶, T.C. Onstott⁷, J.R. Michalski⁸, O. Warr², A.M. Palumbo¹, A.-C. Plesa⁹

5

6 ¹Brown University Department of Earth, Environmental and Planetary Sciences, ²University of

7 Toronto Department of Earth Sciences, ³NASA Jet Propulsion Laboratory, California Institute of

8 Technology, ⁴Department of Geology and Geological Engineering, Colorado School of Mines,

9 ⁵Space Resources Program, Colorado School of Mines, ⁶Université de Nantes Laboratoire de

10 Planétologie et Géodynamique de Nantes, ⁷Princeton University Department of Geosciences,

11 ⁸University of Hong Kong Division of Earth & Planetary Science, ⁹German Aerospace Center

12 (DLR) Institute of Planetary Research

13

14 **Abstract**

15 In Earth's deep continental subsurface, where groundwaters are often isolated for $>10^6$ -

16 10^9 years, energy released by radionuclides within rock produces oxidants and reductants that

17 drive metabolisms of non-photosynthetic microorganisms. Similar processes could support past

18 and present life in the martian subsurface. Sulfate-reducing microorganisms are common in

19 Earth's deep subsurface, often using hydrogen derived directly from radiolysis of pore water and

20 sulfate derived from oxidation of rock-matrix-hosted sulfides by radiolytically-derived oxidants.

21 Radiolysis thus produces redox energy to support a deep biosphere in groundwaters isolated

22 from surface substrate input for millions to billions of years on Earth. Here we demonstrate that

23 radiolysis by itself could produce sufficient redox energy to sustain a habitable environment in

24 the subsurface of present-day Mars, one in which Earth-like microorganisms could survive
25 wherever groundwater exists. We show that the source localities for many martian meteorites are
26 capable of producing sufficient redox nutrients to sustain up to millions of sulfate-reducing
27 microbial cells per kg rock via radiolysis alone, comparable to cell densities observed in many
28 regions of Earth's deep subsurface. Additionally, we calculate variability in supportable sulfate-
29 reducing cell densities between the martian meteorite source regions. Our results demonstrate
30 that martian subsurface groundwaters, where present, would largely be habitable by sulfate-
31 reducing bacteria from a redox energy perspective via radiolysis alone.

32

33 **1.0 Introduction**

34 The surface of Mars is an extremely hostile environment characterized by freezing
35 temperatures, desiccating conditions, high levels of ionizing radiation, oxidizing chemicals, low
36 pressures, and a lack of liquid water that preclude any Earth-like organisms from surviving
37 without adaptation that is unprecedented on Earth. Though some extremophiles on Earth have
38 evolved to tolerate certain conditions relevant for Mars, any martian life formed in earlier eras
39 would have taken refuge in the subsurface for survival (National Academies of Sciences,
40 Engineering, and Medicine et al., 2018; Michalski et al., 2018), perhaps temporarily inhabiting
41 transiently habitable near-surface environments formed by meteorite impacts into ice-rich rock
42 (Schwenzer et al., 2012). Within the last three decades, researchers have discovered that fluids
43 preserved within Earth's kilometers deep subsurface contain significant quantities of biomass
44 utilizing a wide diversity of redox reactions to drive their microbial metabolisms (Onstott et al.,
45 2019). Similar habitable subsurface environments could potentially have existed on ancient Mars
46 (Onstott et al., 2019; Tarnas et al., 2018), may still exist on modern Mars (Michalski et al.,

47 2013), and could exist on other planetary objects including ocean worlds, small bodies like
48 Ceres, and exoplanets, making them an exciting frontier of planetary exploration (National
49 Academies of Sciences, Engineering, and Medicine et al., 2018; Stamenković et al., 2019).
50 Planetary objects with lower surface gravity than Earth, including Mars, likely have higher
51 volumes of habitable pore and fracture space that extend deeper into the crust compared to Earth
52 (e.g., Goossens et al., 2017; Lewis et al., 2019), increasing the volume of feasibly supportable
53 biomass if sufficient liquid water and redox energy is available (Sleep, 2012).

54 In Earth's subsurface, actively metabolizing microbes can be sustained by fluids that have
55 been isolated from surface substrate input for $>10^6$ - 10^9 years (Lin et al., 2006; Holland et al.,
56 2013; Li et al., 2016; Warr et al., 2018; Lollar et al., 2019)—similar to groundwater isolation
57 timescales expected on Mars (Grimm et al., 2017)—powered solely by redox energy derived
58 from water-rock reactions including radiolysis and serpentinization (Sherwood Lollar et al.,
59 2014; Warr et al., 2019). Radiolysis, the breaking of H₂O molecules in pore spaces by α , β , and γ
60 radiation released from decay of radionuclides within host rock, generates both dissolved
61 reductants (e.g. H₂; Lin et al., 2005b; Warr et al., 2019) and dissolved oxidants (e.g. H₂O₂; ref.
62 Lefticariu et al., 2010; Li et al., 2016). The dissolved H₂ generated by radiolysis can be used
63 directly as a reductant by subsurface microbes, while the primary oxidant byproducts of
64 radiolysis can oxidize sulfides within the host rock matrix to form dissolved sulfate (SO₄)
65 (Lefticariu et al., 2010; Li et al., 2016) that can then be used by microbes as an oxidant (Lin et
66 al., 2005b; Lin et al., 2006; Chivian et al., 2008; Li et al., 2016; Lollar et al., 2019). Sulfate-
67 reducing microorganisms are found kilometers deep on Earth in many groundwaters that have
68 been isolated for $>10^7$ years (Lin et al., 2005b; Lin et al., 2006; Chivian et al., 2008; Li et al.,
69 2016; Lollar et al., 2019), and a significant component of the H₂ and SO₄ in especially ancient

70 groundwaters is typically derived from radiolysis (Li et al., 2016; Warr et al., 2019). Radiation
71 doses from decay of rock-matrix-hosted radionuclides are insufficient to sterilize microbial
72 communities, as demonstrated by the presence of microbes in these subsurface settings (e.g., Lin
73 et al., 2005a; Lin et al. 2005b; Lin et al. 2006; Chivian et al., 2008; Lollar et al., 2019). Thus,
74 these Earth systems prove that in any subsurface environment containing radionuclides, liquid
75 H₂O, and sulfides, radiolysis alone has the potential to provide sufficient biologically-supportive
76 redox energy to sustain a subsurface biome (Onstott et al., 2006; Sherwood Lollar et al., 2014).
77 There are many alternative potential pathways to generate additional redox couples for life in
78 subsurface martian environments, including in fluid mixing zones that create chemical
79 disequilibria (Sherwood Lollar et al., 2007), which will not be considered here. Accordingly, the
80 gas production rates and supportable cell density ranges presented in this work should be
81 considered conservative estimates.

82

83 **2.0 Methods**

84 *2.1 Summary*

85 The model used to estimate H₂ production rates, sulfate production rates, dissolved H₂
86 concentrations, dissolved sulfate concentrations, and supportable sulfate-reducing cell densities
87 is similar to models employed in Lefticariu et al., (2010); Li et al., (2016); and Altair et al.,
88 (2018) (Section 2.2). Porosity (Section 2.3), sulfide concentrations (Section 2.4), average sulfide
89 grain sizes (Section 2.5), radionuclide concentrations (Section 2.6), and sulfate-reducing
90 metabolic rates are the key parameters necessary to constrain the model. H₂ production rates are
91 dependent on porosity and radionuclide concentrations, increasing as porosity and radionuclide
92 concentrations increase. Sulfate production rates are dependent on porosity, radionuclide

93 concentrations, sulfide concentrations, and average sulfide grain size. They increase as porosity,
94 radionuclide concentrations, and sulfide concentrations increase and decrease as sulfide grain
95 size increases, as less reactive surface area is exposed when sulfide grains are larger. Dissolved
96 H₂ and sulfate concentrations increase as porosity decreases, as there is less fluid in which to
97 dissolve the H₂ and sulfate that is produced. Supportable sulfate-reducing cell densities in units
98 of cells (kg rock)⁻¹ increase as porosity and sulfate production rates increase, since sulfate is the
99 limiting redox nutrient in all model results presented here. Supportable sulfate-reducing cell
100 densities in units of cells L⁻¹ decrease as porosity increases because there is less fluid per volume
101 rock for the cells to reside in. Supportable sulfate-reducing cell densities are higher when the
102 assumed sulfate-reducing metabolic rate is lower.

103 Porosity is constrained using density estimates for the martian crust from orbital
104 gravimetry (Goossens et al., 2017) that are supported by rover gravimetry (Lewis et al., 2019)
105 (Section 2.3). Sulfide concentrations (Section 2.4), average sulfide grain sizes (Section 2.5), and
106 radionuclide concentrations (Section 2.6) are constrained by their measured values in martian
107 meteorites. The maximum and minimum assumed sulfate-reducing metabolic rates are based on
108 the fastest and slowest sulfate-reducing metabolic rates estimated for fracture fluids in the gold
109 mines of the Witwatersrand Basin (Lin et al., 2006; Chivian et al., 2008). The calculated sulfate-
110 reducing cell densities are compared to those found in Earth's deep subsurface (Onstott et al.,
111 2003; Cockell et al., 2012; Magnabosco et al., 2018; Onstott et al., 2019) (Fig. 2).

112 The amount of radiolytic hydrogen and sulfate that would be produced in each of the
113 different martian meteorite source regions is calculated based on their individual K, Th, and U
114 concentrations (Fig. 1e, Table 2), sulfide concentrations (Fig. 1f, Table 1), average sulfide grain
115 sizes (Fig. 1f, Table 1), and estimated range of porosity (Fig. 1c, Table 2) before the impact

116 events that ejected these rocks from the martian gravity well. Based on the produced amounts of
117 these redox nutrients, the number of supportable sulfate-reducing cells per kg rock is calculated
118 for each of these meteorite source regions using the range of sulfate-reducing metabolic rates
119 observed in Earth's deep subsurface (Lin et al., 2006; Chivian et al., 2008). Porosity range
120 assumptions are based on density estimates of different martian terrains from orbital gravimetry
121 (Fig. 1c-d; Section 2.2) (Goossens et al., 2017), a range that is supported by rover gravimetry
122 (Lewis et al., 2019). The radiolysis model applied here (Section 2.3) is similar to the one
123 employed in Lin et al., (2005b); Li et al., (2016); Dzaugis et al., (2018); Tarnas et al., (2018); and
124 Warr et al., (2019).

125

126 *2.2 Radiolysis model*

127 The model used here follows the radiolytic H₂ and SO₄ production models presented in Lin
128 et al., (2005a); Lin et al., (2005b); Li et al., (2016); Tarnas et al., (2018); and Warr et al., (2019).

129 This calculates radiogenic dose rate as

$$130 \quad (1) E_{net,i} = \frac{E_i \times W \times S_i}{1 + W \times S_i}$$

131 where i is α , β , or γ radiation, E_{net} is the net dosage absorbed by pore water (eV kg⁻¹ s⁻¹), E is
132 the apparent dosage from radioactive element decay (eV kg⁻¹ s⁻¹), W is the weight ratio of pore
133 water to rock, and S is the stopping power of minerals within the rock matrix with respect to
134 each type of radiation particle/ray, where $S_\alpha = 1.5$, $S_\beta = 1.25$, $S_\gamma = 1.14$ (Lin et al., 2005a; Lin
135 et al., 2005b; Tarnas et al., 2018; Warr et al., 2019). The stopping power of minerals can be
136 estimated using Bragg's law (Bragg and Kleeman, 1905) and typically varies by <0.5 for silicate
137 minerals (Nogami and Hurley, 1948).

138 W , the weight ratio of pore water to rock, primarily depends on the rock porosity with
 139 additional minor variations induced by differences in water density in solid ($\sim 900 \text{ kg m}^{-3}$) or
 140 liquid ($\sim 1000 \text{ kg m}^{-3}$) phase. Values from Adamiec and Aitken, 1998 are used for E for 1 wt. %
 141 K, 1 ppm Th, and 1 ppm U for α , β , and γ radiation, which are $E_{K,\alpha} = 0 \text{ (Gy ka}^{-1}\text{)}$, $E_{K,\beta} = 0.782$
 142 $\text{(Gy ka}^{-1}\text{)}$, $E_{K,\gamma} = 0.243 \text{ (Gy ka}^{-1}\text{)}$, $E_{Th,\alpha} = 0.061 \text{ (Gy ka}^{-1}\text{)}$, $E_{Th,\beta} = 0.027 \text{ (Gy ka}^{-1}\text{)}$, $E_{Th,\gamma} =$
 143 $0.048 \text{ (Gy ka}^{-1}\text{)}$, $E_{U,\alpha} = 0.218 \text{ (Gy ka}^{-1}\text{)}$, $E_{U,\beta} = 0.146 \text{ (Gy ka}^{-1}\text{)}$, $E_{U,\gamma} = 0.113 \text{ (Gy ka}^{-1}\text{)}$. 1 Gy
 144 $= 6.2415 \times 10^{18} \text{ eV kg}^{-1}$.

145 H_2 yield is calculated as

$$146 \quad (2) Y_{H_2} = \sum E_{net,i} \times G_{H_2,i}$$

147 where Y_{H_2} is the total yield of H_2 molecules ($\text{molecules kg}^{-1} \text{ s}^{-1}$) and G_{H_2} is the number of H_2
 148 molecules produced per 100 eV of radiation. The conversion factor between (kg water) and (kg
 149 water and rock matrix) is W , the weight ratio of pore water to rock. For H_2O , $G_{H_2,\alpha} = 0.96 \text{ H}_2$
 150 $\text{molecules (100 eV)}^{-1}$, $G_{H_2,\beta} = 0.6 \text{ H}_2 \text{ molecules (100 eV)}^{-1}$, $G_{H_2,\gamma} = 0.4 \text{ H}_2 \text{ molecules (100 eV)}$
 151 $^{-1}$ (Lin et al., 2005a; Lin et al., 2005b; Tarnas et al., 2018; Warr et al., 2019). The value of G_{H_2}
 152 changes depending on the liquid or solid substance filling the pore space, and has received
 153 extensive study due to its importance in the context of nuclear waste storage. It is typically
 154 higher for chloride brines than for pure water (LaVerne and Tandon, 2005; Buck et al., 2012)
 155 due to the reaction of dissolved brine ions (e.g. Cl^-) with ions formed via radiative destruction of
 156 neutral H_2O molecules (e.g. $\text{OH}\cdot$). This causes a depletion of ions that would react with $\text{H}\cdot$
 157 generated by radiolysis, leaving more $\text{H}\cdot$ in solution, which reacts to form H_2 ($2\text{H}\cdot \rightarrow \text{H}_2$) (Klein
 158 et al., 2020). Hydrated chloride salts, embedded in either pore space or within the rock matrix,
 159 will also produce H_2 , though at lower production efficiencies relative to pure water, ice, or brine
 160 (LaVerne and Tandon, 2005). These different production efficiencies are summarized in Tarnas

161 et al., 2018 and references therein. Presence of certain minerals, such as zeolites, can
 162 significantly increase H₂ and oxidant production efficiency from radiolysis (Kumagai et al.,
 163 2013). Zeolites are found in both bedrock (Ehlmann et al., 2009) and dust (Ruff, 2004) on Mars,
 164 thus this mineral is expected to enhance radiolytic H₂ and oxidant production in some parts of the
 165 crust, but this effect is not modeled here in order to focus on the most conservative estimates
 166 possible.

