## The search for clues to abiogenesis on Mars

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#### 12 [First Paragraph]

13 Few traces of Earth's geologic record are preserved from the time of life's emergence, over 14 3800 million years ago. Consequently, what little we understand about abiogenesis-the 15 origin of life on Earth-is based primarily on laboratory experiments and theory. The best 16 geological lens for understanding the early Earth might actually come from Mars, a planet 17 with a crust that's overall far more ancient than our own. On Earth, surface sedimentary 18 environments are thought to best preserve evidence of ancient life, but this is mostly because 19 our planet has been dominated by high photosynthetic biomass production at the surface for 20 the last  $\sim 2500$  million years or more. By the time oxygenic photosynthesis evolved on Earth, 21 Mars had been a hyperarid, frozen desert with a surface bombarded by high-energy solar and 22 cosmic radiation for more than a billion years, and as a result, photosynthetic surface life may 23 never have occurred on Mars. Therefore, one must question whether searching for evidence of 24 life in martian surface sediments is the best strategy. This paper explores the possibility that 25 the abundant hydrothermal environments on Mars might provide more valuable insights into 26 life's origins.

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#### 29 [main text]

30 Following planetary accretion, early delivery via impact of extraterrestrial materials and their 31 payload of volatiles and organic matter may have provided a vast amount of exogenous raw 32 ingredients for abiogenesis<sup>1</sup>. Although the details of the post-accretionary impact period 33 termed the "Late Heavy Bombardment<sup>2</sup>" are intensely debated, consensus is that large 34 impacts were relatively common in the early inner solar system. The catastrophic effects of 35 the impact events would have been a major impediment to the formation, evolution, and 36 preservation of early life, particularly surface life<sup>3</sup>. Yet a mere 800 Myr after the Earth-Moon 37 formed, at the time that the impact rate seems to have diminished, some manner of microbial 38 life appears to have existed<sup>4</sup>.

40 Hints of early life on Earth are found as isotopically light graphitized carbon captured in 41 metamorphic rocks representing the ancient seafloor in what is now Canada<sup>5</sup> and Greenland<sup>6</sup>, 42 and conical stromatolite-like structures within slightly younger rocks<sup>7</sup>. Cryptic evidence in the 43 form of graphite trapped in zircons from the Jack Hills region could push life's origins even 44 earlier into the Hadean  $eon^8$ . These remnants of the Archean and Hadean eons that are so 45 relevant to understanding the temporal and taphonomic window for life's emergence comprise only about ~0.001 vol.% of the terrestrial crust<sup>9</sup> and have been intenselv thermally 46 47 and chemically altered due to their long crustal residence times.

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Because the Earth's early geologic record is so poorly preserved, our limited understanding of how early organic chemistry may have assembled the building blocks of life is largely based upon laboratory experiments<sup>10–15</sup>. But definitive clues to the chemical steps leading to life's origins probably require empirical evidence. The fundamental question of how abiogenesis occurred on Earth may only be answerable through finding better preserved "cradle of life" chemical systems beyond Earth. Indeed, this question of how life originates is one of the fundamental drivers of international space exploration.

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57 Which objects beyond Earth could potentially unlock the mystery of abiogenesis? Europa and 58 Enceladus are high priority targets because they likely contain subsurface oceans even 59 today<sup>16,17</sup>. Yet, it is not simply a subsurface ocean itself that is intriguing in terms of the 60 origin of life perspective-it is the reaction between fluids and silicate rocks at the oceansilicate interface<sup>18</sup> that might hold promise for energetic pathways for chemotrophic life 61 forms<sup>19</sup>. However, all of the icy satellites are far from Earth and access to subsurface fluids 62 63 will either require deep drilling, or we will be limited to collection of ejected molecules from transient cryo-volcanism<sup>20</sup>,<sup>21</sup>, which may not sample fluids from the deep rock-water interface 64 65 of primary interest. In addition, there is a growing interest in the possibility that terrestrial life 66 originated not within an ocean environment but rather in vapour-dominated inland geothermal 67 systems, where shallow pools of fluid may have interacted with porous silicate minerals and metal sulphides<sup>22,23</sup>. While the icy worlds are clearly a high-priority target for understanding 68 69 abiogenesis, Mars is the only Solar System object with an ancient, preserved, accessible crust 70 containing clear evidence of water-rock reactions dating to the time when life appeared on 71 Earth (Figure 1).

