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Concentrated perchlorate at the Mars Phoenix landing site: Evidence for thin film liquid water on Mars

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[1] NASA's Phoenix mission, which landed on the northern plains of Mars in 2008, returned evidence of the perchlorate anion distributed evenly throughout the soil column at the landing site. Here, we use spectral data from Phoenix's Surface Stereo Imager to map the distribution of perchlorate salts at the Phoenix landing site, and find that perchlorate salt has been locally concentrated into subsurface patches, similar to salt patches that result from aqueous dissolution and redistribution on Earth. We propose that thin films of liquid water are responsible for translocating perchlorate from the surface to the subsurface, and for concentrating it in patches. The thin films are interpreted to result from melting of minor ice covers related to seasonal and long-term obliquity cycles.

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1. Introduction

[2] One of the primary goals of NASA's Phoenix mission, which landed on the northern plains of Mars on 25 May 2008, was to characterize the past and present water-ice cycle on Mars, including the concentrations of salt species in the soil [Smith *et al.*, 2009]. To search for soluble salts, three soil samples were added to Phoenix's Wet Chemistry Laboratory (WCL) [Kounaves *et al.*, 2009], which registered a strong signal that was interpreted as resulting from perchlorate [Hecht *et al.*, 2009]. Because perchlorates are strongly deliquescent salts, their homogeneous distribution through the soil column was cited as evidence that the Phoenix soils have not interacted extensively with liquid water [Hecht *et al.*, 2009].

[3] In this paper, we use data from Phoenix's Surface Stereo Imager (SSI) to map the perchlorate distribution at the Phoenix landing site. Because several minerals are spectrally similar to perchlorate (including bischofite and some zeolite and phyllosilicate minerals), we also use a

geochemical model to assess the stability of these minerals under conditions at the Phoenix landing site.

2. Methods

2.1. Spectral Mapping

[4] Phoenix's SSI instrument acquired images and spectra (0.445 to 1.001 μm) of the landing site (68.2188N, 234.2508E, IAU 2000 areocentric), including the 12 trenches that Phoenix excavated using its Robotic Arm (RA, Figure 1) [Arvidson *et al.*, 2009]. With SSI, we examined soil features in each of the 12 trenches and compared them to undisturbed surface soils.

[5] For identification purposes, a perchlorate spectrum was obtained with an ASD spectrometer in a Mars-like chamber filled with a dry N₂ atmosphere (0.4% relative humidity, 100 ppmv H₂O, T ~ 23°C) after purging dry N₂ for 1176 hours. The perchlorate sample was tested with powder XRD and confirmed to be Mg(ClO₄)₆H₂O.

2.2. Geochemical Modeling

[6] Because zeolite minerals are spectrally similar to perchlorate, a model was created to evaluate the plausibility of zeolite formation at the Phoenix landing site. In this model, a fluid of the composition reported by the WCL measurements that was in equilibrium with calcite [Boynton *et al.*, 2009] at a partial pressure of CO₂ of 5.7 mbar [Tamppari *et al.*, 2009] and contained an estimated sulfate content for charge balance was reacted with equal amounts of hematite and mineral components of basaltic sand [McSweeney *et al.*, 2006]. This assemblage was chosen to approximate the chemical environment present at the Phoenix lander site.

[7] All calculations were performed using The Geochemist's Workbench® [Bethke, 2009]. The Lawrence Livermore National Laboratory thermochemical database [Delaney and Lundeen, 1990] was employed with an extended Debye-Huckel activity correction model that is parameterized to be accurate in up to 3 m NaCl solution and approximately 0.5–1 m ionic strengths of other electrolytes [Helgeson, 1969; Helgeson and Kirkham, 1974a, 1974b]. All calculations were performed at 25°C as the database lacked thermodynamic data for zeolites at other temperatures. Chabazite was not included in the database and was thus not considered in the calculations.