167 SO₄ production is calculated as

$$168 \quad (3) Y_{SO_4} = \sum E_{net,i} \times G_{SO_4,i}$$

169 where Y_{SO_4} is the total yield of SO₄ molecules (moles m⁻² sulfide year⁻¹) and G_{SO_4} is the number
 170 of SO₄ molecules produced per m² sulfide per Gy radiation. A G_{SO_4} value from Leticariu et al.,
 171 (2010) is used that is also used in Li et al., (2016) and Altair et al., (2018), which is 2.1×10^{-9}
 172 mol m⁻² Gy⁻¹. However, while the experiments of Leticariu et al., (2010) were performed under
 173 high-energy radiation, they showed that G_{SO_4} increases under low-radiation-dose conditions,
 174 which correspond to typical radiation doses in natural systems (Li et al., 2016). Furthermore,
 175 these experiments only investigated the oxidation, structural damage, and chemical weathering
 176 of sulfides exposed to γ radiation. As Leticariu et al., (2010) note, accounting for increased
 177 oxidation, structural damage, and chemical weathering of sulfides from α and β radiation—
 178 which typically represent >90% of total irradiated energy—would increase the G_{SO_4} value,
 179 perhaps significantly. This means once again that the calculated sulfate production rates, and
 180 thus supportable sulfate-reducing cell densities herein, are likely conservative estimates. The
 181 concentration of sulfate in fluid from radiolytic production is expressed as

$$182 \quad (4) C_{SO_4} = Y_{SO_4} \times C_{sulfide} \times S_{sulfide} / W$$

183 where C_{SO_4} is the production rate of dissolved sulfate ($M \text{ year}^{-1}$), $C_{sulfide}$ is the concentration of
184 sulfide in the rock matrix (volume ratio), and $S_{sulfide}$ is the average specific surface area of
185 sulfides in the rock matrix ($m^{-2} \text{ sulfide/kg}$). $C_{sulfide}$ values used in this study are listed in Table
186 2, which encompasses the values found in martian meteorites (Greenwood et al., 2000b;
187 Greenwood et al., 2000a; Rochette et al., 2001; Lorand et al., 2005; Rochette et al., 2005;
188 Chevrier et al., 2011; Lorand et al., 2015; Baumgartner et al., 2017; Jean-Pierre Lorand et al.,
189 2018b; Jean-Pierre Lorand et al., 2018a). $S_{sulfide}$ values ranging from $2.26 \times 10^1 \text{ m}^2 \text{ kg}^{-1}$ and
190 $1.41 \times 10^3 \text{ m}^2 \text{ kg}^{-1}$ are used, which correspond to average sulfide grain sizes of 2-125 μm . The
191 $S_{sulfide}$ values for each meteorite class are assigned based on the range of sulfide grain sizes
192 measured in those martian meteorites (Fig. 1f; Tables 1 & 2). Finally, following Altair et al.,
193 (2018), minimum and maximum sulfate-reducing metabolic rates of $5.5 \times 10^{-18} - 3.6 \times 10^{-17}$
194 moles $\text{cell}^{-1} \text{ year}^{-1}$ are assumed, which correspond to the minimum and maximum sulfate-
195 reducing metabolic rates estimated in deep subsurface fracture fluids of the gold mines from the
196 Witwatersrand Basin (Lin et al., 2006; Chivian et al., 2008).

197

198 *2.3 Porosity model*

199 The average porosity ranges for the martian meteorite source regions are estimated using
200 density estimates for the different regions of Mars' crust from orbital gravimetry (Goossens et
201 al., 2017) that are validated by rover gravimetry in Gale crater that estimates a porosity range of
202 $40 \pm 6\%$ for the top ~300 meters of bedrock (Lewis et al., 2019). Porosity in this context includes
203 both fracture and pore space within rocks. The orbital gravimetry estimates give average
204 densities of $\sim 1600 \text{ kg m}^{-3}$ for the southern highlands, 2400 kg m^{-3} for the northern lowlands, and
205 $\sim 2800 \text{ kg m}^{-3}$ for the volcanic bulges. It is assumed that the volcanic bulges and northern

206 lowlands are the possible source regions for the SNCs and the southern highlands are the source
207 region for the regolith breccias (Cassata et al., 2018). In the estimation, an average solid crust
208 matrix density range of 2000-3200 kg m⁻³ is used for the southern highlands crust, consistent
209 with its mineralogy (Ehlmann and Edwards, 2014), and 2600-3200 kg m⁻³ is used for the
210 northern lowlands and volcanic bulges, consistent with volcano-specific orbital gravimetry
211 (Broquet and Wieczorek, 2019). It is assumed the difference between the gravimetry density
212 measurements and rock matrix density estimates is caused by pore space filled with ice or water.
213 The densities add linearly such that $\rho_{bulk} = \rho_{rock}(1 - \theta) + \rho_{H_2O}(\theta)$ where θ is porosity. This
214 means the composition of the southern highlands rock is assumed to vary, on average, between
215 pure phyllosilicate (~2000 kg m⁻³) and dense basalt (~3200 kg m⁻³), while the composition of the
216 northern lowlands and volcanic bulges rocks are assumed to vary between 50-50 phyllosilicate-
217 basalt mixture (~2600 kg m⁻³) and dense basalt (~3200 kg m⁻³). Given uncertainties in density
218 estimates from orbital gravimetry, to be conservative, estimated porosities were reduced by 10%.
219 This does not model depth dependence of porosity, but rather the average porosity of the source
220 regions for the individual martian meteorites. This gives approximate average porosity ranges of
221 15-35% for the regolith breccias, 5-15% for the SNCs. The SNC porosity assumptions are
222 supported by images of porous float vesicular basalt rocks in Gusev crater imaged by the Spirit
223 rover (McMahon et al., 2013; McSween, 2015).

224

225 *2.4 Sulfide concentrations*

226 Martian meteorites contain sulfides of both magmatic (pyrrhotite) (Lorand et al., 2005;
227 Lorand et al., 2018b) and hydrothermal (pyrite) (Lorand et al., 2015; Wittmann et al., 2015;
228 Lorand et al., 2018) origins (Sections 2.4 & 2.5). Sulfide abundances range from <0.01-1.0 wt.

229 %. The assumed sulfide concentration ranges are derived based on these sulfide concentrations
230 found in martian meteorites (Greenwood et al., 2000b; Greenwood et al., 2000a; Rochette et al.,
231 2001; Lorand et al., 2005; Rochette et al., 2005; Chevrier et al., 2011; Franz et al., 2014; Lorand
232 et al., 2015; Hewins et al., 2017; Wang and Becker, 2017; Baumgartner et al., 2017; Jean-Pierre
233 Lorand et al., 2018b; Lorand et al., 2018a; Franz et al., 2019; Hewins et al., 2020) (Fig. 1f; Table
234 1). Martian meteorites contain variable abundances of sulfides with relatively consistent grain
235 sizes for grains >10 μm . Published abundances for sulfides in martian meteorites, and reported
236 average grain sizes and grain size range, are presented in Table 1. Magmatic sulfides (pyrrhotite)
237 and hydrothermal sulfides (pyrite) are typically associated with SNCs and regolith breccias,
238 respectively (Greenwood et al., 2000b; Greenwood et al., 2000a; Rochette et al., 2001; Lorand et
239 al., 2005; Rochette et al., 2005; Chevrier et al., 2011; Lorand et al., 2015; Baumgartner et al.,
240 2017; Lorand et al., 2018b; Lorand et al., 2018a). Pyrite is much less abundant than pyrrhotite as
241 it was described only in an orthopyroxenite (ALH84001), two chassignites and a few nakhlites
242 (as minor pyrrhotite replacement product), as well as in regolith breccias (as primary
243 hydrothermal precipitate). Sulfide abundances are typically higher in regolith breccias than in
244 SNCs. The listed sulfide abundances are also minimum abundances, as desert weathering of
245 martian meteorite samples causes oxidation of sulfides and partial S losses (Greenwood et al.,
246 2000b; Greenwood et al., 2000a; Rochette et al., 2001; Lorand et al., 2005; Rochette et al., 2005;
247 Chevrier et al., 2011; Lorand et al., 2015; Baumgartner et al., 2017; Lorand et al., 2018b; Lorand
248 et al., 2018a).

249 The original report of sulfides in Gale crater via CheMin XRD data is no longer supported
250 after recalibration of the data (Morrison et al., 2018), but evolved gas analysis-mass spectroscopy
251 (EGA-MS) (Wong et al., 2020) and sulfur isotope measurements using the Sample Analysis at

252 Mars (SAM) instrument (Franz et al., 2017) support the presence of sulfides in multiple Gale
253 crater samples. Sulfides in Gale crater are diagenetic/epigenetic, likely resulting from thermal
254 reduction of preexisting sulfates (Franz et al., 2017).

255

256 *2.5 Sulfide grain sizes*

257 Average grain sizes for sulfides in martian meteorites range from 5-100 μm with
258 occasional outliers up to 400 μm (Fig. 1f; Table 1 and refs. therein). Sulfide grain sizes are
259 relatively consistent across martian meteorite samples (Fig. 1f). A compilation of published
260 measured sulfide average grain sizes and grain size ranges in martian meteorites is presented in
261 Table 1 and plotted in Fig. 1f. It is noteworthy that many of these studies do not consider sulfides
262 $<10 \mu\text{m}$ in diameter, thus these results may skew average sulfide grain sizes towards larger-than-
263 true values. Since sulfate production in the calculations increases as sulfide grain size decreases,
264 as a result of greater specific surface area, this ensures the calculations are conservative. Sulfide
265 grain sizes depend on a wide array of parameters, including temperature, supersaturation of
266 fluids, nucleation rates, and fracturing. For each individual martian meteorite class, the range of
267 measured average sulfide grain sizes in those meteorites are used as the basis for the calculations.

268

269

270

271

272

273

274

275
276
277
278
279
280
281
282
283

<i>Sample</i>	<i>Sulfide abundance (wt. %)</i>	<i>Average grain size (μm)</i>	<i>Grain size min (μm)</i>	<i>Grain size max (μm)</i>	<i>Publications</i>	<i>Type</i>
<i>NWA 998</i>	0.13	-	20	120	a	Nakhlite
<i>Nakhla</i>	0.07 (0.08)	-	10	50	a,b	Nakhlite
<i>Governador Valadares</i>	0.06	-	2	25	a	Nakhlite
<i>NWA 817</i>	0.03	-	2	30±15	a	Nakhlite
<i>MIL 03346</i>	0.05	-	-	-	c	Nakhlite
<i>And paired samples</i>						
<i>Y000593</i>	0.08	-	-	-	b	Nakhlite
<i>ALH A77005</i>	0.3 ^d , 0.12 ^c , 0.19 ^b	100	-	-	b-d	Lherzolitic shergottites
<i>LEW 88516</i>	0.3	17.5	-	-	d	Lherzolitic shergottites
<i>Y-293605</i>	0.05	-	-	-	d	Lherzolitic shergottites
<i>Tissint</i>	0.6 ^e , 0.35 ^c	-	<1	200	c,e	Olivine-phyric shergottite
<i>EET</i>	0.52 ^b , 0.28 ^c	-	-	-	c	Olivine-phyric shergottite
<i>A79001-A</i>						
<i>DaG 476/489</i>	0.45	50	10	100	d	Olivine-phyric shergottites
<i>SaU 005/094</i>	0.30 ^d , 0.16 ^c , 0.38 ^b	30	10	50	b-d	Olivine-phyric shergottites
<i>Dhofar 019</i>	0.5 ^d , 0.20 ^c	25	-	-	c, d	Olivine-phyric shergottites
<i>NWA 1068/1110</i>	0.8	-	-	-	c, d	Olivine-phyric shergottites
<i>Y-980459</i>	0.1 ^c , 0.4 ^b	30	5	50	b-d, f	Olivine-phyric shergottites
<i>Zagami</i>	0.7 ^d , 0.24 ^c , 0.35 ^b	<10	100	200	b-d	Basaltic shergottite
<i>Shergotty</i>	0.4	-	100	200	d	Basaltic shergottite
<i>EET</i>	0.12	-	-	-	c	Basaltic shergottite
<i>A79001-B</i>						
<i>QUE 94201</i>	0.3 ^d , 0.13 ^c	-	-	-	d	Basaltic shergottite
<i>NWA 480</i>	0.24	-	100	200	d	Basaltic shergottite
<i>NWA 856</i>	0.1	-	-	-	d	Basaltic shergottite
<i>Los Angeles</i>	0.67 ^d , 0.32 ^c	-	200	400	d	Basaltic shergottite
<i>Y000097</i>	0.12	-	-	-	b	Lherzolitic shergottite
<i>LAR 12011</i>	0.35	-	-	-	b	Lherzolitic shergottite
<i>LAR12095</i>	0.5	-	-	-	b	Lherzolitic shergottite
<i>Mean of 37</i>	0.21±0.18	-	-	-	c, g	Shergottites

<i>shergottites</i>						
NWA 7533	1±0.1	35	-	-	h, i	Regolith breccia
Chassigny	0.02	20	1-5	30	j	Chassignite
NWA 2737	0.015 ± 0.005	-	3	60	k	Chassignite
NWA 8693	<0.01	-	1	20	l	Chassignite

284 **Table 1 | Sulfide concentrations and grain sizes in martian meteorites. Assume density of**

285 **5000 kg/m³ for sulfide and 3000 kg/m³ for host rock to convert from vol % to wt %.** Franz

286 et al. (2014, 2019) and Wang and Becker (2017) data are calculated from whole-rock S contents

287 assuming 38 wt.% S for pyrrhotite and 54 wt. % for pyrite. Other sulfide abundances are from

288 image analyses on polished thick sections. ^a(Chevrier et al., 2011), ^b(Wang and Becker, 2017),

289 ^c(Franz et al., 2014), ^d(Lorand et al., 2005 and refs. therein), ^e(Gattacceca et al., 2013),

290 ^f(Baumgartner et al., 2017), ^g(Franz et al., 2019), ^h(Lorand et al., 2015), ⁱ(Lorand et al., 2018a),

291 ^j(Lorand et al., 2018b), ^k(Lorand et al., 2012), ^l(Hewins et al., 2020).

292

293 2.6 Radionuclide concentrations

294 Martian meteorites and remote measurements by the Gamma Ray Spectrometer (GRS)

295 onboard Mars Odyssey (Boynton et al., 2007), which is sensitive to the top ~tens of cm of the

296 martian crust, provide first-order constraints for radionuclide concentrations in the martian

297 subsurface (Fig. 1). Radionuclide concentrations in the top ~tens of cm in the martian crust vary

298 with latitude and longitude (Fig. 1a-b), and volcanic shergottite-nakhlite-chassignite (SNC)

299 martian meteorites generally have lower radionuclide contents than regolith breccias (Fig. 1e)

300 that are likely representative of the Noachian crust (Cassata et al., 2018) (Fig. 1). Elemental

301 concentrations in martian meteorites range from ~0.01-0.4 wt. % K, ~0.01-2 ppm Th, and

302 ~0.003-0.36 ppm U (Onstott et al., 2006; Meyer, 2016) (Fig. 1e). Some U enrichment in these

303 samples is possible through desert contamination (Tarnas et al., 2018). K and Th concentrations

304 estimated from GRS measurements range from ~0.07-0.6 wt. % and ~0.015-1.1 ppm (Boynton et

305 al., 2007; Tarnas et al., 2018), respectively (Fig. 1a-b,e).

