

Unusual Sulfate Cave Mineral Deposits at Craters of the Moon National Monument, Idaho: Potential Analogue for Meridiani Planum, Mars

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Abstract

This study analyzes sulfate-rich precipitates in basaltic caves at Craters of the Moon National Monument (COM), Idaho. Basaltic lava tubes are likely common on Mars, and could preserve biosignatures from earlier, more habitable periods in Mars' history. COM basalts are good Mars analogues because of their unusually high iron concentrations and the hematite and sulfate minerals found in their weathering products. Their weathering over a short time in a volcanic environment could address the origins of the sulfate-rich sedimentary rocks and silica-rich rock coatings at the MER Opportunity landing site.

This project employed XRD, XRF, and SEM analyses of basalts and sulfate-rich precipitates collected from COM caves. Most mineral coatings analyzed from the walls and ceilings were calcite or silica-dominated, but mounds of Na-sulfate and Na-carbonate precipitates were present on the floors. While jarosite has been previously documented in COM caves, it was not observed during the present study.

Introduction

This study documents the geochemical and mineralogical effects of the weathering of high-iron basalts and sulfate mineral precipitation and at Craters of the Moon National Monument (COM), Idaho. COM basalts are similar in composition to Martian basalts, and their weathering over a short time in a volcanic environment could provide a test for one of the proposed origins for the sulfate-rich sedimentary rocks at the Mars Exploration Rover (MER) Opportunity landing site. A volcanic or groundwater origin for these sediments could help constrain the extent, duration, and necessity of liquid water during Mars' early history. As water is essential to life on Earth, its long-term presence on early Mars could indicate conditions favorable to the development of life. In contrast, a volcanic landscape with limited surface water would be less hospitable.

Objectives

The main objectives of this project are to: 1. Determine the compositions and mineral assemblages of high-Fe basalts altered in a volcanic and groundwater environment at COM. COM basalts are very Fe-rich by Earth standards (Kuntz, 1989), and are thus more comparable to Mars basalts than other previously studied analogues (e.g. Hawaii: Morris et al., 2000). 2. Compare the compositions and mineral assemblages of the altered COM basalts and associated precipitates to the rocks and soils analyzed by the MER Opportunity at Meridiani Planum. Similarities between the Mars and COM compositions and assemblages could suggest similarities in processes of formation. Sulfate minerals (including jarosite), hematite, and secondary silica have been identified in COM basaltic caves (Peck, 1974) and at Meridiani Planum, Mars (Klingelhöffer et al., 2004; Glotch et al., 2006).

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Background

Sulfate minerals and altered basalts on Mars. The Meridiani Planum landing site for the MER Opportunity was selected in part because of the presence of grey hematite (detected remotely), a mineral usually associated with aqueous deposition on Earth (Christensen et al., 2000). The initial reports from the rover supported an aqueous origin for the rocks. Among the lines of evidence for water are: (1) the abundance of sulfate minerals, including jarosite, (2) layered sandstone deposits, and (3) cross-bedding and ripple marks (e.g., Bell et al., 2003; Squyres et al., 2004a). McCollom and Hynek (2005) argued that these features are also consistent with volcanic deposition, and suggested an alternative model in which the “sediments” are actually volcanic surge deposits altered in-situ by sulfur-rich fumaroles. More detailed analyses of the sedimentological features of the landing site reveal bedding features consistent with aqueous deposition and not found in surge or eolian deposits on Earth (Squyres et al., 2006), but only in the upper half meter at Endurance Crater. The remaining sediments display primarily eolian features (Grotzinger et al., 2005). However, a volcanic origin for the sulfate-rich sediments themselves cannot be ruled out (Squyres et al., 2006a).

