

CO<sub>2</sub> and CH<sub>4</sub> have been suggested as important greenhouse gases on the early Earth. Our research focuses on the effects of increased concentrations of atmospheric greenhouse gases on the surface temperature of early Mars, with emphasis on the reduced greenhouse gas, CH<sub>4</sub>. To investigate the possible warming effect of CH<sub>4</sub>, we modified a one-dimensional, radiative-convective climate model used in previous studies of the early martian climate (5). New cloud-free temperature profiles for various surface pressures and CH<sub>4</sub> mixing ratios will be presented. This use of climate modeling is important since it is the fundamental way that the magnitude of possible geochemical and biological CH<sub>4</sub> sources can be related to predicted CH<sub>4</sub> concentrations in the early martian atmosphere.

References: 1) Gough, D. O. *Solar Physics* 74, 21-34 (1981). 2) Carr, M. H. *Water on Mars* (1996). 3) Kasting, J. F. *Icarus* 94, 1-13 (1991). 4) Forget, F., and Pierrehumbert R. T. *Science* 278, 1273-1276 (1997). 5) Mischna, M. A., Kasting J. F., Pavlov A., and Freedman R. *Icarus* 145, 546-554 (2000).

## P52B MCC: 270 Friday 1330h

### Stable Isotopes and the Search for Life in the Solar System I (joint with B, V)

**Presiding:** J P Greenwood, Yale University; L Leshin, Arizona State University

## P52B-01 1330h INVITED

### Vapor Pressure Isotope Effects and the Stable Isotope Geochemistry of the Martian Surface

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The stable isotope geochemistry of light elements (H, C, N, O and S) is a tool in the search for evidence of life on bodies other than the earth for several reasons: the elements in question are used by all known or easily imagined life forms; several mass spectrometric and spectroscopic methods for measuring their isotopic compositions exist and are appropriate for in situ analysis on other planets; it is known that 'vital effects' generate large (percent) isotopic fractionations; and studies of extant and former life on earth provide models for interpreting such data. However, the evidence stable isotope geochemistry provides is generally interpretable as a signature for life only in the context of a rich understanding of isotopic variations produced by non biological processes in the same environments.

Several candidates in the search for extra-terrestrial life (Mars and the Jovian satellites) are bodies having volatile-element geochemical cycles that operate at lower temperatures than the earth's surface and involve phase-changes that do not occur on the earth (e.g., condensation/sublimation of CO<sub>2</sub>). We review new experimental data (both recently published and unpublished) describing isotopic fractionations accompanying phase changes of CO<sub>2</sub>, H<sub>2</sub>O below 240 K, NH<sub>3</sub> and CO, compile them with previous data to derive general principles of low-temperature stable isotope fractionations, and discuss their significance for the stable isotope geochemistry of the surface of Mars. One example of the issues constrained by these data (the Martian CO<sub>2</sub> cycle) is discussed here.

The Martian surface is strongly influenced by condensation/sublimation and adsorption/desorption of CO<sub>2</sub>. The isotopic consequences of these processes are an attractive goal for in situ observations of atmosphere, ice and/or adsorbed gas because they provide a record of Martian atmospheric dynamics and atmosphere-surface interactions over a range of time-scales. Furthermore, these isotopic signals naturally must be understood before one could confidently identify carbon or oxygen isotope evidence for life. Recent experiments indicate that both ice/vapor and adsorbate/vapor phase changes produce oxygen isotope fractionations of the same direction and magnitude as those for condensation/evaporation of water vapor in the earth's atmosphere. This suggests that seasonal and longer-term cycles of condensation and release of CO<sub>2</sub> produce variations of order 10 ‰ in δ<sup>18</sup>O. The current seasonal cycle of ice-cap formation and sublimation likely produces a similar-magnitude latitudinal gradient in the δ<sup>18</sup>O of CO<sub>2</sub>. This gradient must be sensitive to mixing between high- and low-latitude air and therefore provides a constraint on martian atmospheric circulation. In contrast, carbon isotopes of CO<sub>2</sub> are not measurably fractionated by condensation/sublimation of ice and should be invariant in a martian atmosphere in which this is the only major phase change. Adsorption significantly (ca. 1 to 2 ‰) concentrates <sup>13</sup>CO<sub>2</sub> into the vapor phase and <sup>12</sup>CO<sub>2</sub> into adsorbate. This is opposite in direction to all familiar vapor-pressure isotope effects. Thus, cycles of adsorption/desorption of CO<sub>2</sub>

have a unique isotopic signature that should be distinguishable for other processes influencing the Martian atmosphere. Measurements of the variation in δ<sup>13</sup>C of CO<sub>2</sub> would constrain the extent to which the atmosphere and surface interact by adsorption/desorption processes.