3. Results

3.1. Spectral Mapping Results

[8] We identified small, relatively high albedo soil patches (Figure 2) with unique spectral properties in material excavated during generation of six trenches: Dodo-Goldilocks,

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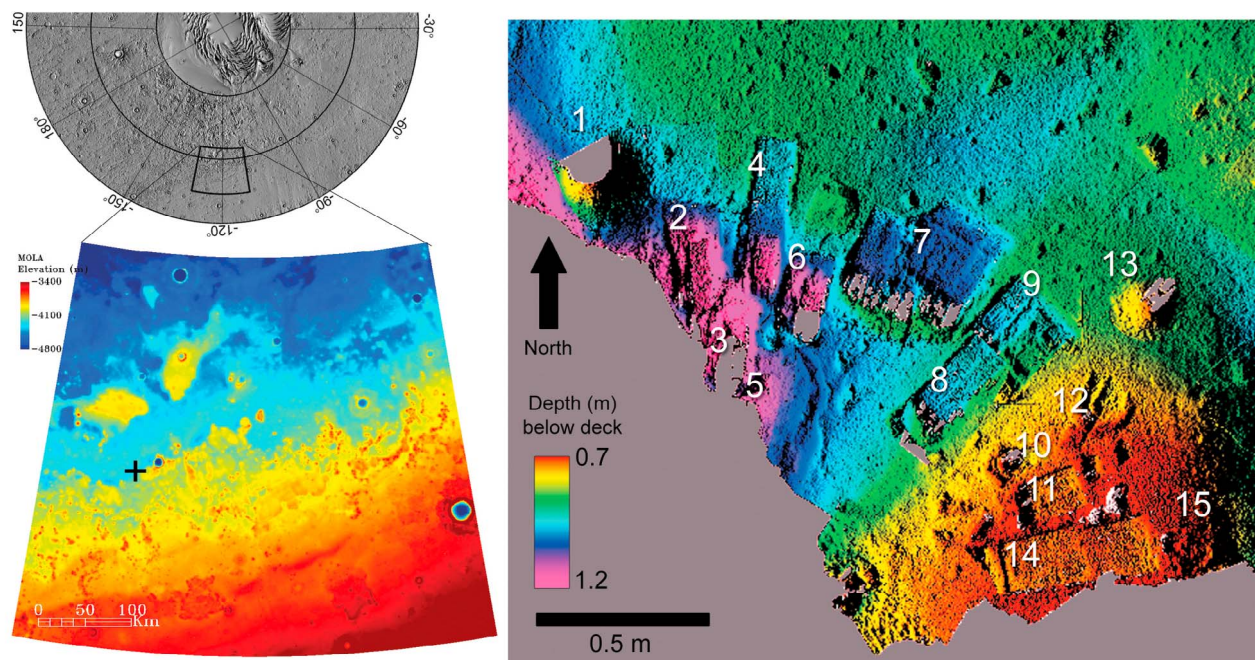


Figure 1. (left) Phoenix landing site marked by a cross. (right) The lander excavated 12 trenches and produced three dump piles along its northeast side: Caterpillar Dump (1), Dodo-Goldilocks (2), Stone Soup (3), Upper Cupboard (4), Lower Cupboard (5), Ice Man (6), La Mancha (7), Neverland (8), Pet Donkey (9), Bear's Lodge (10), Burn Alive 3 (11), Runaway (12), Bee Tree Dump (13), Snow White (14), and Croquet Ground Dump (15).

Upper Cupboard, Snow White, Ice Man, Neverland, and Stone Soup. SSI spectra of the patches have a steep positive slope between 0.445 and $0.65\ \mu\text{m}$, typical of the “red edge” resulting from nanophase iron oxide materials [Morris *et al.*, 2006]. Unlike other Phoenix soils, which are featureless and relatively flat from 0.65 to $1.001\ \mu\text{m}$, the spectra of these patches also have a pronounced minimum reflectance at $0.967\ \mu\text{m}$ (Figure 3).

[9] The patches with the $0.967\ \mu\text{m}$ feature are each several millimeters across and appear as distinct surface patches on soil clods and exposed trench floors. The clods are subangular fragments that keep their form under Mars surface conditions over the Phoenix mission timescale. The patches on trench floors occur as rounded lumps, morphologically similar to smoothed trench floor soil. The patches are found only in areas of disturbed soil (trenches and dump piles); none are observed on undisturbed surface soil, and none are found in contact with ice.

[10] Spectra of soils with the $0.967\ \mu\text{m}$ feature did not change through time. Each patch was uncovered during an RA excavation, exposed for two or more sols, and in some cases re-buried by new trenching. One patch was left undisturbed for 113 sols, during which no changes were observed to the shape of its spectrum, its overall albedo, or the depth of the $0.967\ \mu\text{m}$ feature. In addition, the size and shape of the clods did not change. Thus, the patches are stable on the surface on timescales of hundreds of sols.