306 For calculations of SO₄ and H₂ production in martian meteorites, the measured values for K,
 307 Th, and U in each individual meteorite class were used (Onstott et al., 2006; Meyer, 2016),
 308 integrating both the maximum and minimum radionuclide concentration values measured (Fig.
 309 1e; Table 2). The K and Th concentration ranges for SNCs are substantially lower than K and Th
 310 concentrations measured in GRS data (Fig. 1e), meaning the majority of the martian crust at
 311 depths shallower than ~0.5 m typically has higher radionuclide concentrations than are
 312 represented by the SNCs. Following Hahn et al., (2011) and Tarnas et al., (2018), the GRS data
 313 are normalized to correct for contributions from Cl, H, and S. K concentrations measured by
 314 Mars rovers range from 0.05 wt. % measured by Spirit to 4.4 wt. % measured by Curiosity
 315 (Gellert et al., 2004; Deit et al., 2016).

316

317

<i>Category</i>	<i>K₂O concentration (wt %)</i>	<i>Th concentration (ppm)</i>	<i>U concentration (ppm)</i>	<i>Sulfide concentration (vol %)</i>	<i>Sulfide grain size (um)</i>	<i>Porosity (%)</i>
<i>Regolith breccia</i>	0.42-0.51 ^{a,b,c}	1.1-2 ^{a,b,c}	0.29-0.36 ^{a,b,c}	0.6±0.1 ^{a,b,c}	2-100 ^{a,b,c}	15-35
<i>Chassignite</i>	0.036-0.05 ^d	0.057-0.13 ^d	0.018-0.056 ^d	0.006-0.012 ^{e,f}	2-60 ^{e,f}	5-15
<i>Nakhilite</i>	0.11-0.27 ^d	0.15-0.42 ^d	0.052-0.23 ^d	0.018-0.078 ^g	2-50 ^g	5-15
<i>Shergottite</i>	0.0156-0.35 ^d	0.0117-0.57 ^d	0.00303-0.12 ^d	0.03-0.6 ^h	10-200 ^h	5-15

318 **Table 2 | Range of values for martian meteorites calculations.** ^aAgee et al. (2013), ^bWittman

319 et al. (2015), ^cLorand et al. (2018a), ^dMeyer et al. (2016), ^eLorand et al. (2018b), ^fLorand et al.

320 (2012), ^gChevrier et al. (2011), ^hLorand et al. (2005), ⁱLe Deit et al. (2016), ^jBoynton et al.

321 (2007), ^kHahn et al. (2011), ^lTarnas et al. (2018), ^mVaniman et al. (2014). Sulfide grain size

322 excludes the few unusually large sulfide grains reported in each meteorite class.

323

324

325 **3.0 Results**

326 *3.1 Habitable redox conditions in martian meteorite source regions*

327 The results presented here show that the shergottite, nakhlite, chassignite, and regolith
328 breccia martian meteorite source localities would all produce sufficient redox energy to support
329 sulfate-reducing life where liquid groundwater exists within these lithologies. In all cases, based
330 on the calculations herein, sulfate is the limiting redox nutrient in comparison with H₂. The
331 regolith breccia source locality could support significantly more sulfate-reducing life than the
332 SNC localities because it has higher concentrations of both radionuclides and sulfides. Of the
333 SNCs, the shergottite and nakhlite source localities could support the highest densities of sulfate-
334 reducing cells. Key findings are illustrated in Fig. 2, displayed in Table 3, and are summarized
335 below as:

336 (1) *Regolith breccia source locality*: The sulfate production rates for the regolith breccia
337 source locality vary from $\sim[2 \times 10^{-14}] - [7 \times 10^{-12}]$ moles (kg rock)⁻¹ year⁻¹, while H₂
338 production rates range from $\sim[1-3] \times 10^{-11}$ moles (kg rock)⁻¹ year⁻¹, which could support
339 $[7 \times 10^2]-[1 \times 10^6]$ sulfate-reducing cells per kg rock. The regolith breccia source locality
340 would generate the most redox energy for sulfate-reducing microorganisms compared to
341 the other martian meteorite source localities.

342 (2) *Shergottite source locality*: The sulfate production rates for shergottite source localities
343 vary from $\sim[8 \times 10^{-18}] - [2 \times 10^{-13}]$ moles (kg rock)⁻¹ year⁻¹, while H₂ production rates
344 range from $\sim[4 \times 10^{-14}]-[4 \times 10^{-12}]$ moles (kg rock)⁻¹ year⁻¹, which could support $[2 \times 10^$
345 $^1]-[3 \times 10^4]$ sulfate-reducing cells per kg rock wherever liquid water exists.

346 (3) *Nakhlite source locality*: The sulfate production rates for nakhlite source localities vary
347 from $\sim[1 \times 10^{-16}] - [2 \times 10^{-13}]$ moles (kg rock)⁻¹ year⁻¹, while H₂ production rates range

348 from $\sim[5 \times 10^{-13}]$ - $[6 \times 10^{-12}]$ moles (kg rock)⁻¹ year⁻¹, which could support $[3]$ - $[3 \times 10^4]$
 349 sulfate-reducing cells per kg rock.

350 (4) *Chassignite source locality*: The sulfate production rates for chassignite source localities
 351 vary from $\sim[1 \times 10^{-17}]$ - $[6 \times 10^{-15}]$ moles (kg rock)⁻¹ year⁻¹, while H₂ production rates
 352 range from $\sim[2 \times 10^{-13}]$ - $[2 \times 10^{-12}]$ moles (kg rock)⁻¹ year⁻¹, which could support $[4 \times 10^$
 353 $^1]$ - $[1 \times 10^3]$ sulfate-reducing cells per kg rock.

354
 355

	<i>Radiolytic H₂ production rate in moles (kg rock)⁻¹ year⁻¹</i>	<i>Radiolytic Sulfate production rate in moles (kg rock)⁻¹ year⁻¹</i>	<i>Supportable sulfate-reducing cell density in cells (kg rock)⁻¹</i>
<i>Regolith breccia</i>	$\sim[1-3] \times 10^{-11}$	$\sim[2 \times 10^{-14}] - [7 \times 10^{-12}]$	$[7 \times 10^2]$ - $[1 \times 10^6]$
<i>Shergottite</i>	$\sim[4 \times 10^{-14}]$ - $[4 \times 10^{-12}]$	$\sim[8 \times 10^{-18}] - [2 \times 10^{-13}]$	$[2 \times 10^{-1}]$ - $[3 \times 10^4]$
<i>Nakhlite</i>	$\sim[5 \times 10^{-13}]$ - $[6 \times 10^{-12}]$	$\sim[1 \times 10^{-16}] - [2 \times 10^{-13}]$	$[3]$ - $[3 \times 10^4]$
<i>Chassignite</i>	$\sim[2 \times 10^{-13}]$ - $[2 \times 10^{-12}]$	$\sim[1 \times 10^{-17}]$ - $[6 \times 10^{-15}]$	$[4 \times 10^{-1}]$ - $[1 \times 10^3]$

356 **Table 3 | Estimated H₂ and sulfate production rates for martian meteorite source regions**
 357 **and associated supportable sulfate-reducing cell densities.**

358

359 The sulfate production rates from these calculations are several orders of magnitude lower
 360 than those of Lefticariu et al., (2010) because those authors assumed 5-10 wt. % pyrite in the
 361 martian crust, while this study uses lower sulfide concentrations based on measured values from
 362 martian meteorites (Fig. 1f, Table 1). Because the model uses the lowest and highest measured
 363 sulfate-reducing metabolic rates in Witwatersrand Basin for calculating cell density estimates
 364 (Lin et al., 2006; Chivian et al., 2008), there is a difference between the estimated maximum and
 365 minimum range for sulfate production rates and the estimated maximum and minimum range for
 366 number of supportable sulfate-reducing cells per kg rock. H₂ production estimates, in units of H₂

367 dissolved in water, range from 0.002-0.165 nM H₂/year. This compares to estimates by Dzaugis
368 et al., (2018) of 0.001-1.2 nM H₂/year for proposed Mars landing sites. The differences in the
369 dissolved H₂ estimates between this study and Dzaugis et al., (2018) derive from differences in
370 radionuclide concentrations and porosity used for calculations in the two studies.

371 The number of supportable sulfate-reducing cells per kg rock in the nakhlite, shergottite, and
372 regolith breccia source localities are comparable to those found in Earth's deep subsurface ($\sim 10^4$
373 $- 10^6$ cells per kg rock; Onstott et al., 2019) (Fig. 2). Estimates for the chassignite source locality
374 meanwhile are slightly lower ($\sim 10^1 - 10^3$ cells per kg rock). In all cases, sulfate is the redox
375 nutrient that limits sustainable cell densities, and as such estimated cell densities are most
376 sensitive to estimated sulfate production rates. The martian regolith breccia NWA 7034 and one
377 of its paired samples, NWA 7533, contain the highest radionuclide and sulfide concentrations of
378 the currently known martian meteorites, and thus their source locality would host sulfate
379 production rates capable of supporting the greatest density of sulfate-reducing microorganisms
380 relative to other known martian meteorites. Regolith breccia meteorites are likely representative
381 of the southern highlands, which is a major fraction of Mars' total crustal volume. The results
382 presented here support growing evidence that the subsurface is potentially the largest, longest-
383 lived, and most consistently habitable environment on Mars (Michalski et al., 2018; Michalski et
384 al., 2013; Onstott et al., 2019; Tarnas et al., 2018) and could serve as a refugia for any life that
385 ever existed on the martian surface (National Academies of Sciences, Engineering, and Medicine
386 et al., 2018; Michalski et al., 2018). Other viable processes could produce redox nutrients to
387 sustain additional metabolisms in the martian subsurface, including H₂ and CH₄ production via
388 serpentinization and Fischer-Tropsch-type synthesis, respectively (e.g. Warr et al., 2019),
389 atmospheric O₂ dissolution (Stamenković et al., 2018), atmospheric H₂ and CO dissolution

390 (Weiss et al., 2000), as well as Fe^{2+} and Fe^{3+} from minerals (Onstott et al., 2019). If sulfides are
391 not present in the rock matrix, these other reactions may be favorable over sulfate reduction.
392 These results provide scientific justification for future missions to search for signs of extant life
393 in the martian subsurface—which is an intriguing next frontier for planetary exploration
394 (National Academies of Sciences, Engineering, and Medicine et al., 2018; Stamenković et al.,
395 2019)—and identifies key characteristics of optimal regions for future landing sites.

396 Even the relatively small sampling of Mars' petrologic diversity via four martian meteorite
397 classes (shergottites, nakhlites, chassignites, regolith breccias) shows a wide range in redox
398 energy production rates from these different rock types/localities (Fig. 2). Some of these
399 localities, such as the source regions for regolith breccias, shergottites, and nakhlites could
400 support comparable concentrations of extant life to those found in Earth's deep subsurface
401 (Onstott et al., 2003; Cockell et al., 2012; Onstott et al., 2019). As is demonstrated by the
402 diversity of radionuclide concentrations, sulfide concentrations, and sulfide grain sizes in martian
403 meteorites, some regions of the martian crust will support higher sulfate-reducing cell densities
404 than others. Because sulfate is likely the limiting nutrient for hydrogenotrophic sulfate-reducing
405 metabolisms in a martian subsurface biosphere rather than H_2 , the highest concentrations of
406 sulfate-reducing microorganisms would exist in a water-bearing crustal section with high sulfide
407 abundance, low sulfide grain sizes, and relatively high radionuclide concentrations. Regions rich
408 in zeolites, which enhance radiolytic gas production (Kumagai et al., 2013), would also sustain
409 higher sulfate-reducing bacteria concentrations. Future missions should focus on characterizing
410 where these criteria are met, as these would be prime landing site targets for extant life
411 investigation from a redox energy perspective.

412

413 **4.0 Discussion**

414 *4.1 Potential for higher cell densities elsewhere on Mars*

415 The possibility of relatively high sulfide concentrations on Mars has been recognized for
416 decades due to the presence of ultramafic lithologies there (Burns and Fisher, 1990; Baumgartner
417 et al., 2015; Humayun et al., 2019) (Section 4.2). Concentration of magmatic sulfides are
418 assumed to derive from assimilation of crustal sulfates and/or S-rich regolith in lava flows, as
419 sampled by nakhlites, which likely crystallized from a sulfide-saturated melt after this
420 assimilation (Franz et al., 2014; Mari et al., 2019). Additional evidence for the existence of
421 concentrated martian sulfides has continued to accrue in the form of sulfides in martian
422 meteorites (Table 1 and refs. therein), in Gale crater (Franz et al., 2017; Wong et al., 2020), the
423 prevalence of sulfate-hematite assemblages on the martian surface (e.g. Wiseman et al., 2008),
424 which may or may not have formed as weathering products of sulfides (e.g. Zolotov and Shock,
425 2005), verifiable orbital detections of serpentine outcrops in Noachian terrains (Leask et al.,
426 2018), which are often associated with formation of reduced sulfides and metal alloys on Earth
427 (e.g. Economou and Naldrett, 1984; Thalhammer et al., 1986; Shiga, 1987; Auclair et al., 1993;
428 Wafik et al., 2001; Marques et al., 2007), Cu concentrations in Gale crater that are likely caused
429 by presence of sulfides (Payré et al., 2019), and the low oxygen fugacity (Mari et al., 2019 and
430 refs. therein) and high sulfur fugacity of the ancient martian mantle relative to Earth's (Wang and
431 Becker, 2017; Mari et al., 2019 and refs. therein). Pyrrhotite hosts magnetization in multiple
432 classes of martian meteorites (Rochette et al., 2005 and refs. therein) and thus at high enough
433 concentrations its subsurface presence could explain Mars' crustal magnetic field anomalies
434 (Langlais et al., 2019; Johnson et al., 2020).

435 High concentrations of sulfides could form via differentiation of sulfide-saturated lavas that
436 were contaminated by crustal assimilation of sulfates (Burns and Fisher, 1990; Baumgartner et
437 al., 2015) as occurred in nakhlite lava flows (Franz et al., 2014; Mari et al., 2019), differentiation
438 of impact melt accompanied by crustal sulfur assimilation as occurred in Sudbury, Ontario (e.g.
439 Therriault et al., 2002), later hydrothermal sulfide precipitation with or without serpentinization,
440 as is recorded in regolith breccia martian meteorites (Lorand et al., 2015), or differentiated
441 intrusions (Fig. 3; Section 4.3). Once formed, fluid fracture networks within such sulfide-rich
442 regions in the subsurface of Mars could support high concentrations of radiolytically-fueled
443 sulfate-reducing microorganisms and would be key locations to investigate the possibility of
444 extant life on Mars.

445

446 *4.2 Evidence for concentrated sulfides on Mars*

447 The conclusions from this study do not require concentrated sulfides on Mars, but rather
448 are based on the large body of data for disseminated sulfides on Mars. As such, the estimates are
449 appropriately conservative and are consistent with the body of lithological and geochemical data
450 available. The presence of concentrated sulfide deposits on Mars can be considered, however,
451 and supporting evidence includes (1) sulfur assimilation and sulfide saturation in nakhlite melts
452 (Franz et al., 2014; Mari et al., 2019), (2) geologic evidence for high sulfide concentrations
453 elsewhere in the martian crust based on expected mantle oxygen (Stanley et al., 2011; Armstrong
454 et al., 2015) and sulfur (Ding et al., 2014) fugacities, (3) surface evidence for sulfide oxidation
455 (Zolotov and Shock, 2005; Dehouck et al., 2012; Vaniman et al., 2014), and (4) orbital
456 detections of past serpentinization (Ehlmann et al., 2010; Leask et al., 2018), which produces
457 reducing fluids and is often associated with generation of hydrothermal sulfide deposits on Earth

458 (e.g. Economou and Naldrett, 1984; Thalhammer et al., 1986; Shiga, 1987; Auclair et al., 1993;
459 Wafik et al., 2001; Marques et al., 2007).