## P52B-02 1345h INVITED

### Organic and Isotopic Signatures of Life: Lessons from the Early Earth

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In the study of life on earth, isotopic analyses of organic biomarkers provide essential insight to their biological and environmental provenance. Isotopic analyses of organic materials on other planets present a number of challenges, both analytical and interpretive. Prebiotic planetary organic materials can derive from condensation reactions and by delivery through meteorites or interplanetary dust, with the relative importance of each influenced by the oxidation state of the atmosphere. Material delivered to planets can have an interstellar origin, although it is dominated by compounds influenced by the formation of the solar system. Each of these processes impact molecular isotopic signatures and must be considered in life-detection strategies. Pronounced effects are observed for hydrogen isotopes, with smaller fractionations observed for other elements. Theoretical, laboratory and observational studies of non-terrestrial materials are essential to further understand molecular isotopic heterogeneity associated with these exclusively abiotic processes.

Studies of Archean-aged samples provide an important resource for interpreting molecular isotopic patterns as signatures of life processes. Carbon assimilation and biomass synthesis from simple precursor compounds typically discriminate against <sup>13</sup>C. This generality, however, is complicated by the observations of a wide range of fractionation factors associated with important microbial carbon-uptake processes. Metabolic processes further distribute isotopic signatures, such that wide isotopic heterogeneity is observed among cellular biochemical constituents. In addition, preservation/contamination concerns dominate studies of very ancient organic matter, as they likely will in life-detection studies. However, both biochemical heterogeneity and sample integrity can be addressed by considering patterns from different paleoenvironments. Molecular results demonstrate that Late Archean microbial life on this planet was diverse, and ecological controls on element cycling dominate interpretations of molecular isotopic signatures. Is ecology universal? The answer may be the key to understanding life signatures elsewhere.

## P52B-03 1400h INVITED

### Pre-3.5 Ga terrestrial sediments as test cases in the search for life in the solar system

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The general approach to understanding the early development of life on Earth has been to establish the antiquity of rocks by identifying radiometrically dateable sequences containing morphologically classifiable fossils (morphofossils). While this approach has served us well for nearly a century, classical micropaleontological methods used to interpret the Proterozoic (2.5 - 0.54 Ga) record of life are unsatisfactory in studying the early Archean (>3.2 Ga) record, which has been obscured by metamorphism. To gather insights into earlier traces of life, we must see past the limitations of the morphofossil record and recognize the value of chemical fossils. One such approach has been to utilize the strong fractionation that metabolic activity of organisms imparts to light stable isotope ratios (<sup>13</sup>C/<sup>12</sup>C, <sup>15</sup>N/<sup>14</sup>N, <sup>34</sup>S/<sup>32</sup>S). The importance of such searches is that they provide a natural test bed for planning the kinds of analyses to be performed on samples returned from elsewhere, even if such searches have had mixed success in ancient terrestrial rocks. For example, the atmosphere, carbon products of mantle degassing and carbonate in water on Earth define the inorganic pool of carbon from which bioorganic carbon is isotopically fractionated. Mass balance calculations demonstrate that the average isotopic composition of terrestrial carbon is ~ δ<sup>13</sup>C = -6 ‰ and therefore typical metabolic fractionations from this starting value results in δ<sup>13</sup>C (biomass) < -27 ‰. However, problems arise

when applying the assumptions based on the terrestrial chemofossil record to another planet. Mars is the strongest candidate for a second planetary biosphere in the solar system. If an ancient biosphere did exist on Mars, returned samples might be expected to yield data that challenge many assumptions about what constitutes an isotopic biosignature. Mars appears to be different from the Earth. The isotopic values for the various reservoirs of carbon, nitrogen and sulfur on Mars have been extrapolated from the study of martian meteorites and from remote spectroscopic measurements of the atmosphere. Carbonate carbon in martian meteorites has values that range at least between δ<sup>13</sup>C = +7 to +42 ‰, which are interpreted to represent the isotopic values of carbonate formed in equilibrium with isotopically heavy (~ +40 ‰) CO<sub>2</sub> in the Mars atmosphere and carbon released from martian meteorites at the high temperature stage of step-heating experiments (above 700 C) has δ<sup>13</sup>C values between -30 and -15 ‰, which may represent magmatic values. Depending on the different starting values of the inorganic reservoirs of carbon on Mars described above, possible martian biotic carbon residues would be expected to be either isotopically heavier on average (δ<sup>13</sup>C > +5 ‰) or much lighter (δ<sup>13</sup>C < -40 ‰) than terrestrial life (average δ<sup>13</sup>C = -40 ‰ in the early Archean). Interpreting isotopic values for N and S on Mars present their own unique difficulties. Furthermore, all of these values may have changed over time as the martian atmosphere evolved, which behaves us to link the age of a sample and its geologic context with a specific isotopic signal as we would do with ancient terranes on the Earth. A silver-lining to all of this is that exploring the above possibilities might have the added benefit of differentiating between terrestrial contamination of a returned sample of Mars and the true remains of past martian biota.