[11] The feature is interpreted as an absorption feature due to the third overtone of the asymmetric OH stretch, which can result from either bound water or OH within the mineral structure [Roush *et al.*, 1997].

3.2. Geochemical Modeling Results

[12] Zeolites with potential spectral matches to the $0.967\ \mu\text{m}$ feature include heulandite, chabazite, laumontite, mesolite, natrolite, scolecite, and stilbite [Crowley, 1991]. Of these, only heulandite, chabazite, laumontite, and natrolite are reported to form under low-temperature conditions consistent with pedogenic formation at the Phoenix lander site [Passaglia and Sheppard, 2001; Ming and Boettinger, 2001]. Zeolite crust formation would require aqueous pedogenic processes, with weathering of basaltic sand material originating as dust providing the main chemical components required for zeolite formation.

[13] Our geochemical modeling demonstrates that this reaction will not produce any of the plausible zeolite phases. All zeolites that potentially match the feature are thermodynamically unstable in this system (Figure 4a). Mesolite is the most likely zeolite to form under Phoenix conditions; however, this phase has not been reported to occur or form in any soil or sedimentary environments on Earth [Ming and Boettinger, 2001; Sheppard and Hay, 2001], suggesting that formation of this phase under non-hydrothermal conditions is inhibited. Our modelling shows that smectites (Fe/Mg saponite and nontronite) and minor gibbsite are the most thermodynamically stable weathering products at the Phoenix site (Figure 4b). Gibbsite is likely a proxy for Al incorporation into the predicted smectites, which is not well accounted for in the thermodynamic model as data is only available for smectites of fixed end-member composition.

4. Discussion

4.1. Patch Spectral Properties

[14] The high-albedo patches all have pronounced absorptions at $0.967\ \mu\text{m}$. The $0.967\ \mu\text{m}$ filter is prone to