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#### 73 Mars as a Rosetta Stone for Early Earth

Mars, a planet without plate tectonics and with much lower weathering rates through most of its history<sup>24</sup>, contains a much older and better-preserved geologic record than the Earth

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(Figure 2). At only ~10% of Earth's mass, Mars began with far less primordial and radiogenic
heat<sup>25</sup>. By about 4000 Mya, Mars had cooled sufficiently to cause cessation of the magnetic
dynamo<sup>26</sup>. The loss of the martian magnetic field marked the timing of its clearest divergence
from the evolution of the Earth and its biosphere. It exposed the martian surface to punishing
radiation<sup>27</sup>, and the atmosphere began to be sputtered away by solar wind<sup>28</sup>.

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82 Mars may have been cold, arid, oxidizing and generally inhospitable at the surface for much 83 of its history, however hydrothermal conditions in the near surface or subsurface might have 84 been considerably more clement. Infrared remote sensing has revealed the presence of 85 thousands of deposits of hydrated silicate minerals as well as various salts throughout the 86 martian surface<sup>29</sup>. While essentially all of the salts and some of the hydrated silicates seemingly formed in surface environments during what may have been short lived climate 87 excursions<sup>30</sup>, many of the deposits represent materials that were seemingly exhumed from the 88 subsurface<sup>31</sup>. Among the exhumed phases are serpentines, Fe and Mg-rich smectite clavs, 89 90 chlorites, carbonates, and amorphous silica that seemingly indicate widespread subsurface 91 hydrothermal alteration (Figure 3).

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In 2008, the Spirit Rover also stumbled upon soils of nearly pure opaline silica (>90 wt %
SiO<sub>2</sub>) in the vicinity of Home Plate in Gusev crater<sup>32</sup> providing compelling evidence for
fumarolic hydrothermal activity. Similar materials were also detected in at least one younger
caldera, that on Nili Patera<sup>33</sup>.

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It is difficult to strongly constrain the timing of near surface and deep subsurface hydrothermal alteration other than to state that it was primarily in the Noachian (>3600 Mya)<sup>31</sup>. Whereas some studies document impact-induced hydrothermal activity in Hesperian crater deposits (3000-3600 Mya) e.g.<sup>34</sup>, there is little to no evidence for similar alteration in craters formed in Amazonian age terranes<sup>35</sup> (<3000 Mya) (Figure 3).

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104 The subsurface - from metres to kilometres depth - is potentially the largest, longest-lived, and most stable habitable environment on Mars<sup>36</sup>. A significant fraction of Earth's biomass 105 consists of prokaryotic microbial life in a deep biosphere<sup>37</sup>, a habitat that was essentially 106 107 disregarded more than 30 years ago and remains largely unexplored today<sup>38</sup>. The primary 108 producers in deep subsurface ecosystems are anaerobic chemoautotrophs, or SLiMEs, that 109 oxidize H<sub>2</sub> and reduce CO<sub>2</sub> to produce CH<sub>4</sub> (i.e. methanogens) and acetate (i.e. acetogens)<sup>39,40</sup>. 110 The life-sustaining  $H_2$  has been shown to be generated through abiotic hydrolysis of ferrous minerals in basalt<sup>41</sup> and ultramafic rock (e.g. serpentinization)<sup>42</sup> and through radiolysis of 111 water<sup>49</sup>. Other potential sources of H<sub>2</sub> include exsolved gases from basaltic magmas, 112

113 decomposition of  $CH_4$  at  $T > 600^{\circ}C$ , reactions between  $CH_4$ ,  $H_2O$  and  $CO_2$  at elevated 114 temperatures and silicate cataclasis. Just as important, radiolysis has been shown to generate 115 electron acceptors such as sulphate along with  $H_2$ , which can be utilized to sustain sulphate 116 reducing bacteria indefinitely<sup>43</sup>.

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118 On Earth, the extent of the deep biosphere is controlled not only by energy sources and 119 nutrients, but by availability of pore space. While porosity is strongly dependent on rock type, 120 continental rocks typically have <1-5% porosity at depth of 3-4 km. But due to the lower 121 gravity on Mars the rocks are less compacted; similar values of porosity extend to ~10 km in 122 depth<sup>44</sup>.