The rocks analyzed at Meridiani Planum consist of a mixture of siliciclastic material, sulfate minerals, and hematite (Squyres et al., 2004). The minerals jarosite and hematite were both detected by Mössbauer spectroscopy in the bedrock outcrops of Eagle Crater (Klingelhöffer et al., 2004). Hematite, sulfate minerals, and opaline silica were also detected at the light-toned outcrop at Meridiani Planum (Glotch et al., 2006) and at Paso Robles in Gusev Crater (MER Spirit, Ming et al., 2006). Hematite and sulfate minerals are normally formed at the Earth’s surface in the presence of liquid water (Hynek et al., 2002). Sulfate minerals are most often formed in evaporitic environments on Earth, but also form in volcanic regions by the interaction of basalts with sulfur-rich fumaroles (e.g., Morris et al., 2000) or groundwater (e.g., Peck, 1974). The evaporitic model is not well suited to the sulfate minerals formed at COM, or in other volcanically active areas on Earth.

The model preferred by the MER Opportunity rover team for the origin of the Meridiani Planum deposits involves, in chronological order: (1) the acid-sulfate diagenesis of basalt, (2) eolian transport and deposition of sulfate-rich sands derived from this altered basalt, and (3) the in-situ groundwater-related formation of hematite-rich spherules (Squyres et al., 2006a, b). Aqueous features formed when groundwater periodically reached the surface in low-lying areas. The proposed study provides an analogue for steps one and three: the sulfate alteration of Martian basalt, and the formation of hematite in a groundwater environment. Where and how the sulfate-rich altered basaltic sands of Meridiani Planum formed is unknown. One possibility is a dirty evaporite, where basalt was altered in contact with saline-acidic surface water in an evaporitic environment (Grotzinger et al., 2005). This model would require the presence of long-standing surface water. Another possible explanation is alteration through acid-sulfate weathering in a volcanic environment, as suggested by McCollom and Hynek (2005). Both models can be studied and tested by comparing the Mars deposits to those in similar environments on Earth.

Basaltic caves as Mars analogues. If basaltic volcanism on Mars resembles basaltic volcanism on Earth, then lava tubes must be (or must have been) ubiquitous features. The lower gravity on Mars could make lava tubes a more persistent feature on Mars since they would be less likely to collapse over time. In addition, recent imaging by the Mars Odyssey Thermal

Emission Imaging System (THEMIS) revealed several potential skylights into caves on Mars on and near Arsia Mons (Cushing et al., 2007). Because of their high altitude these imaged caves might make poor astrobiological targets, but other caves at lower altitudes may be great places to look for signs of ancient life. Launching an expedition into the Martian subsurface is far beyond our current technological capabilities. However if there was once life on Mars, its biosignatures would be more likely to be preserved in protective caves than at the surface, where they would be exposed to solar radiation and degraded over time.

Geologic setting: Craters of the Moon. Craters of the Moon lava caves were selected for this project because of their high-Fe basaltic compositions and the previously documented presence of sulfate minerals including jarosite (Peck et al., 1974). COM lava flows erupted during eight eruptive periods between 15,000 and 2100 years ago (Kuntz, 1989). Volcanism at COM appears to be related to the more regional volcanism associated with the nearby Snake River Plain, but with slightly more evolved basaltic compositions (high-Fe basalts, compared to higher-Mg basalts, Stout and Nicholls, 1977; Stout et al., 1994). COM lavas, especially the younger Blue Dragon flows (Figure 1), contain numerous lava tube caves. These caves provide a sheltered