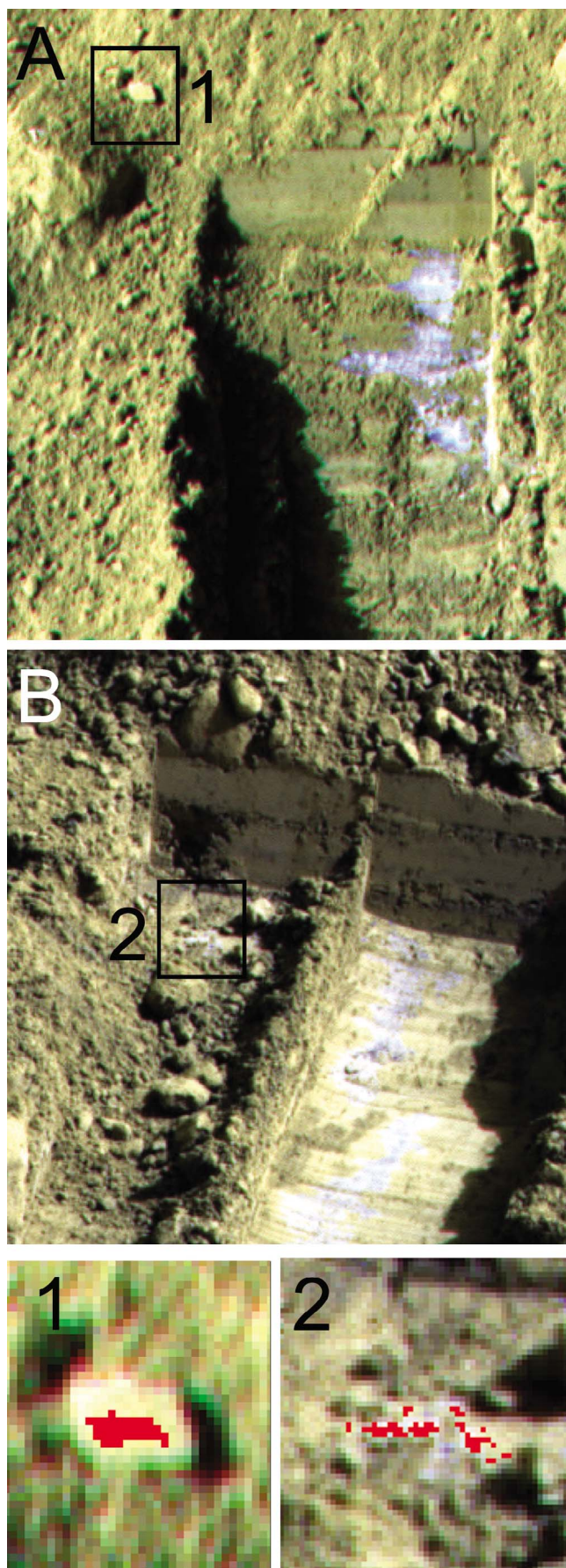


Figure 2

artifacts, because it was the final filter taken during the sequence of images, and illumination conditions can change. However, we have used several criteria to rule out the possibility of artifacts. First, the pixels are clumped together, not randomly distributed across an image. Second, they are not located in or near shadows. Third, they were observed repeatedly and have the same feature on multiple sols (up to 113 sols). Fourth, these pixels have the same feature in images taken at multiple phase angles (Figure 3), ruling out the possibility of a viewing geometry artifact. Fifth, each image had a slightly different pointing, so the pixels in question are not confined to specific parts of the detector.

[15] We conclude that the $0.967\ \mu\text{m}$ feature is not an artifact, but rather is caused by a material exposed during RA operations. Additionally, because the feature is found only as patches on the clods, we conclude that the material is a surface coating or crust.

4.2. Patch Spectral Candidates

[16] Water ice, which is also observed in trenches, has absorptions at 0.98 and $1.04\ \mu\text{m}$, which cause a negative slope from 0.85 to $1.001\ \mu\text{m}$. Water ice is a poor spectral fit to spectra of the patches described here, thus is an unlikely candidate. Additionally, the long-term stability of these patches rules out water ice, which was observed to sublime in a matter of hours to sols when exposed at the surface [Smith *et al.*, 2009].

[17] Several minerals (e.g., Figure 3c) produce a $0.967\ \mu\text{m}$ feature, including hydrated Mg- and Ca-perchlorates, some zeolite and phyllosilicate minerals, and at least one hydrated chloride mineral (bischofite [Crowley, 1991]). WCL could detect and measure Cl ions, and did not detect bischofite [Kounaves *et al.*, 2010a, 2010b]. Our geochemical modeling indicates that zeolites are unlikely candidates for the patches described here, because they are unlikely to form under Phoenix site conditions (see Section 3.1 above). Zeolite formation generally requires an aqueous environment with $\text{pH} > 9$ [Sheppard and Hay, 2001], and though the Phoenix landing site may have experienced aqueous activity in its past [Smith *et al.*, 2009], the 7.7 pH of the soil appears to be buffered by calcite [Boynton *et al.*, 2009], making a highly alkaline aqueous environment unlikely.

[18] In one soil sample, Phoenix's Thermal and Evolved Gas Analyzer (TEGA) recorded a very small H_2O release between 700 and 800°C that was interpreted as possible evidence of a phyllosilicate mineral (possibly smectites [Boynton *et al.*, 2008]). However, material discussed here is present only in concentrated subsurface patches, indicating that some mechanism is translocating them down in the soil

Figure 2. Two examples of patches that display the $0.967\ \mu\text{m}$ feature. (a) A clod (inset 1, pixels with feature marked in red) at Dodo-Goldilocks (sol 18, image token 125C) does not appear in images taken on before the sol 18 excavation. We conclude that the clod was either dropped by the RA on its way to the Caterpillar Dump (left and below the image), or rolled off the dump pile after dumping. (b) A patch at Snow White (sol 16, image token 1314) appeared after the sol 24 excavation (inset 2, pixels with features marked in red). This patch is close to, but does not appear to be in contact with, the Snow White ice.