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124 Although heat flow in the terrestrial crust is heterogeneous, geothermal gradients (10-40 125 K/km) in continental and oceanic crustal settings suggest that the terrestrial deep biosphere 126 likely does not extend passed  $\sim$ 3-4 km depth, beyond which the most tolerant thermophiles 127 are no longer viable ( $\sim$ 120°C)<sup>41</sup>. However, on Mars the lower surface temperature and lower 128 crustal heat flow add up to a more favourable thermal regime within the crust. Assuming a 129 thermal gradient of 20 K/km on Noachian Mars, the 120°C temperature limit would not have 130 been reached until nearly twice the depth where it occurs on Earth (Figure 4).

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132 Most of the martian crust is ultramafic or mafic, and likely contains interlayered volcanics 133 and impactites. Given the lower temperature gradient on Mars compared to Earth, it is likely that Lost City-type<sup>45</sup> (low-temperature, alkaline) serpentinization reactions<sup>19</sup> occurred over a 134 large range of depths on Mars, producing bioavailable  $H_2^{46}$ . Although this mafic-rich crust is 135 136 less radiogenic that average Earth continental crust, H<sub>2</sub> production rates from radiolysis 137 should be as great as that for subsurface environments on the Earth because of the greater 138 porosity of the martian subsurface<sup>47</sup>. Exhumed subsurface carbonates, and the presence of 139 vein carbonates in martian meteorites exhumed from the subsurface<sup>48</sup> suggest that these 140 reactions happened in the presence of CO<sub>2</sub> and may have produced abiogenic hydrocarbons. 141 Consequently, the subsurface habitable volume and abiotic energy sources would likely have 142 been as readily available, if not more so, on Mars as on Earth.

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144 It is probable that fluids within alkaline crustal hydrothermal systems would have mixed with 145 descending acidic, sulphur- (H<sub>2</sub>S, SO<sub>2</sub>) and CO<sub>2</sub>-rich fluids from surface and near-surface 146 environments through taliks, areas of unfrozen ground surrounded by permafrost<sup>44</sup>. Likewise, 147 alkaline fluids might have emerged in deep basins and interfaced with acidic lakes and 148 meltwater from acidic ice deposits, resulting in mixing scenarios which may have been a source of redox energy<sup>19,49</sup>. A test for such an origin of life scenario would be invaluable to
 earth and planetary scientists alike.

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#### 152 Dim prospects for surface life on Mars

153 The evolutionary innovation of oxygenic photosynthesis by cyanobacteria was a turning point 154 in the history of life on Earth<sup>50</sup>. Although the timing remains controversial, oxygenic 155 photosynthesis appeared late within cyanobacterial evolution<sup>51</sup>, well after their divergence at 2.5 to 2.6 billion years<sup>52</sup> and after the rise of its evolutionary precursor, Mn-oxidizing 156 phototrophy<sup>53</sup> and before the Great Oxidation Event at 2.3-2.45 Ga<sup>54,55</sup>. Production of 157 158 atmospheric  $O_2$  led to the formation of ozone, which shielded the immediate surface zone 159 from harmful UV rays. The success of cyanobacteria not only led to marked increases in 160 biomass production and deposition in shallow water environments (shelf, coastal marine, and 161 lacustrine) where high sedimentary rates prevail, but also to the colonization of arid and cold surface environments by endolithic communities<sup>56</sup>. Our paleontologic record over the last 162 163 ~3000 Mya is dominated by carbonaceous sedimentary rocks from such environments<sup>57</sup>.

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165 On Mars, there may have never been an evolutionary drive to inhabit the surface. During the 166 Noachian, Mars was most likely cold, arid and oxidizing<sup>58,59</sup>. Fluvial channels and most crater 167 lakes on Noachian Mars once thought to have required some form of greenhouse atmosphere 168 in order to stabilize liquid water over geologic time scales, are now considered by some to 169 have formed within thousands of years<sup>60,61</sup>, perhaps under a tenuous atmosphere in the 170 Noachian. The surface seemingly shifted from a cold but episodically wet landscape to a 171 frozen, hyperarid desert at the Noachian-Hesperian transition ca. 3600 Mya<sup>62</sup>.

