

# Contrasting styles of water–rock interaction at the Mars Exploration Rover landing sites

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## Abstract

The nature of ancient hydrological systems on Mars has been the subject of ongoing controversy, driven largely by a disconnect between observational evidence for flowing water on the Martian surface at multiple scales and the incompatibility of such observations with theoretical models that predict a cold early Martian environment in which liquid water is unstable. Here we present geochemical data from the Mars Exploration Rovers to evaluate the hydrological conditions under which weathering rinds, soils, and sedimentary rocks were formed. Our analysis indicates that the chemistry of rinds and soils document a water-limited hydrologic environment where small quantities of S-bearing fluids enter the system, interact with and chemically alter rock and soil, and precipitate secondary mineral phases at the site of alteration with little to no physical separation of primary and secondary mineral phases. In contrast, results show that the sedimentary rocks of the Burns Formation at Meridiani Planum have a chemical composition well-described as a mixture between siliciclastic sediment and sulfate-bearing salts derived from the evaporation of groundwater. We hypothesize that the former may be derived from the recently investigated Shoemaker Formation, a sequence of impact breccias that underlie the Burns Formation. This result has important implications for the style of chemical weathering and hydrology recorded by these sedimentary materials, revealing long-range transport of ions in solution in an open hydrological system that is consistent only with subsurface or overland flow of liquid water.

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## 1. INTRODUCTION

The Martian geologic record contains a wide variety of geomorphological features consistent with flowing and standing water on the surface (e.g., outflow channels, valley networks, alluvial fans and deltas, sediment-filled craters and basins), with the generation of water-related landforms generally accepted to have been more prevalent in early Martian history and waning towards the modern day (for recent reviews, see Carr, 2012; Grotzinger and Milliken, 2012). The discovery of a broad suite of aqueous secondary minerals produced from alteration of the Martian crust by fluids has added further constraints on the nature of

water–rock interactions. Syntheses of these mineralogical observations from orbital data have driven a hypothesis for fundamental secular changes in secondary mineral forming processes over time, from early clay-mineral forming environments to intermediate sulfate-mineral forming environments to more modern environments in which anhydrous ferric iron oxides are produced (Bibring et al., 2005; Mustard et al., 2008; Murchie et al., 2009; Ehlmann et al., 2011). These geomorphic and mineralogical observations are broadly thought to reflect a change in environmental conditions at the Martian surface from an early wet (and possibly warm) state to the pervasive dry, cold conditions observed today.

Despite geomorphological, sedimentological, and mineralogical evidence for the presence of liquid water at the Martian surface, at least in its early geologic history, significant questions remain regarding the state and evolution of the Martian atmosphere (e.g., Greenwood et al.,

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2008; Cassata et al., 2012), specifically whether it was sufficiently thick and had an adequate greenhouse budget for liquid water to have been stable at the surface of Mars. A number of models have been proposed that can provide the warm, clement conditions required for liquid water stability (e.g., Forget and Pierrehumbert, 1997; Sagan and Chyba, 1997; Halevy et al., 2007), though none of these models have garnered universal acceptance (for a recent review see Niles et al., 2013). The apparent inconsistency between observations and theory has led to suggestions that Mars may only have experienced transient warm climate episodes driven by volcanism (e.g., Johnson et al., 2008, 2009) or large impact events (Segura et al., 2002). Others have questioned the sedimentary interpretations for liquid water (e.g., Gaidos and Marion, 2003; Niles and Michalski, 2009; Kite et al., 2013), hypothesizing instead that widespread sedimentary deposits were produced in a cold, polar-type climate for much of Martian geologic history. In these conceptual models, the previously described geomorphic and mineralogic features are largely generated by processes such as groundwater escape from beneath a global cryosphere, melting of snowpacks, or glacial melting, as examples.