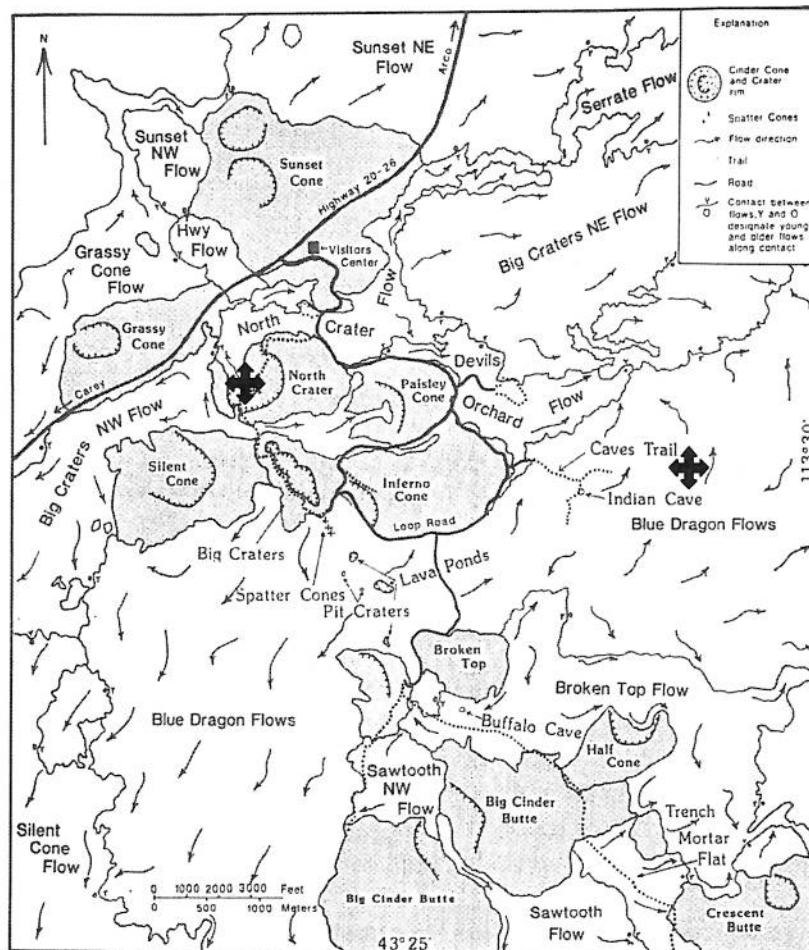


Figure 1: Map of COM lava field, from Kuntz 1989. The Blue Dragon (BD) and North Crater flows are indicated. Many of the 2008 samples came from Arco Tunnel in the BD flow, near the Caves Trail (labeled). Most 2007 samples are from Wilderness Caves, slightly off the map to the east in the BD flow.

environment for the formation and preservation of sulfate minerals. Reports indicate that Crystal Pit, a cave not accessed during this project, contains significant deposits of coarse-grained sulfate minerals including jarosite (Peck, 1974). Other sulfates, especially Na-sulfates such as thenardite and mirabilite, are present in caves throughout the park.

While the Fe content of CoM and Mars basalts are similar (see comparison in Table 1), it should be noted that they differ substantially in other potentially important elements. CoM basalts are higher in Al, P, Ti, Na, and K and lower in Mg, Ca, and S. Despite these differences, they are still closer than other frequently used Mars analogues (e.g. Hawaii, Morris et al. 2000) that suffer from the same limitations and also have lower Fe.

Table 1: comparison of Mars, COM, and Hawaii basalt compositions.

Oxide	Mars basalts				COM flows, this study		Hawaii
	Sherg ^{1*}	Spirit ²	Bounce ³	Meridiani ⁴	BD ⁵	NC ⁵	HI ⁶
SiO ₂	50.10	45.40	50.80	38.10	47.79	48.74	49.74
TiO ₂	0.80	0.45	0.78	0.85	2.95	2.74	2.77
Al ₂ O ₃	6.70	10.90	10.10	6.00	12.51	12.70	17.35
Fe₂O₃T	21.00	20.00	17.30	19.60	17.12	16.89	12.03
MnO	0.50	0.38	0.43	0.32	0.21	0.27	0.21
MgO	9.10	11.90	6.40	7.40	3.32	3.05	3.93
CaO	9.40	7.42	12.50	4.50	7.04	6.72	6.60
Na ₂ O	1.40	2.70	1.30	1.10	3.14	2.90	4.33
K ₂ O	0.20	0.06	0.10	0.56	1.89	2.08	1.90
P ₂ O ₅	0.70	0.54	0.95	1.00		1.78	0.85
Cr ₂ O ₃	0.20	0.59	0.12	0.22	-	-	-
Cl	0.20	0.13	0.06	0.39	-	-	-
SO ₃	0.48	1.15	0.52	21.00	0.10*	-	0.09
Total	100.00	-	-	-	98.28	97.87	99.71

* calculated as volatile-free

¹ Shergotty meteorite, as reported in Lodders (1998)

² MER Spirit analysis of Adirondack (RAT). As reported in Gellert et al., 2004.