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173 The success of surface life on Earth can be traced back to the evolution of oxygenic 174 photosynthesis in the Archean. The most recent molecular clocks have placed the origin of 175 photosynthesis at ~3000 Myr and the origin of oxygenic photosynthesis later than 2500 to 176 2600 Mya on Earth. Martian phototrophs would have had to attain these evolutionary 177 benchmarks by 3600 Mya, despite the generally frozen and arid surface conditions, fainter 178 sunlight, and the intense radiation flux from solar UV, Solar Energetic Particles (SEPs) and 179 Galactic Cosmic Rays (GCRs). By contrast, the evolution of methanogenesis, an important 180 metabolic pathway for subsurface life, occurred prior to the divergence of Euryarcheota and 181 Crenarcheota and represents one of the most ancient forms of metabolism<sup>63</sup>.

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183 Considering these challenges, it seems prudent to consider the possibility that photosynthesis
184 never evolved on Mars. Unless high-energy radiation could be harnessed as a form of energy,
185 as has been reported for certain fungal species<sup>64</sup>, the radiated surface environment is an

impediment to the existence of surface life and an obstacle for the preservation of organic
 materials<sup>36</sup>. With all this in mind, it seems time to reconsider the current Mars exploration
 philosophy.

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# 190 A Mars exploration strategy focused on abiogenesis

191 Much of the thinking about candidate landing sites for future landed missions has been aimed 192 at maximizing taphonomic potential by targeting sedimentary environments such as lacustrine 193 delta deposits. While this Mars exploration strategy is understandable, such an approach 194 suffers a major epistemological problem: Mars is not Earth. We must recognize that our entire 195 perspective on how life has evolved and how evidence of life is preserved is colored by the 196 fact that we live on a planet where photosynthesis evolved. Even if photosynthesis did evolve 197 on Mars, questions remain as to how successful surface life would have been, and whether 198 evidence of that life could have been captured in the sedimentary record.

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200 Considering that some of the most ancient analogue habitats on Earth, hydrothermal and 201 subsurface environments, are mirrored on Mars, it is logical to search for the signs of 202 primitive life there in settings analogous to where it may have emerged here. We thereby not 203 only maximize our chances of finding chemotrophic life, but also of finding the evidence of 204 prebiotic chemistry that might have led to the formation of life in a sustained habitable 205 setting.

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The search for biosignatures in hydrothermal deposits must also be questioned. For example, silica sinter deposits of the type found in Gusev crater<sup>32</sup> are widely considered deposits with high preservation potential for textural and chemical biomarkers on Earth. But, a significant fraction of the biomass and biosignatures associated with silica sinters correspond to photosynthetic bacteria that thrive in fluid mixing zones<sup>65</sup> and therefore the effects of a possible absence of photosynthesis on biosignature preservation should also be considered in this context.

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215 Potential biosignatures in exhumed deep crustal rocks include the following: 1) isotopic 216 signatures of gasses (e.g. CH<sub>4</sub>) trapped in fluid inclusions, 2) isotopic signatures of minerals, 217 fluids and organic matter trapped in veins and diagenetic replacements<sup>66</sup>, 3) metal or 218 carbonate accumulations at redox gradients—especially indicating disequilibrium conditions, 4) biotextures in fractures and pores, 5) microfossils preserved in mineralized veins<sup>66</sup> or 219 220 diagenetic cements and concretions, and 6) important organic molecules such as nucleic 221 acids, lipids, and amino acids in fractures, fluid inclusions, and within mineral aggregates<sup>67,68</sup>. 222 The detection of disequilibrium chemistry implicating life may perhaps be less satisfying than the detection of fossilized microbial mats in lacustrine sediments, but such an approach might actually teach us more about the origin of life. Because the chemical signatures from the dawn of life have been entirely obliterated on Earth, finding these clues on Mars, a unique site within the Solar System, would provide an invaluable window into our own history.

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Given how little we understand about the origin of life on Earth, it makes sense to adopt a broader plan to seek signs of life. In other words, it is perhaps more logical to seek evidence of prebiotic chemistry that might have led to the formation of life in sustained habitable settings rather than searching directly for evolved forms of surface life in ephemeral environments. We could search for the signs of primitive life on Mars in settings analogous to where it may have formed on Earth.