460 1. Nakhrites likely crystallized from a sulfide saturated melt after S assimilation from
461 the crust/regolith based on $\Delta^{33}\text{S}$, $\delta^{34}\text{S}$, and highly siderophile element (HSE) patterns
462 (Mari et al., 2019). Some amount of sulfur assimilation from S-rich regolith occurred
463 after the nakhlite lavas were emplaced on or near the martian surface (Franz et al.,
464 2014; Mari et al., 2019). Nakhrites are the only SNC meteorite class known to likely
465 have crystallized from a sulfide-saturated melt, as shergottites crystallized from
466 sulfide undersaturated parent magmas (Wang and Becker, 2017). Still, 1/3 of volcanic
467 martian meteorite classes exhibiting geochemical characteristics consistent with
468 crystallization from a sulfide saturated melt implies that characteristically similar
469 volcanism could have occurred throughout Mars' history. Sulfates are prevalent
470 across the surface of Mars (Ehlmann and Edwards, 2014), S-rich regolith is
471 effectively ubiquitous across the surface (Clark et al., 1976; Foley et al., 2003), and
472 elemental sulfur concentrations measured by GRS range from 0.7-3.2 wt. % at GRS
473 spatial resolution (McLennan et al., 2010), thus a substantial amount of sulfur for
474 accumulation by lavas commonly exists on the martian surface. Therefore, it is likely
475 that lavas saturated with respect to sulfide after assimilation of S from the
476 crust/regolith—similar to those from which nakhlites crystallized—were common
477 throughout Mars' history, thus resulting in the formation of high sulfide concentration
478 zones.

479 2. The martian mantle is generally expected to be more reducing than Earth's (Stanley et
480 al., 2011; Armstrong et al., 2015), with estimates for mantle oxygen fugacity based on

481 basalts measured in Gusev crater varying from iron-wüstite (IW) to IW+1 (Stanley et
482 al., 2011). Oxygen fugacity estimates from shergottites are estimated to be [QFM-4]-
483 [QFM-1] (Herd, 2003), and from nakhlites are ~QFM (Mari et al., 2019). Earth's
484 mantle has an oxygen fugacity range of [QFM-1]-[QFM+1] (Blundy et al., 1991;
485 Brounce et al., 2017). Still, on Earth, pyrrhotite is found in volcanic rocks derived
486 from partial melting of a more oxidized mantle relative to Mars (e.g. Desborough et
487 al., 1968; Whitney, 1984), though concentrations are typically higher in silicic
488 magmas than basalt, the latter of which is more common on Mars. Thus, magmatic
489 sulfides are expected in some volcanic rocks derived from the wide variability in melt
490 oxygen fugacities from the martian mantle. Martian mantle sulfur abundance
491 estimates range from ~400-2200 ppm (Wang and Becker, 2017), while Earth's mantle
492 sulfur abundance is ~250 ppm (McDonough and Sun, 1995). Additional sulfur is
493 incorporated into martian melts via crustal assimilation of sulfates and/or S-rich
494 regolith, as occurred in the nakhlite lavas (Franz et al., 2014; Mari et al., 2019). As
495 such, sulfur activity in martian melts is typically higher than in most melts on Earth,
496 while oxygen fugacity is lower than in most melts on Earth, both of which increase
497 the likelihood of sulfide precipitation.

498 3. Sulfide oxidation has been invoked to explain multiple mineral assemblages found on
499 the martian surface (King and McSween, 2005), including the hematite enrichment in
500 Meridiani Planum (Zolotov and Shock, 2005), the Burns formation –a sulfate-rich
501 sandstone in Meridiani Planum (Zolotov and Shock, 2005), sulfate-carbonate
502 assemblages found using orbital hyperspectral data (Dehouck et al., 2012), akaganeite
503 detections made from orbit (Carter et al., 2015), and acidic fluid diagenetic mineral

504 assemblages detected by the Curiosity rover (Rampe et al., 2017). In its type locality
505 on Earth, akaganeite forms as an alteration product of pyrrhotite (Nambu, 1968). To
506 concentrate the abundances of sulfur needed to form the Burns formation, pre-
507 concentration of sulfur via magmatic or hydrothermal sulfides may be required
508 (Zolotov and Shock, 2005). Furthermore, oxidation of sulfides to form the acid-mine-
509 drainage-like mineral assemblages of sulfates and oxides seen on the martian surface
510 provides a functional hypothesis for generation of these assemblages. Formation of
511 concentrated sulfides could have occurred on Mars via differentiation/crustal
512 assimilation from ultramafic lavas (Burns and Fisher, 1990), the existence of which is
513 supported by possible observations of ultramafic volcanism on the martian surface
514 (Ruff et al., 2014; Ruff et al., 2019; Kremer et al., 2019), ultramafic lithologies in
515 martian meteorites (Burns and Fisher, 1990), and evidence that nakhlites crystallized
516 from sulfide-saturated melts after S assimilation from the crust/regolith (Franz et al.,
517 2014; Mari et al., 2019).

518 4. Mg-rich serpentine has been detected on Mars using CRISM hyperspectral images
519 (Ehlmann et al., 2010; Leask et al., 2018), but most of these detections have been
520 called into question after discovery of a 2.1 μm spectral artifact in CRISM TRR3 data
521 that can resemble the diagnostic absorption feature of Mg-rich serpentine (Leask et
522 al., 2018). Still, at least 3 Mg-rich serpentine detections have been validated as
523 unrelated to this artifact (Leask et al., 2018), demonstrating that serpentinization has
524 occurred on Mars. The fluids generated via serpentinization are highly reducing,
525 which can cause precipitation of reduced alloys and minerals such as awaruite
526 (Lorand, 1985; Sleep et al., 2004) (NiFe alloy) and sulfides (Economou and Naldrett,

527 1984; Thalhammer et al., 1986; Shiga, 1987; Auclair et al., 1993; Wafik et al., 2001;
528 Marques et al., 2007). As such, serpentinization would have generated reducing fluids
529 on Mars that could have precipitated and concentrated sulfide minerals, making
530 regions containing serpentine potential ‘hotzones’ for both ancient chemolithotrophic
531 life that consumed H₂ from serpentinization, as well as modern chemolithotrophic life
532 that may consume H₂ and SO₄ from radiolysis in sulfide-rich zones.

533

534 *4.3 Possible settings of concentrated sulfides on Mars*

535 1. *Differentiated impact melt sheets:* The Sudbury Impact Structure in Ontario, Canada
536 is one of the three largest discovered impact structures on Earth—the others being
537 Chicxulub and Vredefort—and hosts concentrations of sulfides of high economic
538 value. Sulfides were concentrated via gravitational differentiation from the impact
539 melt sheet (Zieg and Marsh, 2005). Sulfur was likely introduced to the impact melt
540 via assimilation from the surrounding country rock during the depressurization
541 melting that formed the Sudbury Igneous Complex (Walker et al., 1991; Dickin et al.,
542 1992; Mungall et al., 2004). Because of the prevalence of sulfur on Mars in the form
543 of sulfates and S-rich regolith, and the larger number of preserved impact structures
544 on Mars relative to Earth, Sudbury-type differentiated impact melt sheet sulfide
545 concentrations may be more common on Mars than they are on Earth (West and
546 Clarke, 2010) (Fig. 3 Setting 1).

547 2. *Layered intrusions:* Sulfides are commonly concentrated in ultramafic-to-mafic
548 layered intrusions on Earth, such as the Bushveld Igneous Complex in South Africa
549 (Gain and Mostert, 1982), Voisey’s Bay in Canada (Evans-Lamswood et al., 2000),

550 Jinchuan in China (Chai and Naldrett, 1992), and the Noril'sk-Talnakh deposits in
551 Siberia (Arndt et al., 2003). Sulfides are typically concentrated as residue in restricted
552 conduits/channels, and thus are often concentrated at the base of the layered intrusion,
553 in sills, or in stratiform reef-style deposits (Lightfoot and Evans-Lamswood, 2015).
554 Such intrusions typically form in intracratonic settings, often via mantle plume
555 activity (Barnes et al., 2017). This is similar to settings of non-subvolcanic and
556 subvolcanic intrusions formed on Mars, which is a single-plate planet. There is
557 evidence for ultramafic volcanism on Mars (Ruff et al., 2019; Kremer et al., 2019)
558 and basaltic volcanism is prevalent across the entire planet (Bandfield et al., 2000).
559 Mantle plumes have caused volcanism on Mars, likely forming the Tharsis and
560 Elysium volcanic bulges (Fuller and Head, 2003; Redmond and King, 2004; Hynek et
561 al., 2011). It is thus plausible that both subvolcanic and non-subvolcanic ultramafic-
562 to-mafic magmatic intrusions exist on Mars, providing structures for potential
563 concentration of sulfides (Fig. 3 Settings 2 & 7).

564 3. *Hydrothermal sulfides.* Sulfides are concentrated by hydrothermal fluids that are S-
565 rich and reducing. Many hydrothermally-formed sulfide deposits are associated with
566 serpentinites, due to the highly reducing fluids formed during the process of
567 serpentinization (Lorand, 1985; Sleep et al., 2004) causing formation of metal alloys
568 (Lorand, 1985) and sulfides (Economou and Naldrett, 1984; Thalhammer et al., 1986;
569 Shiga, 1987; Auclair et al., 1993; Wafik et al., 2001; Marques et al., 2007).

570 Hydrothermal sulfides are found in regolith breccia martian meteorites (Lorand et al.,
571 2015; Wittmann et al., 2015; Hewins et al., 2017; Jean-Pierre Lorand et al., 2018a).
572 Minerals potentially produced by hydrothermal alteration, including smectites,

573 carbonates, chlorite, hydrated silica, and zeolites (Ehlmann et al., 2009), and small
574 amounts of Mg-serpentine (Ehlmann et al., 2010; Leask et al., 2018) are detected on
575 the martian surface from orbit using CRISM. Evidence for past hydrothermal
576 alteration has been seen by every Mars rover. NASA’s Opportunity rover observed
577 evidence for multiple diagenetic episodes in the Burns Formation (McLennan et al.,
578 2005), as well as hydrothermal alteration of bedrock due to meteorite impact at
579 Endeavour crater (Squyres et al., 2012; Arvidson et al., 2014). NASA’s Spirit rover
580 observed carbonate-bearing olivine-rich rocks (Comanche) formed via alteration of
581 ultramafic tuff (Algonquin) (Morris et al., 2010; Ruff et al., 2014), silica deposits
582 (Ruff et al., 2011) that likely formed in hot spring settings (Ruff and Farmer, 2016;
583 Ruff et al., 2019), and hydrothermal alteration of Home Plate resulting in
584 precipitation of silica (Squyres et al., 2008). NASA’s Curiosity rover has found
585 evidence for multiple diagenetic episodes in the stratigraphy of Mount Sharp,
586 including cross-cutting Ca-sulfate veins (Yen et al., 2017), jarosite veins dated to 2.1
587 ± 4 Gyr (Martin et al., 2017), and evidence for diagenetic episodes that cross-cut
588 multiple stratigraphically distinct units (Sheppard et al., 2019). Sulfides could also be
589 concentrated via hydrothermal activity during submarine volcanism –as likely
590 occurred in Eridania Basin (Michalski et al., 2017). As such, hydrothermal alteration
591 has occurred on Mars in a wide variety of geochemical settings, including some that
592 precipitated pyrite in regolith breccias (Lorand et al., 2015; Wittmann et al., 2015;
593 Hewins et al., 2017; Jean-Pierre Lorand et al., 2018a), and some that formed
594 serpentinite (Ehlmann et al., 2010; Leask et al., 2018). It is plausible that higher
595 concentrations of sulfides than those found in NWA 7533 and NWA 7034— which

596 were ejected from Mars by the same impact event (Wittmann et al., 2015)—could
597 form from hydrothermal alteration in other regions of the martian crust (Fig. 3
598 Settings 3 & 5).

599 4. *Sulfide-bearing sediments.* Mount Sharp in Gale crater is an example of a sulfide-
600 bearing sediment mound on Mars (McAdam et al., 2014; Franz et al., 2017; Wong et
601 al., 2020) (Fig. 3 Setting 4). These sulfides are diagenetic/epigenetic, likely resulting
602 from thermal reduction of preexisting sulfates (Franz et al., 2017). It is possible that a
603 similar sulfate thermal reduction process could occur in other sulfate-bearing martian
604 sediments, which are widespread (Ehlmann and Edwards, 2014), thus generating a
605 higher concentration of sulfides in those localities. Cu-enrichments in Gale crater are
606 potentially due to presence of detrital sulfides (Payré et al., 2019). It is therefore
607 plausible that sulfides could be concentrated in martian sediments in detrital form and
608 also via diagenetic processes.

609 5. *Differentiated ultramafic-to-mafic lava flows.* Examples of sulfide concentrations in
610 komatiitic lava flows on Earth include the Kambalda and Perseverance deposits in
611 Australia. Sulfides are also concentrated picrites, including the Pechenga deposits in
612 Russia (Barnes et al., 2017). Due to the presence of ultramafic lithologies in martian
613 meteorites, the possibility of sulfide concentration in komatiite-type settings has been
614 known for decades (Burns and Fisher, 1990). Since then, further evidence for
615 ultramafic volcanism on Mars has been presented (Ruff et al., 2014; Ruff et al., 2019;
616 Kremer et al., 2019). The magmas that precipitated the nakhlites assimilated sulfur
617 from the crust, forming a sulfide-saturated melt (Franz et al., 2014; Mari et al., 2019).
618 Thus, there is already evidence for some degree of sulfide concentration in

619 differentiated ultramafic-to-mafic lava flows on Mars via the martian meteorites (Fig.
620 3 Setting 6).