³ MER Opportunity analysis of Bounce Rock (RAT). As reported in Rieder et al., 2004.

⁴ MER Opportunity analysis of McKittrick (RAT). As reported in Rieder et al., 2004.

⁵ Blue Dragon (BD) and North Crater (NC) flows, Craters of the Moon, this study.

⁶ Average unaltered Hawaiian tephra from Mauna Kea, as reported in Morris et al., 2000.

Methods

Samples of mineral coatings from the ceilings, walls, and floors of basaltic caves from COM, as well as associated lavas, were collected in October 2007 and June 2008. All caves sampled and analyzed for this project are in the Blue Dragon flows (Figure 1), a series of recent (~2100 yr), extensive high-Fe lava flows. Samples from the following categories were selected for analysis: hard, white ceiling coatings, brown ceiling coatings, yellow coatings, dendritic mineral deposits, powdery or snow-like mounds on cave floors, and associated basalts (Figure 2).

Thin coatings were separated from the substrate and powdered using a tungsten carbide dental drill, and then finely powdered using a mortar and pestle. The fine powders were then mounted in a cavity mount and gently flattened for X-ray Diffraction (XRD) analysis. Samples were analyzed using a Bruker D8 Focus system (Cu K α radiation, 4 s per 0.01° 2 θ , 2° to 60° range,

Sol-X energy dispersive detector). Peak identification was conducted by comparison to the ICDD database using Bruker's EVA software package. Relative proportions of each mineral were determined qualitatively by comparing peak heights and quantitatively where possible using Bruker's TOPAS Reitveld refinement software package. This was not possible for samples containing some of the more unusual minerals identified (such as burkeite), as these minerals were not in the structure database. Samples 07-CM 1 to 5 and 7, 08-CM3, 5, 6, 7, 10, and 11 were analyzed by XRD.

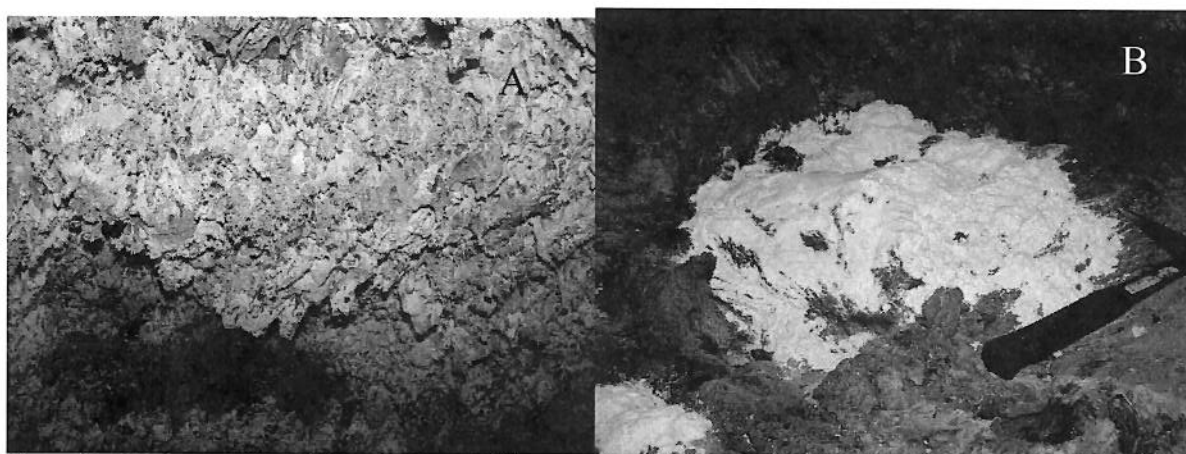


Figure 2: COM cave mineral deposits from Wilderness Caves, Blue Dragon Flows. A: white (07-T1) and dendritic (07-T2) ceiling coatings. B: Na-sulfate and carbonate mound on floor (08-CM8).