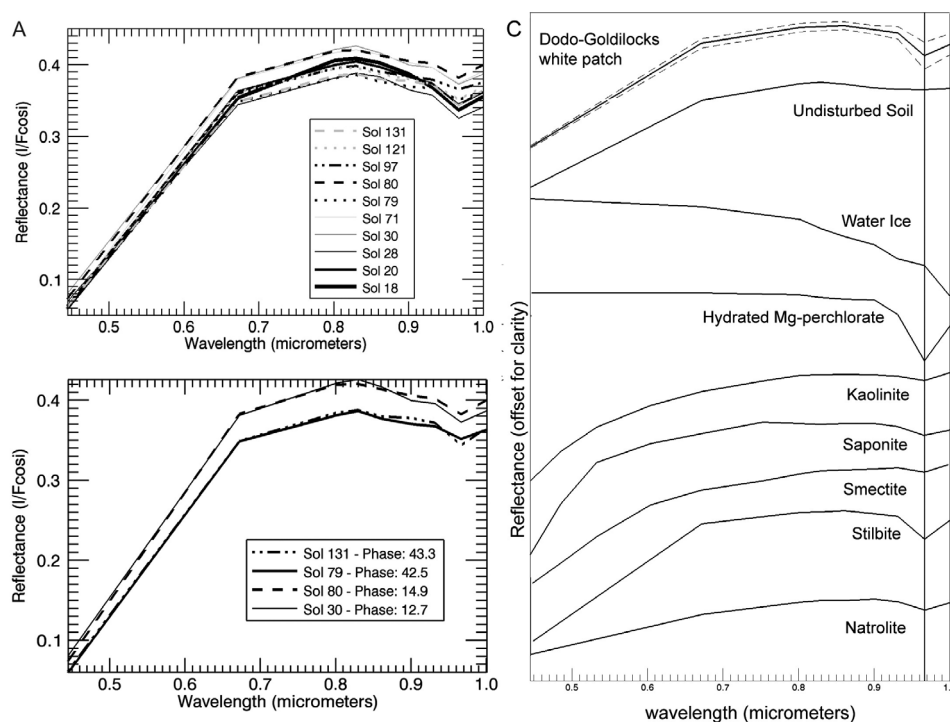


Figure 3. (a) Spectra of the clod pictured in Figure 2a, taken over 113 sols. The feature at $0.967 \mu\text{m}$ does not appear to change during that time. (b) The brightness of the spectrum does vary through time; however, this is clearly due to differences in viewing geometry. Each of these spectra is generated from averaging together the same group of approximately 30 pixels. The standard deviation for this group of pixels is shown in Figure 3 and is approximately ± 0.017 . (c) Spectra of candidate minerals for the $0.967 \mu\text{m}$ feature. A representative patch from Dodo-Goldilocks is shown on top, with the dotted lines illustrating the standard deviation of patch spectra.

column, and concentrating them into patches. Phyllosilicate minerals have a very low solubility and would not be expected to be redistributed by dissolution and reprecipitation in water. Aqueous clay illuviation can physically move phyllosilicates from the surface to subsurface, forming argillic horizons or clay coatings on peds; however, this process requires repeated flushings with a substantial quantity of water [e.g., *Eswaran and Sys*, 1979]. Additionally, Phoenix soils undergo pedoturbation resulting from seasonal

freeze-thaw cycles [*Mellon et al.*, 2008]; hence, the high-volume wetting events would have had to occurred recently in order for the argillic horizons to remain intact. We see no evidence to suggest that the Phoenix landing site has been repeatedly flushed with large volumes of liquid water in the geologically recent past, and so conclude that physically translocated phyllosilicates are unlikely candidates for the patches reported here.

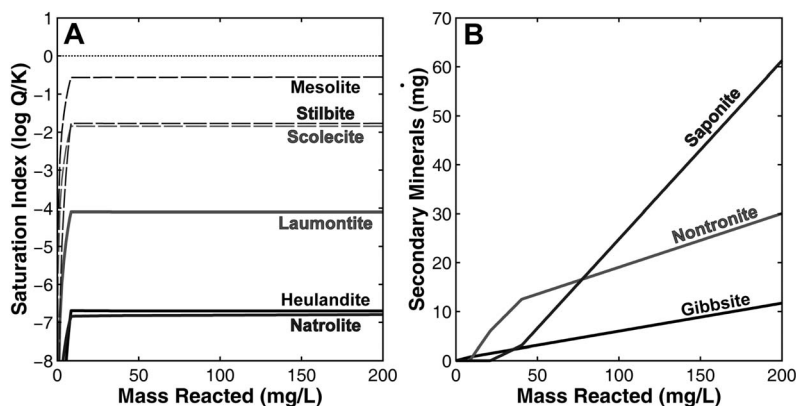


Figure 4. (a) Zeolite mineral saturation indices and (b) mass of secondary minerals produced as a function of the mass of a 1:1 mixture of hematite and basaltic sand reacted with a fluid phase representative of the site. K is the equilibrium constant for the reaction, Q is the reaction quotient, and the mineral saturated in the system when $K = Q$. None of the minerals shown here is saturated in the system.