621

622 **5.0 Conclusions**

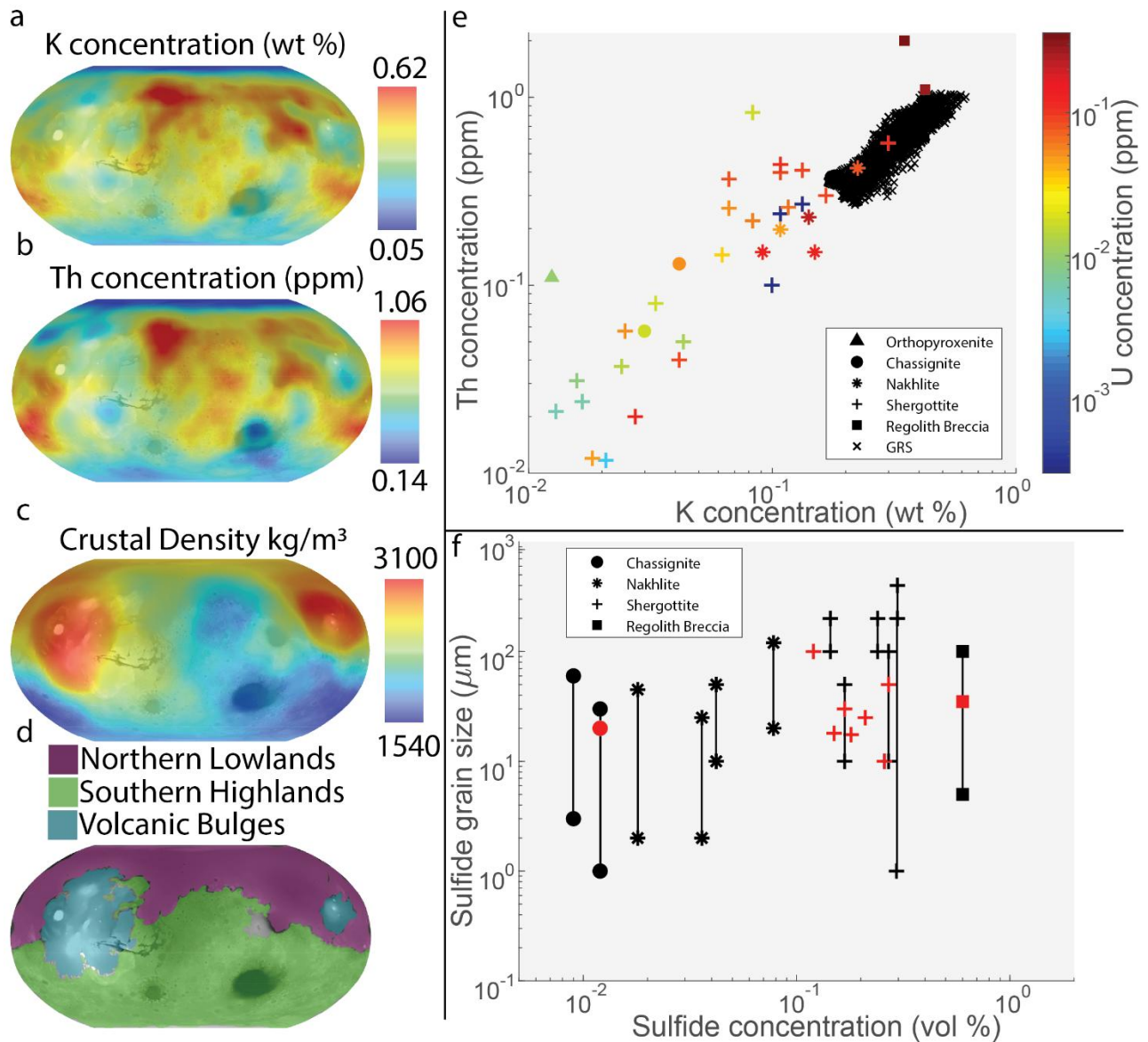
623 The results presented here demonstrate that radiolysis in the martian meteorite source
624 lithologies would produce sufficient H₂ and sulfate to sustain sulfate-reducing bacteria wherever
625 groundwater is present. The regolith breccia source lithology, which is within the southern
626 highlands (Cassata et al., 2018), could sustain the highest concentrations of sulfate-reducing
627 bacteria, with shergottite and nakhlite lithologies supporting the second highest concentrations.
628 The calculated supportable sulfate-reducing bacteria densities in the regolith breccia, shergottite,
629 and nakhlite source lithologies are comparable to those measured in Earth's deep subsurface
630 (Table 3; Fig. 2).

631 If Earth-like chemolithotrophic life ever existed on Mars and survived until present, it could
632 potentially be sustained in these subsurface regions by harnessing the same radiolytically-derived
633 energy that drives the metabolisms of microbial ecosystems on Earth sustained in groundwaters
634 >10⁶-10⁹ years old (Lin et al., 2006; Chivian et al., 2008; Lollar et al., 2019). Furthermore, this
635 subsurface environment has likely remained habitable since the Noachian (Tarnas et al., 2018),
636 making it the longest-lived habitable environment on Mars and the most likely refugia for any
637 life on the planet (National Academies of Sciences, Engineering, and Medicine et al., 2018;
638 Michalski et al., 2018). The parameters outlined here—radionuclide abundance, sulfide
639 abundance, average sulfide grain size, and porosity—along with characterization of the locations
640 of liquid groundwater, can be used to constrain an optimal landing site for a mission to detect
641 Earth-like extant sulfate-reducing martian life in the subsurface. Additional microbial

642 metabolisms, for which redox energy availability is more difficult to model, could also exist in
 643 the martian subsurface, as they do in Earth's subsurface, potentially increasing the measured
 644 biomass in any subsurface region that contains groundwater. This should be considered in the
 645 context of future extant life detection missions to Mars.

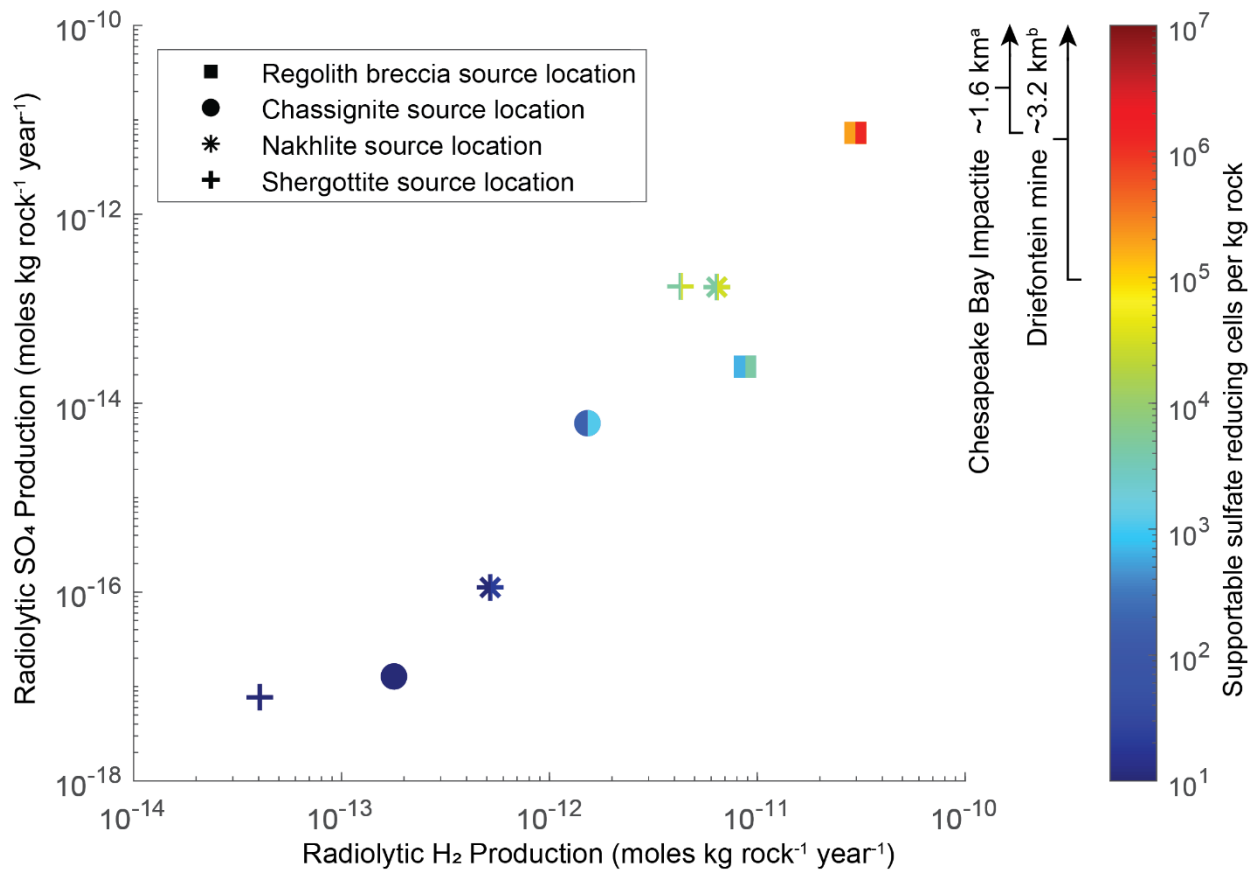
646

647 **Figures**



648

649 **Figure 1 | Model input data.** (a) K concentration (wt. %) from GRS data, corrected for
650 contributions Cl, H, and S (Boynton et al., 2007; Tarnas et al., 2018). (b) Th concentration (ppm)
651 from GRS data, corrected for contributions Cl, H, and S (Boynton et al., 2007; Tarnas et al.,
652 2018). (c) Possible crustal density estimates from orbital gravimetry (Goossens et al., 2017) that
653 are supported by rover gravimetry (Lewis et al., 2019). (d) Division of Mars into 3 different
654 regions. (e) Radionuclide abundances in martian meteorites (Meyer, 2016) and GRS data from
655 60°S-60°N, where near surface ice does not contaminate measurements (Boynton et al., 2007;
656 Tarnas et al., 2018). The lower spatial resolution of GRS data (100s of km) means the calculated
657 K and Th concentrations are more closely clustered than in the martian meteorites. There are also
658 no U concentration measurements from GRS data, thus the color is black. (f) Sulfide
659 concentrations and grain sizes in martian meteorites are compiled and referenced in Table 1.
660 Black symbols show the maximum and minimum reported grain sizes, while red symbols show
661 the reported average grain size.
662



663

664 **Figure 2 | Radiolytic redox energy production and number of supportable sulfate-reducing**

665 **cells in Mars meteorite source regions.** The datapoints represent the highest and lowest

666 endmembers for each given category, with the assumed values for the calculations presented in

667 Table 2. The different colors of the same datapoints represent the supportable sulfate-reducing

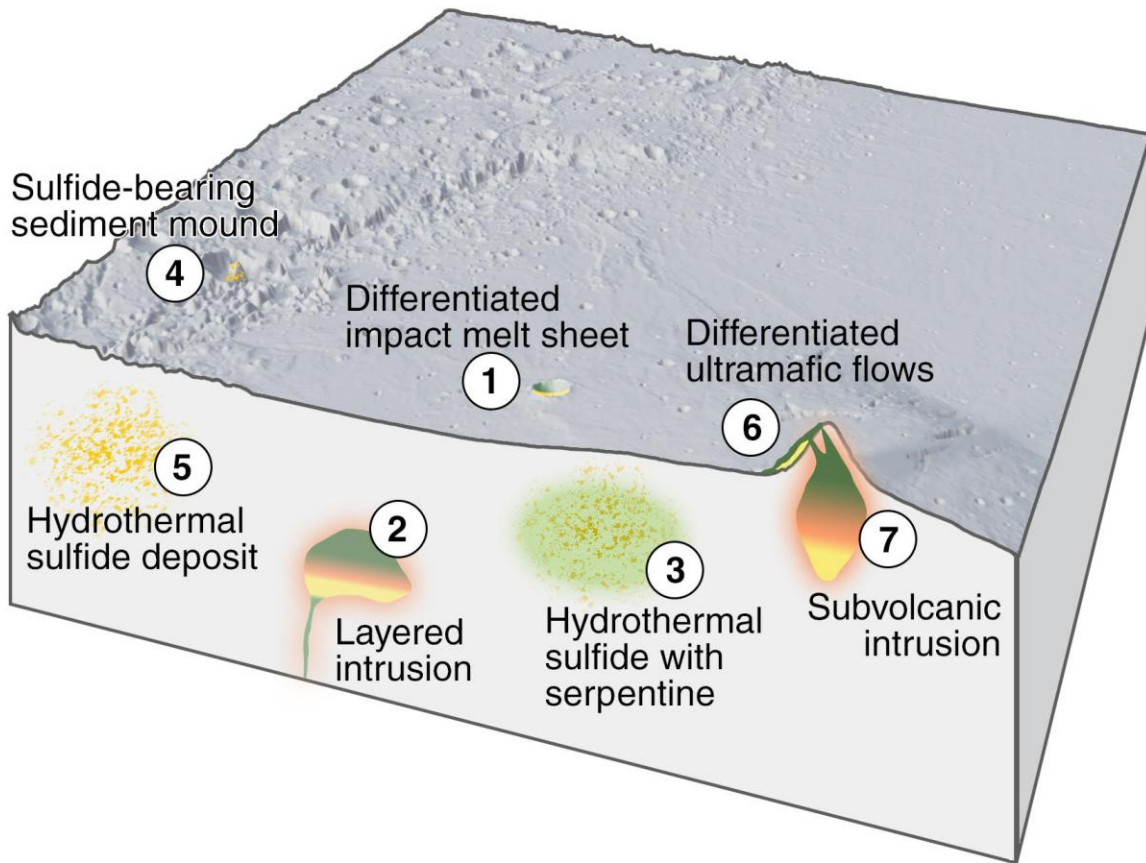
668 cells per kg rock assuming the highest and lowest metabolic rates observed in Earth's deep

669 subsurface ($5.5 \times 10^{-18} - 3.6 \times 10^{-17}$ moles cell⁻¹ year⁻¹) (Lin et al., 2006; Chivian et al., 2008).

670 Cell density measurements from Earth's deep subsurface come from ^a(Cockell et al., 2012),

671 ^b(Onstott et al., 2003).

672



673

674 **Figure 3 | Possible sulfide concentration settings on Mars in addition to those found in**
 675 **martian meteorites.** Topographic cross-section of Mars using data from the Mars Orbital Laser
 676 Altimeter (MOLA) from 10°S, 141°E; 29°N, 149°E; 4°S, 104°E; 35°N, 96°E. See Materials and
 677 Methods for discussion of possible sulfide concentration settings on Mars, in addition to sulfide
 678 concentrations found in martian meteorites that are used for calculations in this study. (1)
 679 Differentiated impact melt sheet similar to sulfide concentrations at the Sudbury Impact
 680 Structure in Canada (Therriault et al., 2002). (2) Sulfide concentration in layered intrusion,
 681 similar to Bushveld Igneous Complex in South Africa and many other localities on Earth. (3)
 682 Sulfide hydrothermally concentrated in serpentinite, similar to many ophiolite localities on Earth.
 683 Mg-serpentine has been detected on Mars (Leask et al., 2018). (4) Sulfide concentrations in

684 sediments, such as those in Gale crater (Franz et al., 2017; Wong et al., 2020). (5) Sulfide
685 hydrothermally concentrated without associated serpentinite minerals, such as those in regolith
686 breccia martian meteorites (Lorand et al., 2005; Lorand et al., 2015; Jean-Pierre Lorand et al.,
687 2018b; Jean-Pierre Lorand et al., 2018a). (6) Concentrations of sulfides in differentiated
688 ultramafic-to-mafic lava flows, as occurs in many localities on Earth including the Kambalda and
689 Perseverance deposits in Australia. Sulfides are also concentrated in picrites, including the
690 Pechenga deposits in Russia. This is a possible formation setting for nakhlites, which crystallized
691 from sulfide saturated magmas after assimilation of S from the crust/regolith (Franz et al., 2014;
692 Mari et al., 2019). (7) Concentration of sulfides in ultramafic-to-mafic subvolcanic intrusions
693 beneath volcanic provinces such as Tharsis, Elysium, and Syrtis Major.

694

695 **Acknowledgements**

696 We thank Norman Sleep and an anonymous reviewer for feedback that improved the quality of
697 this manuscript. We thank Paul Niles, Ralph Milliken, and Steven D'Hondt for discussions.
698 Thanks to the Astromaterial Curation Group at NASA JSC, especially Kevin Righter, for
699 providing a compilation table of martian meteorite elemental abundances. We thank Paul Niles,
700 Ralph Milliken, and Steven D'Hondt for discussions. J.D.T. gratefully acknowledges support
701 from the NASA Postdoctoral Program and a Brown University Dissertation Fellowship. J.F.M.,
702 B.S.L., V.S., and J.R.M. acknowledge support from the Canadian Institute for Advanced
703 Research (CIFAR).

704

705 **References**

706 Adamiec G. and Aitken M. (1998) Dose-rate conversion factors: Update: Ancient TL, v. 16.
707 *ArcheoSciences, revue d'archéométrie* **32**, 51–58.