Chips of mineral coatings and their associated substrates were also prepared for Scanning Electron Microscopy (SEM) for the 2007 samples. These were mounted in epoxy, polished, carbon coated, and then analyzed using a Hitachi S-3400N variable pressure SEM at the University of Wisconsin-Madison. The system was operated in vacuum at 15 kV. Backscattered electron images (BSE) and Energy Dispersive Spectra (EDS) were collected for thirteen cave ceiling precipitate samples.

Select samples (07-CM1 to 5, 7) were also prepared for analysis by X-ray Fluorescence (XRF) to determine their bulk chemical compositions. One gram of finely ground sample powder was combined with 10g of flux (50:50 LiT:LiM with integrated LiBr non-wetting agent) and ~1g of an oxidizer (ammonium nitrate). The mixture was then fused in a platinum crucible in a Claisse M4 fluxer using a predetermined 21-minute fusion routine (high temperature ~1050° C). The prepared beads were then analyzed using a Bruker S4 Pioneer WD-XRF. The instrument was calibrated for major and minor elements using 11 USGS rock standards. Sulfur was determined semiquantitatively using Bruker's standardless analytical routine.

Results

Sulfate minerals were surprisingly rare among the surface coatings, which are instead dominated by calcite. Abundant sulfate minerals are limited to deposits collected from the cave floors (samples 07-CM4 and 08-CM7, 10, and 11). Where present, the sulfates were dominated by Na-sulfate minerals (thenardite, mirabilite, burkeite) associated with Na-carbonate and bicarbonate phases (trona, natrite, thermonatrite).

The XRD-determined mineralogical results are summarized in Table 2. The hard, white ceiling coatings and the dendritic samples are dominated by calcite with only minor contaminants. The brown ceiling coatings contained abundant hematite with or without associated quartz (e.g. 07-CM3, Figure 3). The XRD pattern of the yellow mineral deposit (08-CM5) contains peaks that could indicate jarosite, but at concentrations likely under 1%. It instead contains abundant hematite and likely derives its color from goethite. One of the powdery samples from the floor mounds was mostly calcite (08-CM3), but the other four analyzed were dominated by Na-sulfate and/or Na-carbonate minerals (e.g., 07-CM4, Figure 4). Non-Na sulfate minerals were not observed in significant amounts in any sample.

The SEM-EDS results confirmed the dominance of calcite and a high-Si phase in ten additional samples of the ceiling coatings. Sulfur was not detected in the EDS spectra of any of the ceiling coating samples.

Table 2: Mineral assemblages as determined by XRD

		Calcite	Quartz	Hematite	Na- carbonate	Na- sulfate	Basalt minerals	Goethite
Hard, white ceiling coatings								
07-CM1		XXX	--	+	--	--	+	--
07-CM7		XXX	--	--	+	-	-	--
Dendritic minerals on ceilings								
07-CM2		XXX	--	--	--	--	+	--
Brown ceiling coatings								
07-CM3		--	XX	XX	--	+	XX	--
08-CM6		--	--	XX	--	--	XX	--
Yellow mineralization								
08-CM5		--	--	XX	--	--	XX	X
Powdery or snowy white floor deposits								
07-CM4		--	--	--	XXX	X	--	--
08-CM3		XXX	+	--	--	--	--	--
08-CM7		--	--	--	--	XXX	+	--
08-CM10		--	--	--	XX	XX	+	--
08-CM11		--	--	--	XX	XX	--	--
Basalt								
07-CM5		--	--	--	--	--	XXX	--

XXX = abundant, XX = common, X = between common and rare, + = rare, -- = not detected.