## P52B-04 1415h INVITED

### Strategies for Life Detection Using Sulfur Isotopes

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Accurate evaluation of sulfur isotope fractionations as tracers of biological activity requires knowledge of: (1) the isotopic composition of reservoirs of inorganic sulfur species (e.g., sulfide, sulfate, elemental sulfur); (2) competing mechanisms that may produce sulfur isotope variations of non-biological origin; and (3) any isotopic signatures that can be uniquely linked to specific biological processes. We show how consideration of these three characteristics in terms of multiple sulfur isotopes may validate claims that certain sulfur isotopic signatures are biological.

For the most part, studies that have focused on the biological implications of variations in sulfur isotope compositions have only considered the quantity δ<sup>34</sup>S. We propose that it is critical to take into account δ<sup>33</sup>S variations as well. The consideration of multiple sulfur isotopes may help identify the sources of sulfur used by organisms because certain sulfur reservoirs possess anomalous and characteristic Δ<sup>33</sup>S compositions (Δ<sup>33</sup>S expresses the deviation from a mass fractionation array through δ<sup>33</sup>S = 0 and δ<sup>34</sup>S = 0 and is approximated by δ<sup>33</sup>S<sub>measured</sub> - 0.515 × δ<sup>34</sup>S<sub>measured</sub>). Measurement of δ<sup>33</sup>S adds an additional mass-balance constraint on an organism's metabolic reaction network and may identify a particular pathway for sulfur through this network. Most importantly, the observation of systematic Δ<sup>33</sup>S variations may produce definitive evidence for or against specific biological fractionation processes. The latter of these is possible because of small but measurable differences in mass dependent fractionation arrays that are associated with different fractionation processes. For example, it has been shown that the kinetic fractionation attending certain biological processes imparts Δ<sup>33</sup>S signatures that are distinct from those produced by abiotic processes. Although further experimental study will be required to determine whether similar Δ<sup>33</sup>S differences are characteristic of all biological fractionation processes, many biological processes have kinetic components and, accordingly, should produce a unique, case-specific fractionation of multiple sulfur isotopes.

## P52B-05 1430h INVITED

### OXYGEN ISOTOPE BIOMARKERS IN THE SEARCH FOR EXTRATERRESTRIAL LIFE

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Of the five key light stable isotope biomarkers, C, H, O, N and S, oxygen has received relatively limited application to the search for life and previous oxygen isotope studies of extraterrestrial materials have often focused on oxygen in carbonates and silicates. This is due, in part, to the limited study and development of other oxygen isotope ratio systems relevant to biological activity (e.g., phosphate, sulfate, nitrate) specifically for application to the search for extraterrestrial life. An overview of oxygen isotope biomarker systematics will be presented with emphasis on development of O isotope ratios of phosphate as a new biomarker in the search for life on Mars and Europa. Phosphate is central to life on Earth and has been widely recognized and applied as a biomarker molecule in the form of phosphate minerals, in studies of ancient life on Earth. The lack of multiple stable isotopes of P precludes direct stable isotope ratio studies of P, however, its occurrence as predominantly orthophosphate, PO<sub>4</sub>, permits the use of oxygen isotope ratios of PO<sub>4</sub> to trace enzymatic reactions and biological cycling of P in natural environments. The unique chemical properties of PO<sub>4</sub> also make  $\delta^{18}O$  values of inorganic environmental PO<sub>4</sub> (e.g., PO<sub>4</sub> in soils, rocks; dissolved PO<sub>4</sub>) an ideal signature of the presence of both extant and extinct biological (enzymatic) activity as well as hydrothermal activity. Fundamental processes underlying the recording of biological signals in oxygen isotope ratios of environmental phosphate and development and application of  $\delta^{18}O_P$  as a biomarker for Mars will be discussed, including characterization of  $\delta^{18}O_P$  signatures of key Martian PO<sub>4</sub> source reservoirs by analysis of Martian meteorites, and consideration of the co-evolution of  $\delta^{18}O_P$  signatures with life on Mars and in terrestrial analogue systems.