- 708 Altair T., de Avellar M. G. B., Rodrigues F. and Galante D. (2018) Microbial habitability of Europa
709 sustained by radioactive sources. *Scientific Reports* **8**, 1–8.
- 710 Armstrong L. S., Hirschmann M. M., Stanley B. D., Falksen E. G. and Jacobsen S. D. (2015)
711 Speciation and solubility of reduced C–O–H–N volatiles in mafic melt: Implications for
712 volcanism, atmospheric evolution, and deep volatile cycles in the terrestrial planets.
713 *Geochimica et Cosmochimica Acta* **171**, 283–302.
- 714 Arndt N. T., Czamanske G. K., Walker R. J., Chauvel C. and Fedorenko V. A. (2003) Geochemistry
715 and Origin of the Intrusive Hosts of the Noril’sk-Talnakh Cu-Ni-PGE Sulfide Deposits.
716 *Economic Geology* **98**, 495–515.
- 717 Arvidson R. E., Squyres S. W., Bell J. F., Catalano J. G., Clark B. C., Crumpler L. S., Souza P. A. de,
718 Fairén A. G., Farrand W. H., Fox V. K., Gellert R., Ghosh A., Golombek M. P., Grotzinger J.
719 P., Guinness E. A., Herkenhoff K. E., Jolliff B. L., Knoll A. H., Li R., McLennan S. M., Ming
720 D. W., Mittlefehldt D. W., Moore J. M., Morris R. V., Murchie S. L., Parker T. J., Paulsen
721 G., Rice J. W., Ruff S. W., Smith M. D. and Wolff M. J. (2014) Ancient Aqueous
722 Environments at Endeavour Crater, Mars. *Science* **343**. Available at:
723 <https://science.sciencemag.org/content/343/6169/1248097> [Accessed July 9, 2020].
- 724 Auclair M., Gauthier M., Trottier J., Jebrak M. and Chartrand F. (1993) Mineralogy,
725 geochemistry, and paragenesis of the Eastern Metals serpentinite-associated Ni-Cu-Zn
726 deposit, Quebec Appalachians. *Economic Geology* **88**, 123–138.
- 727 Bandfield J. L., Hamilton V. E. and Christensen P. R. (2000) A Global View of Martian Surface
728 Compositions from MGS-TES. *Science* **287**, 1626–1630.
- 729 Barnes S. J., Holwell D. A. and Vaillant M. L. (2017) Magmatic Sulfide Ore Deposits. *Elements* **13**,
730 89–95.
- 731 Baumgartner R. J., Fiorentini M. L., Baratoux D., Micklethwaite S., Sener A. K., Lorand J. P. and
732 McCuaig T. C. (2015) Magmatic controls on the genesis of Ni–Cu±(PGE) sulphide
733 mineralisation on Mars. *Ore Geology Reviews* **65**, 400–412.
- 734 Baumgartner R. J., Fiorentini M. L., Lorand J.-P., Baratoux D., Zaccarini F., Ferrière L., Prašek M.
735 K. and Sener K. (2017) The role of sulfides in the fractionation of highly siderophile and
736 chalcophile elements during the formation of martian shergottite meteorites.
737 *Geochimica et Cosmochimica Acta* **210**, 1–24.
- 738 Blundy J. D., Brodholt J. P. and Wood B. J. (1991) Carbon–fluid equilibria and the oxidation state
739 of the upper mantle. *Nature* **349**, 321–324.
- 740 Boynton W. V., Taylor G. J., Evans L. G., Reedy R. C., Starr R., Janes D. M., Kerry K. E., Drake D.
741 M., Kim K. J., Williams R. M. S., Crombie M. K., Dohm J. M., Baker V., Metzger A. E.,
742 Karunatillake S., Keller J. M., Newsom H. E., Arnold J. R., Brückner J., Englert P. a. J.,
743 Gasnault O., Sprague A. L., Mitrofanov I., Squyres S. W., Trombka J. I., d’Uston L., Wänke

- 744 H. and Hamara D. K. (2007) Concentration of H, Si, Cl, K, Fe, and Th in the low- and mid-
745 latitude regions of Mars. *Journal of Geophysical Research: Planets* **112**. Available at:
746 <https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/2007JE002887> [Accessed
747 January 15, 2020].
- 748 Bragg M. A. and Kleeman B. Sc. (1905) XXXIX. On the α particles of radium, and their loss of
749 range in passing through various atoms and molecules. *The London, Edinburgh, and*
750 *Dublin Philosophical Magazine and Journal of Science* **10**, 318–340.
- 751 Broquet A. and Wieczorek M. A. (2019) The Gravitational Signature of Martian Volcanoes.
752 *Journal of Geophysical Research: Planets* **124**, 2054–2086.
- 753 Brounce M., Stolper E. and Eiler J. (2017) Redox variations in Mauna Kea lavas, the oxygen
754 fugacity of the Hawaiian plume, and the role of volcanic gases in Earth's oxygenation.
755 *PNAS*. Available at: <https://www.pnas.org/content/early/2017/08/01/1619527114>
756 [Accessed January 21, 2020].
- 757 Buck E. C., Wittman R. S., Skomurski F. N., Cantrell K. J., McNamara B. K. and Soderquist C. Z.
758 (2012) *Radiolysis Process Model*, Pacific Northwest National Lab. (PNNL), Richland, WA
759 (United States). Available at: <https://www.osti.gov/biblio/1069211> [Accessed January
760 17, 2020].
- 761 Burns R. G. and Fisher D. S. (1990) Evolution of sulfide mineralization on Mars. *Journal of*
762 *Geophysical Research: Solid Earth*, 14169–14173.
- 763 Carter J., Viviano-Beck C., Loizeau D., Bishop J. and Le Deit L. (2015) Orbital detection and
764 implications of akaganéite on Mars. *Icarus* **253**, 296–310.
- 765 Cassata W. S., Cohen B. E., Mark D. F., Trappitsch R., Crow C. A., Wimpenny J., Lee M. R. and
766 Smith C. L. (2018) Chronology of martian breccia NWA 7034 and the formation of the
767 martian crustal dichotomy. *Science Advances* **4**, eaap8306.
- 768 Chai G. and Naldrett A. J. (1992) Characteristics of Ni-Cu-PGE mineralization and genesis of the
769 Jinchuan Deposit, Northwest China. *Economic Geology* **87**, 1475–1495.
- 770 Chevrier V., Lorand J.-P. and Sautter V. (2011) Sulfide petrology of four nakhlites: Northwest
771 Africa 817, Northwest Africa 998, Nakhla, and Governador Valadares. *Meteoritics &*
772 *Planetary Science* **46**, 769–784.
- 773 Chivian D., Brodie E. L., Alm E. J., Culley D. E., Dehal P. S., DeSantis T. Z., Gihring T. M., Lapidus
774 A., Lin L.-H., Lowry S. R., Moser D. P., Richardson P. M., Southam G., Wanger G., Pratt L.
775 M., Andersen G. L., Hazen T. C., Brockman F. J., Arkin A. P. and Onstott T. C. (2008)
776 Environmental Genomics Reveals a Single-Species Ecosystem Deep Within Earth. *Science*
777 **322**, 275–278.

- 778 Clark B. C., Baird A. K., Rose H. J., Toulmin P., Keil K., Castro A. J., Kelliher W. C., Rowe C. D. and
779 Evans P. H. (1976) Inorganic Analyses of Martian Surface Samples at the Viking Landing
780 Sites. *Science* **194**, 1283–1288.
- 781 Cockell C. S., Voytek M. A., Gronstal A. L., Finster K., Kirshtein J. D., Howard K., Reitner J., Gohn
782 G. S., Sanford W. E., Horton J. W., Kallmeyer J., Kelly L. and Powars D. S. (2012) Impact
783 Disruption and Recovery of the Deep Subsurface Biosphere. *Astrobiology* **12**, 231–246.
- 784 Dehouck E., Chevrier V., Gaudin A., Mangold N., Mathé P.-E. and Rochette P. (2012) Evaluating
785 the role of sulfide-weathering in the formation of sulfates or carbonates on Mars.
786 *Geochimica et Cosmochimica Acta* **90**, 47–63.
- 787 Desborough G. A., Anderson A. T. and Wright T. L. (1968) Mineralogy of sulfides from certain
788 Hawaiian basalts. *Economic Geology* **63**, 636–644.
- 789 Dickin A. P., Richardson J. M., Crocket J. H., McNutt R. H. and Peredery W. V. (1992) Osmium
790 isotope evidence for a crustal origin of platinum group elements in the Sudbury nickel
791 ore, Ontario, Canada. *Geochimica et Cosmochimica Acta* **56**, 3531–3537.
- 792 Ding S., Dasgupta R. and Tsuno K. (2014) Sulfur concentration of martian basalts at sulfide
793 saturation at high pressures and temperatures – Implications for deep sulfur cycle on
794 Mars. *Geochimica et Cosmochimica Acta* **131**, 227–246.
- 795 Dzaugis M., Spivack A. J. and D’Hondt S. (2018) Radiolytic H₂ Production in Martian
796 Environments. *Astrobiology* **18**, 1137–1146.
- 797 Economou M. I. and Naldrett A. J. (1984) Sulfides associated with podiform bodies of chromite
798 at Tsangli, Eretria, Greece. *Mineral. Deposita* **19**, 289–297.
- 799 Ehlmann B. L. and Edwards C. S. (2014) Mineralogy of the Martian Surface. *Annual Review of*
800 *Earth and Planetary Sciences* **42**, 291–315.
- 801 Ehlmann B. L., Mustard J. F. and Murchie S. L. (2010) Geologic setting of serpentine deposits on
802 Mars. *Geophysical Research Letters* **37**. Available at:
803 <https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/2010GL042596> [Accessed
804 January 21, 2020].
- 805 Ehlmann B. L., Mustard J. F., Swayze G. A., Clark R. N., Bishop J. L., Poulet F., Marais D. J. D.,
806 Roach L. H., Milliken R. E., Wray J. J., Barnouin-Jha O. and Murchie S. L. (2009)
807 Identification of hydrated silicate minerals on Mars using MRO-CRISM: Geologic context
808 near Nili Fossae and implications for aqueous alteration. *Journal of Geophysical*
809 *Research: Planets* **114**. Available at:
810 <https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/2009JE003339> [Accessed
811 April 8, 2019].

- 812 Evans-Lamswood D. M., Butt D. P., Jackson R. S., Lee D. V., Muggridge M. G., Wheeler R. I. and
813 Wilton D. H. C. (2000) Physical Controls Associated with the Distribution of Sulfides in
814 the Voisey's Bay Ni-Cu-Co Deposit, Labrador. *Economic Geology* **95**, 749–769.
- 815 Foley C. N., Economou T. and Clayton R. N. (2003) Final chemical results from the Mars
816 Pathfinder alpha proton X-ray spectrometer. *Journal of Geophysical Research: Planets*
817 **108**. Available at:
818 <https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/2002JE002019> [Accessed July
819 9, 2020].
- 820 Franz H. B., Kim S.-T., Farquhar J., Day J. M. D., Economos R. C., McKeegan K. D., Schmitt A. K.,
821 Irving A. J., Hoek J. and Iii J. D. (2014) Isotopic links between atmospheric chemistry and
822 the deep sulphur cycle on Mars. *Nature* **508**, 364–368.
- 823 Franz H. B., McAdam A. C., Ming D. W., Freissinet C., Mahaffy P. R., Eldridge D. L., Fischer W. W.,
824 Grotzinger J. P., House C. H., Hurowitz J. A., McLennan S. M., Schwenzer S. P., Vaniman
825 D. T., Archer Jr P. D., Atreya S. K., Conrad P. G., Drottin Iii J. W., Eigenbrode J. L., Farley K.
826 A., Glavin D. P., Johnson S. S., Knudson C. A., Morris R. V., Navarro-González R., Pavlov A.
827 A., Plummer R., Rampe E. B., Stern J. C., Steele A., Summons R. E. and Sutter B. (2017)
828 Large sulfur isotope fractionations in Martian sediments at Gale crater. *Nature*
829 *Geoscience* **10**, 658–662.
- 830 Franz H. B., Wu N., Farquhar J. and Irving A. J. (2019) A new type of isotopic anomaly in
831 shergottite sulfides. *Meteoritics & Planetary Science* **54**, 3036–3051.
- 832 Fuller E. R. and Head J. W. (2003) Olympus Mons, Mars: Detection of extensive preareole
833 volcanism and implications for initial mantle plume behavior. *Geology* **31**, 175–178.
- 834 Gain S. B. and Mostert A. B. (1982) The geological setting of the platinoid and base metal sulfide
835 mineralization in the Platreef of the Bushveld Complex in Drenthe, North of
836 Potgietersrus. *Economic Geology* **77**, 1395–1404.
- 837 Gattacceca J., Hewins R. H., Lorand J.-P., Rochette P., Lacroix F., Cournède C., Uehara M., Pont
838 S., Sautter V., Scorzelli R. B., Hombourger C., Munayco P., Zanda B., Chennaoui H. and
839 Ferrière L. (2013) Opaque minerals, magnetic properties, and paleomagnetism of the
840 Tissint Martian meteorite. *Meteoritics & Planetary Science* **48**, 1919–1936.
- 841 Gellert R., Rieder R., Anderson R. C., Brückner J., Clark B. C., Dreibus G., Economou T.,
842 Klingelhöfer G., Lugmair G. W., Ming D. W., Squyres S. W., d'Uston C., Wänke H., Yen A.
843 and Zipfel J. (2004) Chemistry of Rocks and Soils in Gusev Crater from the Alpha Particle
844 X-ray Spectrometer. *Science* **305**, 829–832.
- 845 Goossens S., Sabaka T. J., Genova A., Mazarico E., Nicholas J. B. and Neumann G. A. (2017)
846 Evidence for a low bulk crustal density for Mars from gravity and topography.
847 *Geophysical Research Letters* **44**, 7686–7694.

- 848 Greenwood J. P., Mojzsis S. J. and Coath C. D. (2000a) Sulfur isotopic compositions of individual
849 sulfides in Martian meteorites ALH84001 and Nakhla: implications for crust–regolith
850 exchange on Mars. *Earth and Planetary Science Letters* **184**, 23–35.
- 851 Greenwood J. P., Riciputi L. R., McSween H. Y. and Taylor L. A. (2000b) Modified sulfur isotopic
852 compositions of sulfides in the nakhlites and Chassigny. *Geochimica et Cosmochimica*
853 *Acta* **64**, 1121–1131.
- 854 Grimm R. E., Harrison K. P., Stillman D. E. and Kirchoff M. R. (2017) On the secular retention of
855 ground water and ice on Mars. *Journal of Geophysical Research: Planets* **122**, 94–109.
- 856 Hahn B. C., McLennan S. M. and Klein E. C. (2011) Martian surface heat production and crustal
857 heat flow from Mars Odyssey Gamma-Ray spectrometry. *Geophysical Research Letters*
858 **38**. Available at:
859 <https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/2011GL047435> [Accessed
860 January 21, 2020].
- 861 Herd C. D. K. (2003) The oxygen fugacity of olivine-phyric martian basalts and the components
862 within the mantle and crust of Mars. *Meteoritics & Planetary Science* **38**, 1793–1805.
- 863 Hewins R. H., Humayun M., Barrat J.-A., Zanda B., Lorand J.-P., Pont S., Assayag N., Cartigny P.,
864 Yang S. and Sautter V. (2020) Northwest Africa 8694, a ferroan chassignite: Bridging the
865 gap between nakhlites and chassignites. *Geochimica et Cosmochimica Acta* **282**, 201–
866 226.
- 867 Hewins R. H., Zanda B., Humayun M., Nemchin A., Lorand J.-P., Pont S., Deldicque D., Bellucci J.
868 J., Beck P., Leroux H., Marinova M., Remusat L., Göpel C., Lewin E., Grange M., Kennedy
869 A. and Whitehouse M. J. (2017) Regolith breccia Northwest Africa 7533: Mineralogy and
870 petrology with implications for early Mars. *Meteoritics & Planetary Science* **52**, 89–124.
- 871 Holland G., Lollar B. S., Li L., Lacrampe-Couloume G., Slater G. F. and Ballentine C. J. (2013) Deep
872 fracture fluids isolated in the crust since the Precambrian era. *Nature* **497**, 357–360.
- 873 Humayun M., Yang S., Irving A. J. and Righter K. (2019) Sulfide Assimilation and Mineralization
874 in Ancient (2.4 Ga) Shergottites. **2157**, 6380.
- 875 Hynke B. M., Robbins S. J., Šrámek O. and Zhong S. J. (2011) Geological evidence for a migrating
876 Tharsis plume on early Mars. *Earth and Planetary Science Letters* **310**, 327–333.
- 877 Johnson C. L., Mittelholz A., Langlais B., Russell C. T., Ansan V., Banfield D., Chi P. J., Fillingim M.
878 O., Forget F., Haviland H. F., Golombek M., Joy S., Lognonné P., Liu X., Michaut C., Pan L.,
879 Quantin-Nataf C., Spiga A., Stanley S., Thorne S. N., Wiczorek M. A., Yu Y., Smrekar S. E.
880 and Banerdt W. B. (2020) Crustal and time-varying magnetic fields at the InSight landing
881 site on Mars. *Nature Geoscience* **13**, 199–204.