[19] Additionally, our geochemical modeling suggests that the stable phyllosilicates would all contain substantial iron because of their formation from basaltic material, and they would display a green or brown color, which is inconsistent with the observed crust. Their original formation would also require substantial aqueous alteration of basaltic sand; corroborating evidence for such alteration at the site is lacking.

[20] Perchlorate, on the other hand, is highly soluble in even very small amounts of water, and would be easily transported from the surface to the subsurface by fluids. In the subsurface, it would form concentrated crusts as the water evaporated or froze then sublimated. Given the previous WCL detection of perchlorate at the site, the thermodynamic instability of zeolites, the concentrated morphology of the observed patches, and their location in the soil column, we conclude that a hydrated perchlorate salt is the most likely candidate to explain the $0.967\ \mu\text{m}$ absorption feature.

4.3. Perchlorate at the Phoenix Landing Site

[21] The observation that concentrated perchlorate patches are limited to the subsurface is not inconsistent with previous findings from the Phoenix WCL, which reported perchlorate throughout the soil column, including a sample from near the surface (actually a scoop sample of the upper $\sim 1\ \text{cm}$ of soil) [Hecht et al., 2009]. Taken together, the WCL and SSI observations indicate that the soil column contains low concentrations of evenly dispersed perchlorate, with occasional patches of the highly concentrated perchlorate reported here.

[22] These observations have constrained the cation associated with the perchlorate: only Mg- and Ca-perchlorate produce a $0.967\ \mu\text{m}$ feature, while K-, Na-, Fe^{3+} , and Fe^{2+} perchlorates do not. Interestingly, Mg- and Ca-perchlorates have significantly lower eutectic temperatures, increasing the chances to brine formation [e.g., Chevrier et al., 2009]. This work does not rule out the possibility of additional perchlorate phases at the Phoenix site.

[23] Previous studies have proposed that perchlorate on Mars might form from atmospheric interactions between ozone and volatile chlorine compounds as aerosols or at the surface, as in the Atacama Desert of Chile [Catling et al., 2010] and the Antarctic Dry Valleys [Kounaves et al., 2010a, 2010b]. Dissolution and redistribution of perchlorate could occur when summer mid-day temperatures exceed the perchlorate eutectic point. Chevrier et al. [2009] showed that Mg-perchlorate is metastable above 206K, while Phoenix surface temperatures rose up to 245K during the mission. However, the volume of water in the atmosphere during these times is small: the maximum observed water vapor in the atmospheric column was ~ 55 precipitable- μm [Tamppari et al., 2009]. An alternative redistribution mechanism involves seasonal ices. The Phoenix site is covered in early fall by an ~ 90 micrometer layer of seasonal H_2O ice, which is then topped in winter by a translucent slab of CO_2 ice that reaches $\sim 30\ \text{cm}$ thick [Cull et al., 2010]. At the base of the translucent CO_2 slab, a solid greenhouse effect can increase the temperature at the ice-surface interface, a process that Kieffer et al. [2000] proposed could be responsible for southern hemisphere “spider” features. We propose that this solid greenhouse effect can raise temperatures at the ice-surface interface high enough for small amounts of meltwater

or thin water films to form, dissolving surface perchlorate and transporting it downward through diffusive or gravity-driven fluid transport. Transport into the soil is limited by the rate of transport and the lower boundary of the subsurface ice table. This surface-to-subsurface redistribution process is common in the Antarctic Dry Valleys, where it concentrates soluble sulfates beneath soil clods and rocks. Perchlorate is thus removed from the surface and deposited as salt crusts in the shallow subsurface soil by thin films of water (greater volumes of liquid water are unlikely given the small amount of seasonal water ice involved and the low relative humidity/partial pressure of water in the atmosphere). This scenario implies a geologically recent occurrence of aqueous processes at the site.

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