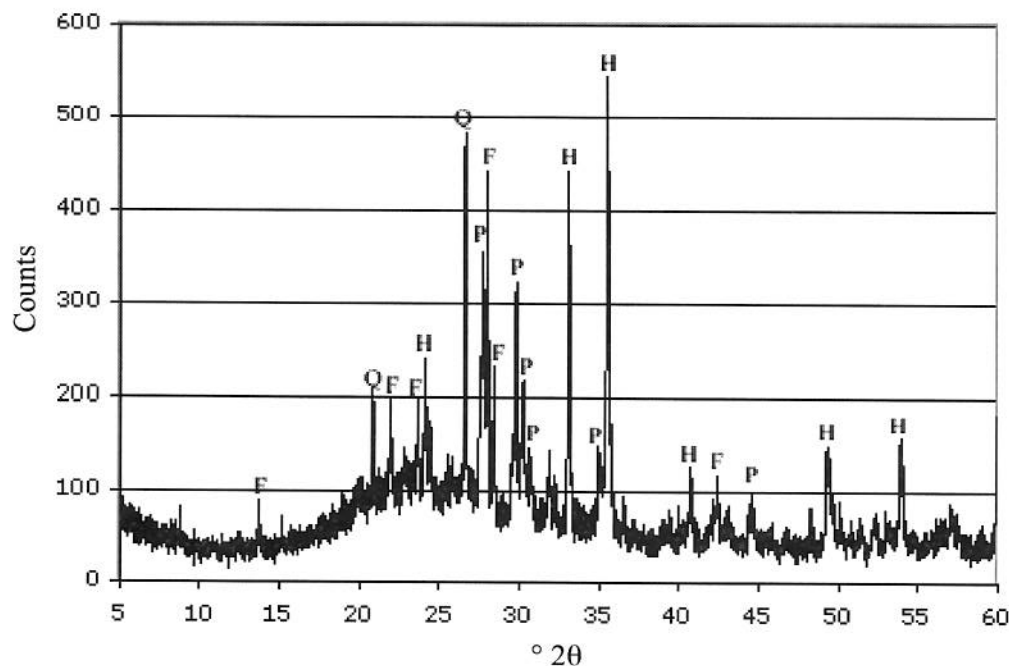


Figure 3: XRD pattern for brown ceiling coating sample 07-CM3. F = feldspar, Q = quartz, H = hematite, and P = pyroxene. Note the dominance of hematite and quartz, overprinting the basalt's feldspar and pyroxene.