- 882 King P. L. and McSween H. Y. (2005) Effects of H₂O, pH, and oxidation state on the stability of
883 Fe minerals on Mars. *Journal of Geophysical Research: Planets* **110**. Available at:
884 <https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/2005JE002482> [Accessed July
885 9, 2020].
- 886 Klein F., Tarnas J. D. and Bach W. (2020) Abiotic Sources of Molecular Hydrogen on Earth.
887 *Elements* **16**, 19–24.
- 888 Kremer C. H., Mustard J. F. and Bramble M. S. (2019) A widespread olivine-rich ash deposit on
889 Mars. *Geology* **47**, 677–681.
- 890 Kumagai Y., Kimura A., Taguchi M., Nagaishi R., Yamagishi I. and Kimura T. (2013) Hydrogen
891 production in gamma radiolysis of the mixture of mordenite and seawater. *Journal of*
892 *Nuclear Science and Technology* **50**, 130–138.
- 893 Langlais B., Thébaud E., Houliez A., Purucker M. E. and Lillis R. J. (2019) A New Model of the
894 Crustal Magnetic Field of Mars Using MGS and MAVEN. *Journal of Geophysical Research:*
895 *Planets* **124**, 1542–1569.
- 896 LaVerne J. A. and Tandon L. (2005) H₂ and Cl₂ Production in the Radiolysis of Calcium and
897 Magnesium Chlorides and Hydroxides. *J. Phys. Chem. A* **109**, 2861–2865.
- 898 Le Deit L., Mangold N., Forni O., Cousin A., Lasue J., Schröder S., Wiens R. C., Sumner D., Fabre
899 C., Stack K. M., Anderson R. B., Blaney D., Clegg S., Dromart G., Fisk M., Gasnault O.,
900 Grotzinger J. P., Gupta S., Lanza N., Mouélic S. L., Maurice S., McLennan S. M., Meslin P.-
901 Y., Nachon M., Newsom H., Payré V., Rapin W., Rice M., Sautter V. and Treiman A. H.
902 (2016) The potassic sedimentary rocks in Gale Crater, Mars, as seen by ChemCam on
903 board Curiosity. *Journal of Geophysical Research: Planets* **121**, 784–804.
- 904 Leask E. K., Ehlmann B. L., Dundar M. M., Murchie S. L. and Seelos F. P. (2018) Challenges in the
905 Search for Perchlorate and Other Hydrated Minerals With 2.1- μ m Absorptions on Mars.
906 *Geophysical Research Letters* **45**, 12,180-12,189.
- 907 Lefticariu L., Pratt L. A., LaVerne J. A. and Schimmelmann A. (2010) Anoxic pyrite oxidation by
908 water radiolysis products — A potential source of biosustaining energy. *Earth and*
909 *Planetary Science Letters* **292**, 57–67.
- 910 Lewis K. W., Peters S., Gonter K., Morrison S., Schmerr N., Vasavada A. R. and Gabriel T. (2019)
911 A surface gravity traverse on Mars indicates low bedrock density at Gale crater. *Science*
912 **363**, 535–537.
- 913 Li L., Wing B. A., Bui T. H., McDermott J. M., Slater G. F., Wei S., Lacrampe-Couloume G. and
914 Lollar B. S. (2016) Sulfur mass-independent fractionation in subsurface fracture waters
915 indicates a long-standing sulfur cycle in Precambrian rocks. *Nat Commun* **7**, 1–9.

- 916 Lightfoot P. C. and Evans-Lamswood D. (2015) Structural controls on the primary distribution of
917 mafic–ultramafic intrusions containing Ni–Cu–Co–(PGE) sulfide mineralization in the
918 roots of large igneous provinces. *Ore Geology Reviews* **64**, 354–386.
- 919 Lin L.-H., Hall J., Lippmann-Pipke J., Ward J. A., Lollar B. S., DeFlaun M., Rothmel R., Moser D.,
920 Gihring T. M., Mislouack B. and Onstott T. C. (2005a) Radiolytic H₂ in continental crust:
921 Nuclear power for deep subsurface microbial communities. *Geochemistry, Geophysics,*
922 *Geosystems* **6**. Available at:
923 <https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/2004GC000907> [Accessed
924 January 6, 2020].
- 925 Lin L.-H., Slater G. F., Sherwood Lollar B., Lacrampe-Couloume G. and Onstott T. C. (2005b) The
926 yield and isotopic composition of radiolytic H₂, a potential energy source for the deep
927 subsurface biosphere. *Geochimica et Cosmochimica Acta* **69**, 893–903.
- 928 Lin L.-H., Wang P.-L., Rumble D., Lippmann-Pipke J., Boice E., Pratt L. M., Lollar B. S., Brodie E. L.,
929 Hazen T. C., Andersen G. L., DeSantis T. Z., Moser D. P., Kershaw D. and Onstott T. C.
930 (2006) Long-Term Sustainability of a High-Energy, Low-Diversity Crustal Biome. *Science*
931 **314**, 479–482.
- 932 Lollar G. S., Warr O., Telling J., Osburn M. R. and Lollar B. S. (2019) ‘Follow the Water’:
933 Hydrogeochemical Constraints on Microbial Investigations 2.4 km Below Surface at the
934 Kidd Creek Deep Fluid and Deep Life Observatory. *Geomicrobiology Journal* **36**, 859–872.
- 935 Lorand J.-P. (1985) The behaviour of the upper mantle sulfide component during the incipient
936 alteration of “Alpine”-type peridotites as illustrated by the Beni Bousera (northern
937 Morocco) and Ronda (southern Spain) ultramafic bodies. *TMPM Tschermaks Petr. Mitt.*
938 **34**, 183–209.
- 939 Lorand J.-P., Barrat J.-A., Chevrier V., Sautter V. and Pont S. (2012) Metal-saturated sulfide
940 assemblages in NWA 2737: Evidence for impact-related sulfur devolatilization in Martian
941 meteorites. *Meteoritics & Planetary Science* **47**, 1830–1841.
- 942 Lorand J.-P., Chevrier V. and Sautter V. (2005) Sulfide mineralogy and redox conditions in some
943 shergottites. *Meteoritics & Planetary Science* **40**, 1257–1272.
- 944 Lorand J.-P., Hewins R. H., Humayun M., Remusat L., Zanda B., La C. and Pont S. (2018a)
945 Chalcophile-siderophile element systematics of hydrothermal pyrite from martian
946 regolith breccia NWA 7533. *Geochimica et Cosmochimica Acta* **241**, 134–149.
- 947 Lorand J.-P., Hewins R. H., Remusat L., Zanda B., Pont S., Leroux H., Marinova M., Jacob D.,
948 Humayun M., Nemchin A., Grange M., Kennedy A. and Göpel C. (2015) Nickeliferous
949 pyrite tracks pervasive hydrothermal alteration in Martian regolith breccia: A study in
950 NWA 7533. *Meteoritics & Planetary Science* **50**, 2099–2120.

- 951 Lorand J.-P., Pont S., Chevrier V., Luguët A., Zanda B. and Hewins R. (2018b) Petrogenesis of
952 martian sulfides in the Chassigny meteorite. *American Mineralogist* **103**, 872–885.
- 953 Magnabosco C., Lin L.-H., Dong H., Bomberg M., Ghiorse W., Stan-Lotter H., Pedersen K., Kieft T.
954 L., van Heerden E. and Onstott T. C. (2018) The biomass and biodiversity of the
955 continental subsurface. *Nature Geoscience* **11**, 707–717.
- 956 Mari N., Riches A. J. V., Hallis L. J., Marrocchi Y., Villeneuve J., Gleissner P., Becker H. and Lee M.
957 R. (2019) Syneruptive incorporation of martian surface sulphur in the nakhlite lava flows
958 revealed by S and Os isotopes and highly siderophile elements: implication for mantle
959 sources in Mars. *Geochimica et Cosmochimica Acta* **266**, 416–434.
- 960 Marques A. F. A., Barriga F. J. A. S. and Scott S. D. (2007) Sulfide mineralization in an ultramafic-
961 rock hosted seafloor hydrothermal system: From serpentinization to the formation of
962 Cu–Zn–(Co)-rich massive sulfides. *Marine Geology* **245**, 20–39.
- 963 Martin P. E., Farley K. A., Baker M. B., Malespin C. A., Schwenzer S. P., Cohen B. A., Mahaffy P.
964 R., McAdam A. C., Ming D. W., Vasconcelos P. M. and Navarro-González R. (2017) A Two-
965 Step K-Ar Experiment on Mars: Dating the Diagenetic Formation of Jarosite from
966 Amazonian Groundwaters. *Journal of Geophysical Research: Planets* **122**, 2803–2818.
- 967 McAdam A. C., Franz H. B., Sutter B., Archer P. D., Freissinet C., Eigenbrode J. L., Ming D. W.,
968 Atreya S. K., Bish D. L., Blake D. F., Bower H. E., Brunner A., Buch A., Glavin D. P.,
969 Grotzinger J. P., Mahaffy P. R., McLennan S. M., Morris R. V., Navarro-González R.,
970 Rampe E. B., Squyres S. W., Steele A., Stern J. C., Sumner D. Y. and Wray J. J. (2014)
971 Sulfur-bearing phases detected by evolved gas analysis of the Rocknest aeolian deposit,
972 Gale Crater, Mars. *Journal of Geophysical Research: Planets* **119**, 373–393.
- 973 McDonough W. F. and Sun S. (1995) The composition of the Earth. *Chemical Geology* **120**, 223–
974 253.
- 975 McLennan S. M., Bell J. F., Calvin W. M., Christensen P. R., Clark B. C., de Souza P. A., Farmer J.,
976 Farrand W. H., Fike D. A., Gellert R., Ghosh A., Glotch T. D., Grotzinger J. P., Hahn B.,
977 Herkenhoff K. E., Hurowitz J. A., Johnson J. R., Johnson S. S., Jolliff B., Klingelhöfer G.,
978 Knoll A. H., Learner Z., Malin M. C., McSween H. Y., Pockock J., Ruff S. W., Soderblom L.
979 A., Squyres S. W., Tosca N. J., Watters W. A., Wyatt M. B. and Yen A. (2005) Provenance
980 and diagenesis of the evaporite-bearing Burns formation, Meridiani Planum, Mars. *Earth
981 and Planetary Science Letters* **240**, 95–121.
- 982 McLennan S. M., Boynton W. V., Karunatillake S., Hahn B. C., Taylor G. J. and Mars Odyssey GRS
983 Team (2010) Distribution of Sulfur on the Surface of Mars Determined by the 2001 Mars
984 Odyssey Gamma Ray Spectrometer. **41**, 2174.
- 985 McMahan S., Parnell J., Ponicka J., Hole M. and Boyce A. (2013) The habitability of vesicles in
986 martian basalt Mars habitability. *A&G* **54**, 1.17-1.21.

- 987 McSween H. Y. (2015) Petrology on Mars. *American Mineralogist* **100**, 2380–2395.
- 988 Meyer C. (2016) The Martian Meteorite Compendium.
- 989 Michalski J. R., Cuadros J., Niles P. B., Parnell J., Rogers A. D. and Wright S. P. (2013)
990 Groundwater activity on Mars and implications for a deep biosphere. *Nature Geosci* **6**,
991 133–138.
- 992 Michalski J. R., Dobrea E. Z. N., Niles P. B. and Cuadros J. (2017) Ancient hydrothermal seafloor
993 deposits in Eridania basin on Mars. *Nature Communications* **8**, 15978.
- 994 Michalski J. R., Onstott T. C., Mojzsis S. J., Mustard J., Chan Q. H. S., Niles P. B. and Johnson S. S.
995 (2018) The Martian subsurface as a potential window into the origin of life. *Nature*
996 *Geoscience* **11**, 21–26.
- 997 Morris R. V., Ruff S. W., Gellert R., Ming D. W., Arvidson R. E., Clark B. C., Golden D. C., Siebach
998 K., Klingelhöfer G., Schröder C., Fleischer I., Yen A. S. and Squyres S. W. (2010)
999 Identification of Carbonate-Rich Outcrops on Mars by the Spirit Rover. *Science* **329**, 421–
1000 424.
- 1001 Morrison S. M., Downs R. T., Blake D. F., Vaniman D. T., Ming D. W., Hazen R. M., Treiman A. H.,
1002 Achilles C. N., Yen A. S., Morris R. V., Rampe E. B., Bristow T. F., Chipera S. J., Sarrazin P.
1003 C., Gellert R., Fendrich K. V., Morookian J. M., Farmer J. D., Marais D. J. D. and Craig P. I.
1004 (2018) Crystal chemistry of martian minerals from Bradbury Landing through Naukluft
1005 Plateau, Gale crater, Mars. *American Mineralogist* **103**, 857–871.
- 1006 Mungall J. E., Ames D. E. and Hanley J. J. (2004) Geochemical evidence from the Sudbury
1007 structure for crustal redistribution by large bolide impacts. *Nature* **429**, 546–548.
- 1008 Nambu M. (1968) New mineral Akaganeite, β -FeOOH, from Akagane Mine, Iwate Prefecture,
1009 Japan. *The Journal of the Japanese Association of Mineralogists, Petrologists and*
1010 *Economic Geologists* **59**, 143–151.
- 1011 National Academies of Sciences, Engineering, and Medicine, Division on Engineering and
1012 Physical Sciences, Space Studies Board and Committee on Astrobiology Science Strategy
1013 for the Search for Life in the Universe (2018) *An Astrobiology Strategy for the Search for*
1014 *Life in the Universe.*, National Academies Press (US), Washington (DC). Available at:
1015 <http://www.ncbi.nlm.nih.gov/books/NBK540091/> [Accessed July 23, 2020].
- 1016 Nogami H. H. and Hurley P. M. (1948) The absorption factor in counting alpha rays from thick
1017 mineral sources. *Eos, Transactions American Geophysical Union* **29**, 335–340.
- 1018 Onstott, T.C., McGown D., Kessler J., Lollar B. S., Lehmann K. K. and Clifford S. M. (2006)
1019 Martian CH₄: Sources, Flux, and Detection. *Astrobiology* **6**, 377–395.