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235 While concerns about the preservation potential of biosignatures in rocks from hydrothermal 236 and subsurface martian environments are important to consider, it is clear that preservation 237 potential does not present an ultimate stumbling block. The preservation of biomolecules 238 associated with hydrothermal activity in the extraterrestrial context has been validated by 239 their common occurrence in hydrous meteorites with signs of ancient hydrothermal 240 processing  $(\leq 150^{\circ}C)^{69}$ . Upon the cessation of the hydrothermal event, plunging temperatures 241 in martian environment would be ideal for preserving biosignatures (e.g. amino acid 242 enantiomeric ratio)<sup>70</sup>. Silica has been recognized for its significance in microfossil 243 preservation, and iron-silicate biomineralization in hot spring environments has been shown 244 to serve as a potent shield to UV radiation<sup>71</sup>. Biomarker preservation in subsurface 245 environments is a field that has hardly been explored, but biomarkers from Cretaceous 246 subsurface environments clearly demonstrate that preservation is possible<sup>67</sup>.

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By focusing our search on non-photosynthetic life, we not only maximize our chances of finding biosignatures on Mars but also uncovering clues to abiogenesis, an aspect that should be a key part of our exploration strategy. The quest to understand life's origins could be described as "Follow the energy sources<sup>46</sup>: sulphur, iron and H<sub>2</sub>." That mantra would lead us to Mars, an iron and sulphur-rich planetary crust with abundant evidence for ancient hydrothermal activity and H<sub>2</sub> production that could have fuelled an early chemosynthetic biosphere.

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## 260 Methods

The map of hydrothermal and subsurface mineral deposits on Mars (Figure 3) was derived from multiple sources. The primary data sources include mineral detections by Carter et al.<sup>29</sup> and Ehlmann et al.<sup>31</sup>, which were created with significant input from the science instrument teams for the Observatoire pour la minéralogie, l'eau, les glaces et l'activité (OMEGA)<sup>72</sup> and the Compact Reconnaissance Imaging Spectrometer for Mars (CRISM)<sup>73</sup>. These instruments have produced 1000s of detections of hydrated minerals on Mars, many of which correspond to contexts in surface environments and many of which correspond to deposits exhumed from the subsurface. All of the detections shown in Figure 3 correspond to detections that have seemingly been exhumed from the subsurface by impact or erosion.

The edited global-scale datasets of Carter and Ehlmann were supplemented with other information pertaining to the detection of subsurface, surface or near-surface hydrothermal deposits. Subsurface carbonate detections were supplemented with data from studies of exhumed carbonates<sup>74</sup> and a global carbonate<sup>75</sup> study. Serpentine deposits include those described in a global search for serpentinized rocks<sup>76</sup>. Fumarolic silica corresponds to silica detected by the Spirit rover<sup>77</sup> and with CRISM. Seafloor-type clays correspond to Fe- and Mg-rich phyllosilicates and carbonates with in the Eridania basin on Mars, which was the site of a large inland sea when the deposits formed  $>3800 \text{ Mya}^{78}$ .

296 Figure Captions:

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298 Figure 1: A comparison of the age of planetary crust. Lines represent the best estimate 299 limits of oldest preserved crust. Dashed lines represent significant uncertainties. The crust of 300 Mars might provide the best window into the time when abiogenesis occurred on Earth.

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302 Figure 2: A comparison of key events in the histories of the Earth and Mars. The area of 303 each time line is an approximation of the amount of crust preserved from over different 304 epochs. The generally unmetamorphosed and well-preserved geologic record of early Mars is 305 an invaluable window into the geology and prebiotic chemistry of the early Earth.

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307 Figure 3: Hydrothermal and exhumed, altered subsurface deposits on Mars. The global 308 occurrence of alteration minerals formed in deep crustal or surface hydrothermal 309 environments detected by infrared remote sensing. See Methods for explanation of data 310 included in the map.

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312 Figure 4: A comparison of the average porosity of thermal gradients of the crusts of 313 Earth and Mars. For similar rock types and surface porosities, the martian crust contains 314 significantly more porosity to greater depth than that of the Earth (left). Estimated thermal 315 gradients for Noachian ( $\phi_N$ ) and modern ( $\phi_m$ ) Mars are lower than that of the modern 316 continental ( $\phi_c$ ) or oceanic ( $\phi_o$ ) crust of Earth (right). A hypothetical 120°C limit is 317 encountered at 3-4 km depth on Earth, where the porosity is 1-2%. The same temperature 318 limit would not be encountered until ~6 km depth on Noachian Mars or much deeper on 319 modern Mars.

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