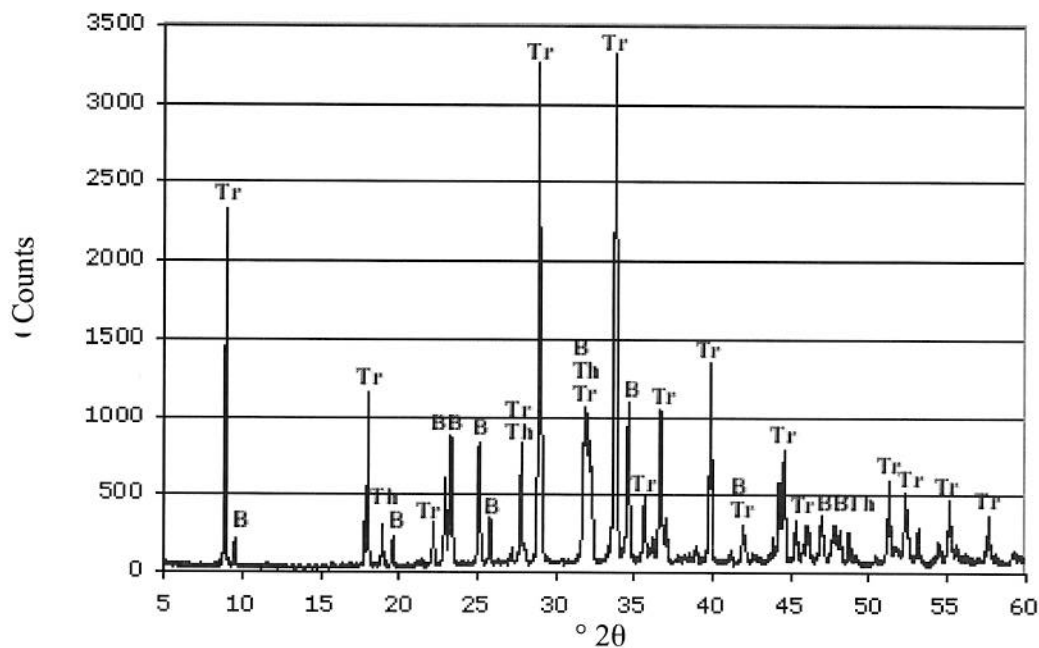


Figure 4: XRD pattern for Na-carbonate/ Na-sulfate mound sample 07-CM4. Tr = trona ($\text{Na}_3\text{H}(\text{CO}_3)_2 \cdot 2\text{H}_2\text{O}$), B = burkeite ($\text{Na}_6(\text{CO}_3)(\text{SO}_4)_2$), and Th = thenardite (Na_2SO_4). Note the complete absence of basalt minerals.

Geochemically, the COM basalts analyzed (from North Crater and the Blue Dragon flows, Table 1) were high in Fe as expected. The precipitates were of variable compositions (Table 3), likely reflecting both the composition of the precipitates and the amount of basalt substrate inadvertently incorporated in the samples of the thin coatings during preparation. As expected,

the calcite-dominated samples (07-CM1, 2, and 7) have higher CaO concentrations than the underlying Blue Dragon basalt. The hematite-bearing sample 07-CM3 does not have elevated Fe₂O₃ relative to the basalt, suggesting that the hematite was derived through in-situ alteration of the Fe-bearing phases present in the basalt substrate. This sample also lacks olivine, an abundant mineral in COM lavas. Na is the only significant cation in the mound sample 07-CM4.

Table 3: Bulk geochemical analysis of coatings, as measured by XRF.

	Mineral coatings				Mound	Fresh BD
	White		Dendritic	Brown		
Oxide	07-CM1	07-CM7	07-CM2	07-CM3	07-CM4	07-CM5
SiO ₂	64.27	34.04	59.44	53.61	1.60	47.79
TiO ₂	0.37	0.07	0.10	2.35	0.01	2.95
Al ₂ O ₃	2.12	0.54	0.73	11.12	0.25	12.51
Fe ₂ O ₃ T	2.59	0.53	0.77	14.34	0.10	17.12
MnO	0.03	0.01	0.02	0.18	LLD	0.21
MgO	2.64	1.60	1.93	2.73	0.26	3.32
CaO	12.92	38.31	18.48	5.82	0.07	7.04
Na ₂ O	1.03	0.25	0.22	3.83	55.31	3.14
K ₂ O	0.49	0.07	0.09	2.21	0.05	1.89
SO ₃ *	0.46	0.79	0.52	0.14	16.98	0.10
Total	87.11	76.53	82.47	98.04	57.99	98.28

* Semiquantitative SO₃ concentrations

Discussion

The sulfate minerals of Craters of the Moon do not appear to be direct alteration products of the weathering basalts. The contact between the authigenic minerals and the basalt substrate is usually abrupt, and basaltic minerals (plagioclase, diopside, olivine) are not found intermixed with the sulfate minerals. This suggests that the sulfate minerals formed as precipitates rather than through replacement. The fact that the sulfates are concentrated in mounds on the floors of the caves could suggest a biotic origin for the deposits- either through microbial activity or the alteration of bat guano. The lack of associated gypsum and phosphate minerals or visible guano deposits makes the latter possibility less likely, as these are the most common minerals formed through the process of bat guano leaching (Hill and Forti, 1997).