- 1020 Onstott T. C., Ehlmann B. I., Sapers H., Coleman M., Ivarsson M., Marlow J. J., Neubeck A. and
1021 Niles P. (2019) Paleo-Rock-Hosted Life on Earth and the Search on Mars: A Review and
1022 Strategy for Exploration. *Astrobiology* **19**, 1230–1262.
- 1023 Onstott T. C., Moser D. P., Pfiffner S. M., Fredrickson J. K., Brockman F. J., Phelps T. J., White D.
1024 C., Peacock A., Balkwill D., Hoover R., Krumholz L. R., Borscik M., Kieft T. L. and Wilson R.
1025 (2003) Indigenous and contaminant microbes in ultradeep mines. *Environmental*
1026 *Microbiology* **5**, 1168–1191.
- 1027 Payré V., Fabre C., Sautter V., Cousin A., Mangold N., Deit L. L., Forni O., Goetz W., Wiens R. C.,
1028 Gasnault O., Meslin P.-Y., Lasue J., Rapin W., Clark B., Nachon M., Lanza N. L. and
1029 Maurice S. (2019) Copper enrichments in the Kimberley formation in Gale crater, Mars:
1030 Evidence for a Cu deposit at the source. *Icarus* **321**, 736–751.
- 1031 Rampe E. B., Ming D. W., Blake D. F., Bristow T. F., Chipera S. J., Grotzinger J. P., Morris R. V.,
1032 Morrison S. M., Vaniman D. T., Yen A. S., Achilles C. N., Craig P. I., Des Marais D. J.,
1033 Downs R. T., Farmer J. D., Fendrich K. V., Gellert R., Hazen R. M., Kah L. C., Morookian J.
1034 M., Peretyazhko T. S., Sarrazin P., Treiman A. H., Berger J. A., Eigenbrode J., Fairén A. G.,
1035 Forni O., Gupta S., Hurowitz J. A., Lanza N. L., Schmidt M. E., Siebach K., Sutter B. and
1036 Thompson L. M. (2017) Mineralogy of an ancient lacustrine mudstone succession from
1037 the Murray formation, Gale crater, Mars. *Earth and Planetary Science Letters* **471**, 172–
1038 185.
- 1039 Redmond H. L. and King S. D. (2004) A numerical study of a mantle plume beneath the Tharsis
1040 Rise: Reconciling dynamic uplift and lithospheric support models. *Journal of Geophysical*
1041 *Research: Planets* **109**. Available at:
1042 <https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/2003JE002228> [Accessed July
1043 9, 2020].
- 1044 Rochette P., Gattacceca J., Chevrier V., Hoffmann V., Lorand J.-P., Funaki M. and Hochleitner R.
1045 (2005) Matching Martian crustal magnetization and magnetic properties of Martian
1046 meteorites. *Meteoritics & Planetary Science* **40**, 529–540.
- 1047 Rochette P., Lorand J.-P., Fillion G. and Sautter V. (2001) Pyrrhotite and the remanent
1048 magnetization of SNC meteorites: a changing perspective on Martian magnetism. *Earth*
1049 *and Planetary Science Letters* **190**, 1–12.
- 1050 Ruff S. W. (2004) Spectral evidence for zeolite in the dust on Mars. *Icarus* **168**, 131–143.
- 1051 Ruff S. W., Campbell K. A., Van Kranendonk M. J., Rice M. S. and Farmer J. D. (2019) The Case
1052 for Ancient Hot Springs in Gusev Crater, Mars. *Astrobiology* **20**, 475–499.
- 1053 Ruff S. W. and Farmer J. D. (2016) Silica deposits on Mars with features resembling hot spring
1054 biosignatures at El Tatio in Chile. *Nature Communications* **7**, 13554.

- 1055 Ruff S. W., Farmer J. D., Calvin W. M., Herkenhoff K. E., Johnson J. R., Morris R. V., Rice M. S.,
1056 Arvidson R. E., Bell III J. F., Christensen P. R. and Squyres S. W. (2011) Characteristics,
1057 distribution, origin, and significance of opaline silica observed by the Spirit rover in
1058 Gusev crater, Mars. *Journal of Geophysical Research: Planets* **116**. Available at:
1059 <https://agupubs.onlinelibrary.wiley.com/doi/10.1029/2010JE003767> [Accessed April 8,
1060 2019].
- 1061 Ruff S. W., Hamilton V. E., Rogers A. D., Edwards C. S. and Horgan B. (2019) Olivine-Rich,
1062 Carbonate-Bearing Ash Deposits Link Jezero and Gusev Craters. **50**, 2775.
- 1063 Ruff S. W., Niles P. B., Alfano F. and Clarke A. B. (2014) Evidence for a Noachian-aged ephemeral
1064 lake in Gusev crater, Mars. *Geology* **42**, 359–362.
- 1065 Schwenzer S. P., Abramov O., Allen C. C., Clifford S. M., Cockell C. S., Filiberto J., Kring D. A.,
1066 Lasue J., McGovern P. J., Newsom H. E., Treiman A. H., Vaniman D. T. and Wiens R. C.
1067 (2012) Puncturing Mars: How impact craters interact with the Martian cryosphere. *Earth*
1068 *and Planetary Science Letters* **335–336**, 9–17.
- 1069
- 1070 Sheppard R. Y., Milliken R. E., Itoh Y. and Parente M. (2019) Mineral Stratigraphy Around Mt.
1071 Sharp Suggests Aqueous Processes Affected the Entire Mound: Directions for Upcoming
1072 Rover Observations from Orbital Data. *LPI Contributions* **2089**, 6289.
- 1073 Sherwood Lollar B., Voglesonger K., Lin L.-H., Lacrampe-Couloume G., Telling J., Abrajano T. a.,
1074 Onstott T. c. and Pratt L. m. (2007) Hydrogeologic Controls on Episodic H₂ Release from
1075 Precambrian Fractured Rocks—Energy for Deep Subsurface Life on Earth and Mars.
1076 *Astrobiology* **7**, 971–986.
- 1077 Sherwood Lollar B., Onstott T. C., Lacrampe-Couloume G. and Ballentine C. J. (2014) The
1078 contribution of the Precambrian continental lithosphere to global H₂ production.
1079 *Nature* **516**, 379–382.
- 1080 Shiga Y. (1987) Behavior of iron, nickel, cobalt and sulfur during serpentinization, with reference
1081 to the Hayachine ultramafic rocks of the Kamaishi mining district, northeastern Japan.
- 1082 Sleep N. H. (2012) Maintenance of permeable habitable subsurface environments by
1083 earthquakes and tidal stresses. *International Journal of Astrobiology* **11**, 257–268.
- 1084 Sleep N. H., Meibom A., Fridriksson T., Coleman R. G. and Bird D. K. (2004) H₂-rich fluids from
1085 serpentinization: Geochemical and biotic implications. *PNAS* **101**, 12818–12823.
- 1086 Squyres S. W., Arvidson R. E., Bell J. F., Calef F., Clark B. C., Cohen B. A., Crumpler L. A., Souza P.
1087 A. de, Farrand W. H., Gellert R., Grant J., Herkenhoff K. E., Hurowitz J. A., Johnson J. R.,
1088 Jolliff B. L., Knoll A. H., Li R., McLennan S. M., Ming D. W., Mittlefehldt D. W., Parker T. J.,
1089 Paulsen G., Rice M. S., Ruff S. W., Schröder C., Yen A. S. and Zacny K. (2012) Ancient
1090 Impact and Aqueous Processes at Endeavour Crater, Mars. *Science* **336**, 570–576.

- 1091 Squyres S. W., Arvidson R. E., Ruff S., Gellert R., Morris R. V., Ming D. W., Crumpler L., Farmer J.
1092 D., Marais D. J. D., Yen A., McLennan S. M., Calvin W., Bell J. F., Clark B. C., Wang A.,
1093 McCoy T. J., Schmidt M. E. and Souza P. A. de (2008) Detection of Silica-Rich Deposits on
1094 Mars. *Science* **320**, 1063–1067.
- 1095 Stamenković V., Beegle L. W., Zacny K., Arumugam D. D., Baglioni P., Barba N., Baross J., Bell M.
1096 S., Bhartia R., Blank J. G., Boston P. J., Breuer D., Brinckerhoff W., Burgin M. S., Cooper I.,
1097 Cormarkovic V., Davila A., Davis R. M., Edwards C., Etiope G., Fischer W. W., Glavin D. P.,
1098 Grimm R. E., Inagaki F., Kirschvink J. L., Kobayashi A., Komarek T., Malaska M., Michalski
1099 J., Ménez B., Mischna M., Moser D., Mustard J., Onstott T. C., Orphan V. J., Osburn M.
1100 R., Plaut J., Plesa A.-C., Putzig N., Rogers K. L., Rothschild L., Russell M., Sapers H., Lollar
1101 B. S., Spohn T., Tarnas J. D., Tuite M., Viola D., Ward L. M., Wilcox B. and Woolley R.
1102 (2019) The next frontier for planetary and human exploration. *Nature Astronomy* **3**,
1103 116–120.
- 1104 Stamenković V., Ward L. M., Mischna M. and Fischer W. W. (2018) O₂ solubility in Martian
1105 near-surface environments and implications for aerobic life. *Nature Geoscience* **11**, 905–
1106 909.
- 1107 Stanley B. D., Hirschmann M. M. and Withers A. C. (2011) CO₂ solubility in Martian basalts and
1108 Martian atmospheric evolution. *Geochimica et Cosmochimica Acta* **75**, 5987–6003.
- 1109 Tarnas J. D., Mustard J. F., Sherwood Lollar B., Bramble M. S., Cannon K. M., Palumbo A. M. and
1110 Plesa A.-C. (2018) Radiolytic H₂ production on Noachian Mars: Implications for
1111 habitability and atmospheric warming. *Earth and Planetary Science Letters* **502**, 133–
1112 145.
- 1113 Thalhhammer O., Stumpfl E. F. and Panayiotou A. (1986) Postmagmatic, hydrothermal origin of
1114 sulfide and arsenide mineralizations at Limassol Forest, Cyprus. *Mineral. Deposita* **21**,
1115 95–105.
- 1116 Therriault A. M., Fowler A. D. and Grieve R. A. F. (2002) The Sudbury Igneous Complex: A
1117 Differentiated Impact Melt Sheet. *Economic Geology* **97**, 1521–1540.
- 1118 Vaniman D. T., Bish D. L., Ming D. W., Bristow T. F., Morris R. V., Blake D. F., Chipera S. J.,
1119 Morrison S. M., Treiman A. H., Rampe E. B., Rice M., Achilles C. N., Grotzinger J. P.,
1120 McLennan S. M., Williams J., Bell J. F., Newsom H. E., Downs R. T., Maurice S., Sarrazin
1121 P., Yen A. S., Morookian J. M., Farmer J. D., Stack K., Milliken R. E., Ehlmann B. L.,
1122 Sumner D. Y., Berger G., Crisp J. A., Hurowitz J. A., Anderson R., Marais D. J. D., Stolper E.
1123 M., Edgett K. S., Gupta S., Spanovich N. and Team M. S. (2014) Mineralogy of a
1124 Mudstone at Yellowknife Bay, Gale Crater, Mars. *Science* **343**. Available at:
1125 <https://science.sciencemag.org/content/343/6169/1243480> [Accessed January 15,
1126 2020].

- 1127 Wafik A., Admou H., Saquaque A., Boukhari A. E. and Juteau T. (2001) Les Minéralisations
1128 Sulfurées à Cu-Fe et les Altérations Associées dans les Ophiolites Protérozoïques de Bou
1129 Azzer et de Khzama (Anti-Atlas, Maroc) (Cu-Fe sulfurous mineralisations and the
1130 associated alterations in the Bou Azzer and Khzama Proterozoic ophiolites, Anti-Atlas,
1131 Maroc). *Ophioliti* **26**, 47–62.
- 1132 Walker R. J., Morgan J. W., Naldrett A. J., Li C. and Fassett J. D. (1991) Re-Os isotope systematics
1133 of Ni-Cu sulfide ores, Sudbury Igneous Complex, Ontario: evidence for a major crustal
1134 component. *Earth and Planetary Science Letters* **105**, 416–429.
- 1135 Wang Z. and Becker H. (2017) Chalcophile elements in Martian meteorites indicate low sulfur
1136 content in the Martian interior and a volatile element-depleted late veneer. *Earth and
1137 Planetary Science Letters* **463**, 56–68.
- 1138 Warr O., Giunta T., Ballentine C. J. and Sherwood Lollar B. (2019) Mechanisms and rates of 4He,
1139 40Ar, and H₂ production and accumulation in fracture fluids in Precambrian Shield
1140 environments. *Chemical Geology* **530**, 119322.
- 1141 Warr O., Sherwood Lollar B., Fellowes J., Sutcliffe C. N., McDermott J. M., Holland G., Mabry J.
1142 C. and Ballentine C. J. (2018) Tracing ancient hydrogeological fracture network age and
1143 compartmentalisation using noble gases. *Geochimica et Cosmochimica Acta* **222**, 340–
1144 362.
- 1145 Weiss B. P., Yung Y. L. and Nealon K. H. (2000) Atmospheric energy for subsurface life on
1146 Mars? *PNAS* **97**, 1395–1399.
- 1147 West M. D. and Clarke J. D. A. (2010) Potential martian mineral resources: Mechanisms and
1148 terrestrial analogues. *Planetary and Space Science* **58**, 574–582.
- 1149 Whitney J. A. (1984) Fugacities of sulfurous gases in pyrrhotite-bearing silicic magmas.
1150 *American Mineralogist* **69**, 68–78.
- 1151 Wiseman S. M., Arvidson R. E., Andrews-Hanna J. C., Clark R. N., Lanza N. L., Marais D. D., Marzo
1152 G. A., Morris R. V., Murchie S. L., Newsom H. E., Dobrea E. Z. N., Ollila A. M., Poulet F.,
1153 Roush T. L., Seelos F. P. and Swayze G. A. (2008) Phyllosilicate and sulfate-hematite
1154 deposits within Miyamoto crater in southern Sinus Meridiani, Mars. *Geophysical
1155 Research Letters* **35**. Available at:
1156 <https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/2008GL035363> [Accessed
1157 March 25, 2020].
- 1158 Wittmann A., Korotev R. L., Jolliff B. L., Irving A. J., Moser D. E., Barker I. and Rumble D. (2015)
1159 Petrography and composition of Martian regolith breccia meteorite Northwest Africa
1160 7475. *Meteoritics & Planetary Science* **50**, 326–352.
- 1161 Wong G. M., Lewis J. M. T., Knudson C. A., Millan M., McAdam A. C., Eigenbrode J. L.,
1162 Andrejkovičová S., Gómez F., Navarro-González R. and House C. H. (2020) Detection of

- 1163 reduced sulfur on Vera Rubin ridge by quadratic discriminant analysis of volatiles
1164 observed during evolved gas analysis. *Journal of Geophysical Research: Planets* **n/a**,
1165 e2019JE006304.
- 1166 Yen A. S., Ming D. W., Vaniman D. T., Gellert R., Blake D. F., Morris R. V., Morrison S. M., Bristow
1167 T. F., Chipera S. J., Edgett K. S., Treiman A. H., Clark B. C., Downs R. T., Farmer J. D.,
1168 Grotzinger J. P., Rampe E. B., Schmidt M. E., Sutter B. and Thompson L. M. (2017)
1169 Multiple stages of aqueous alteration along fractures in mudstone and sandstone strata
1170 in Gale Crater, Mars. *Earth and Planetary Science Letters* **471**, 186–198.
- 1171 Zieg M. J. and Marsh B. D. (2005) The Sudbury Igneous Complex: Viscous emulsion
1172 differentiation of a superheated impact melt sheet. *GSA Bulletin* **117**, 1427–1450.
- 1173 Zolotov M. Y. and Shock E. L. (2005) Formation of jarosite-bearing deposits through aqueous
1174 oxidation of pyrite at Meridiani Planum, Mars. *Geophysical Research Letters* **32**.
1175 Available at: <https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/2005GL024253>
1176 [Accessed January 21, 2020].
- 1177