The ancient sedimentary rocks of the Burns Formation at Meridiani Planum, Mars have recently become a touchstone in the debate over whether climate conditions were ever appropriate to allow for stable liquid water in surface environments at the time these deposits were formed (ca. 3.5–3.7 Ga). Based on extensive *in situ* observations by the Mars Exploration Rover (MER) *Opportunity*, the sedimentary rocks of the Burns Formation were interpreted to have formed under dry, desert-like conditions in which chemically weathered basaltic sediment was transported in an aeolian environment and cemented by evaporating, sulfate-rich groundwater (Grotzinger et al., 2005; Squyres and Knoll, 2005). This interpretation is consistent with models of global groundwater transport that indicate that Meridiani Planum was a locus for groundwater upwelling and evaporation, requiring a climate system that would have supported meteoric recharge to drive groundwater transport (Andrews-Hanna et al., 2007, 2010 although see Michalski et al., 2013). If this interpretation is correct, then the Burns Formation places an important constraint on Martian climate conditions, and indicates that during late Noachian to early Hesperian time, the climate was sufficiently warm and wet to allow for the existence of at least a limited hydrologic cycle in which aquifers could be recharged by rainfall and/or snowmelt.

The Burns Formation is characterized by enigmatic chemistry and mineralogy: its bulk composition has been broadly described as a mixture of “basalt plus sulfur”, and it contains as a ubiquitous mineral component the mineral jarosite ( $\text{K, Na, H}_3\text{O}^+\text{Fe}_3^{3+}(\text{SO}_4)_2(\text{OH})_6$ , which requires acidic (pH = ca. 2–4) conditions to precipitate. This chemistry is somewhat surprising given the process hydrology interpretation of the deposit that implies aquifer recharge and subsequent groundwater upwelling and evaporation. In such a setting, elemental fractionation along the flow path due to differential mineral solubility and the generation of alkalinity from silicate weathering might reasonably be expected. Highlighting the apparent inconsistency between the charac-

teristics of the Burns Formation and an open hydrologic system, a number of alternative hypotheses have been advanced that interpret the Burns Formation as the product of sulfuric acid-driven weathering in volcanoclastic (McCollom and Hynke, 2005), impact (Knauth et al., 2005), or glacial ice (Niles and Michalski, 2009) environments.

These alternative hypotheses, driven by the recognition of a “basalt plus sulfur” bulk chemical composition; invoke chemical weathering processes described as “closed system”, “isochemical”, or “cation-conservative”. What is meant by these terms from a process perspective is: sulfuric acid-charged water is added to basalt, a suite of secondary minerals is generated, the water leaves the system in the gas phase by vaporization or sublimation (and/or is partially retained in hydrated secondary minerals), and left behind is a mixture of newly formed secondary minerals and residue that have not been physically separated from each other. The term “cation-conservative” (Niles and Michalski, 2009) is the most accurate for the hydrochemical conditions described above, which are neither truly “closed” nor truly “isochemical”; accordingly, we employ the term “cation-conservative” here.

“Acid fog” hypotheses also describe a form of cation-conservative chemical weathering that yield basalt plus sulfur chemistry, and are often invoked to explain the chemical composition of soils on Mars. In this model, acidic volcanic aerosols settle out of the atmosphere onto rock, soil, and dust, altering the surfaces they come into contact with through low water-to-rock ratio, low pH weathering (e.g., Settle, 1979; Clark and Van Hart, 1981; Banin et al., 1997; King and McSween, 2005; Hurowitz et al., 2006b; Ming et al., 2008; King and McLennan, 2010). Similar mechanics could arise from impact processes in which the decomposition of sulfate minerals generates “recycled” sulfuric acid to drive silicate weathering, similar to what is thought to have occurred in the terrestrial Chixulub impact event (Kring, 2007; Zolotov and Mironenko, 2007; King and McLennan, 2010). The weathering process associated with the acid fog model is similar to the hypothesis proposed by McCollom and Hynke (2005), and more recently by Michalski and Bleacher (2013), who suggested that the chemistry of the Burns Formation was generated by the co-deposition of acid volatiles and basalt in volcanoclastic base surge deposits.

Building on analogues in terrestrial polar environments, Niles and Michalski (2009) proposed that Burns Formation sediment was generated during alteration in small pockets of meltwater hosted in massive ice sheets in the equatorial latitudes of Mars (including over Meridiani Planum). In these melt pockets, low temperature and low water-to-rock ratio chemical weathering occurred between co-deposited basaltic dust and volcanic acid aerosols trapped in the ice as it accumulated. Later sublimation of the ice left behind altered sediments with a basalt plus sulfur bulk composition.

In this report we present evidence from rover geochemical data that the sedimentary record of Mars preserves evidence for both cation-conservative weathering processes and open system weathering conditions in which there is sufficient liquid water present to mobilize and transport cations in solution over more significant length scales. The geochemistry of modern soils and weathered rock surfaces

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