The sulfate minerals observed are exclusively Na-sulfates and are often associated with Na-carbonates. CoM basalts are concentrated in Na relative to most basalts (and certainly compared to Mars basalts) and thus leaching could be a source for the Na cations as it is for N-sulfates in other basaltic cave settings (Hill and Forti, 1997). The source of the sulfur is more questionable since the caves are overlain entirely by COM basalts that are not especially rich in this element. A potential sulfur source could be volcanic gases, though COM basaltic field has not had any eruptions in 2100 years. Another potential source is, again, bat guano.

The sulfate and Na-carbonate deposits appear to be a seasonal feature. The Wilderness Caves were visited in both October 2007 and June 2008, and there were significant differences in the observed patches of sulfates. In October, few patches were observed and those that were present

were soft, powdery, and dominated by trona (sample 07-CM4). The same cave visited in June had more and larger patches of a more solid consistency, and contained a greater proportion of Na-sulfate minerals (08-CM10, 11). Apparently these sulfate mounds appear and disappear on an annual basis, dissolving away during wet times and reprecipitating when dry. Temperature also plays a factor- mirabilite, a low-temperature Na-sulfate, was present (along with thenardite, a higher-temperature Na-sulfate) in a sample collected in June (08-CM7) but not in October, when thenardite was present without mirabilite. June is earlier in the season and ice still persists in the deeper parts of the caves, while all of the ice had melted by October.

The presence of these deposits in patches on the floors of lava tubes, and not at the deepest spots, suggests that they are not formed directly as precipitates from liquid water. Standing pools of water would have left more uniform deposits at the lowest spots. The ceiling deposits, including the silica and calcite, formed from dripping water as do stalactites in limestone caves.

Comparison to Mars mineralogy and geochemistry. The closest analogue on Mars to the COM mineral precipitates is probably the light-toned outcrop at Meridiani Planum reported by Glotch et al. (2006). Based on data from the MER Mössbauer spectrometer, Mini-TES, and APXS instruments, this outcrop is dominated by opaline silica, Mg-, Ca-, and Fe-bearing sulfate minerals, hematite, and plagioclase. The coexistence of hematite, silica, and plagioclase is reminiscent of the brown ceiling coating sample 07-CM3. At COM, this mineral assemblage likely formed through alteration of high-Fe basalts in contact with seeping groundwater. The Fe-rich olivine typical of COM basalts has altered to hematite, while the plagioclase remains. This mechanism could also be possible at Meridiani Planum on Mars, where olivine is also preferentially destroyed during weathering (Glotch et al., 2006).

However, the sodium-rich carbonate and sulfate minerals observed at COM do not appear to have direct analogues on Mars. This could be related to a difference in environment, as thenardite and burkeite are typical evaporite minerals from saline-alkaline lakes (Spencer, 2000) while Mg and Fe-sulfates (such as melanterite and jarosite) are more typical of acidic environments (Hill and Forti, 1997; Ptacek and Blowes, 2000). This difference is unlikely related to differences in the availability of water, as both jarosite and thenardite are highly soluble and indicate arid conditions. Thenardite has a solubility between the Mg-sulfate minerals epsomite and kieserite (Spencer, 2000), two minerals observed at Gusev Crater on Mars (Peterson et al., 2007). The presence in the COM cave precipitates of carbonate minerals, including calcite and trona, is also unlike Mars.

Conclusions

The precipitates on the lava tube ceilings at COM are dominated by calcite and silica. Na-sulfate and Na-carbonate deposits are limited to mounds on the cave floors, and other sulfate minerals are absent or very rare. Thus, the sulfate deposits of COM are not analogous to the Mg-, Fe-, and Ca-bearing sulfates observed at Meridiani Planum and Gusev Crater on Mars.

However, the hematite and silica-rich weathering rinds on some of the basalt stalactites are potentially analogous to the light-toned outcrop at Meridiani Planum, Mars. These likely formed by the alteration of olivine in basalt in contact with seeping groundwater, a mechanism that has also been suggested for the Mars deposits.

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