P52B-06 1445h

### Oxygen Isotope Composition of Silica in ALH84001: Implications for Water on Early Mars

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The oxygen isotopic compositions of silica and SiO<sub>2</sub>-rich glass have been measured in situ by ion microprobe in the Martian meteorite ALH84001. Two different textural occurrences, a large anhedral (40x60μm) partially crystalline silica grain and a SiO<sub>2</sub>-rich glass-rich region of a granular band, have been studied. The large silica grain has been studied by electron microprobe [1] and micro-Raman spectroscopy [2] and is found to be compositionally similar to the fine-grained SiO<sub>2</sub>-rich regions in the granular band [1]. The SiO<sub>2</sub>-rich glass is finely intergrown with carbonate (of limited compositional range), chromite, feldspar and orthopyroxene. <sup>18</sup>O/<sup>16</sup>O ratios were measured using the UCLA ims 1270 ion microprobe in multicollector mode using quartz mineral standards. Three analyses of the large silica grain in 2 analytical sessions gave a small range in oxygen isotopes (d18O=+24.41±/-0.80per mil to +25.34±/-2.6per mil; 2s); a similar range was measured for four regions of SiO<sub>2</sub>-rich glass in the granular band (d18O=+22.37±/-0.38per mil to +24.08±/-0.58 per mil; 2s). Our data show a limited range in d18O of silica which is in contrast to the large range found for ALH84001 carbonates (d18O: -10 to +28per mil [3-7]) and no evidence of low d18O values that would be indicative of magmatic or high-temperature silica [8]. Our results are heavier (by 2-4per mil) than a value published by [3] probably reflecting our more accurate correction for instrumental mass fractionation. The similar diffusivities for oxygen in calcite and quartz suggest that carbonate was deposited over a wide temperature range but that silica precipitated over a limited temperature interval. In the granular band, silica and carbonate appear to have been co-precipitated, potentially providing an opportunity to constrain the temperature and oxygen isotopic composition of the formation water for these two phases. The carbonate is too fine-grained for ion probe isotopic analysis, however the well-defined correlation of major-element and oxygen isotope compositions of ALH84001 carbonate [3-6] could be used to constrain its oxygen isotope composition. Electron microprobe analyses are in progress and will be reported at the meeting. These data should provide our best proxy for the oxygen isotope composition of water on early Mars. Implications for the search for life using the stable isotopes of oxygen will be discussed.

P52B-07 1500h

### Biological Iron Isotopic Fractionations in Antarctic Endolithic Microbial Communities

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In the McMurdo Dry Valleys, cryptoendolithic microorganisms under sandstone surfaces secrete oxalic acid to leach iron oxides from the rock. A translucent surface rock layer is necessary to transmit sufficient sunlight to support photosynthetic primary production and long-term survival. Part of the mobilized iron is re-deposited on the rock surface as a protective crust; the rest accumulates below the colonized zone. We report here that this weathering process results in redistribution of the iron isotopes, with the microbial zone being enriched in heavy isotopes relative to the rock crust and the accumulation zone. In a simulated laboratory experiment to understand the cause for this isotopic effect, hematite was incubated in 5 mM oxalic acid under light. Analysis of the initial dissolved iron showed that the dissolution in itself could not reproduce the isotopic shifts observed in the rock. Presumably, equilibrium isotopic fractionation between Fe(II) and Fe(III) species is the cause, as both are produced from oxalate-promoted dissolution of iron oxides. Subsequently, microorganisms would recycle oxalate for carbon nutrient and as a result destroy iron oxalate complexes. Without chelation, the ferric iron, which is isotopically heavier, would precipitate first and the ferrous iron later as they are transported downward through the circumneutral endolithic environment, effectively achieving a physical separation of the different isotopes. On Mars, if endolithic microorganisms had occurred and then became extinct as the planet dried and cooled, their iron isotopic biosignatures might be well preserved because subsequent reworking of iron would be unlikely without liquid water.

P52C MCC: 270 Friday 1530h

### Space Weathering of Solid Surfaces in the Solar System and Elsewhere I

(joint with SA, SH, SM)

Presiding: C A Hibbitts, University of Washington; J F Cooper, Raytheon ITSS

P52C-01 1530h INVITED

### Space Weathering on the Surfaces of Planets, Satellites and Asteroids

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The vapor deposition model of space weathering will be discussed. The changes in the optical properties of regoliths of silicate bodies without atmospheres, including spectral darkening, reddening and obscuration of absorption bands, are due to submicroscopic metallic iron. The iron particles are created during the deposition of vapor generated by both solar wind sputtering and meteorite impact vaporization. The history of the model will be briefly reviewed and evidence supporting it presented. It will be shown to be able to account quantitatively for changes in lunar optical properties, and to predict the alteration of spectra of ordinary chondrites to more closely resemble those of S-asteroids. Applied to Mercury, it implies that this body has a regolith in which FeO is low (2-6%), but not completely absent.

P52C-02 1545h INVITED

### Space Weathering of Small Bodies

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Space weathering is defined as any process that wears away and alters surfaces, here confined to small bodies in the Solar System. Mechanisms which possibly alter asteroid and comet surfaces include solar wind bombardment, UV radiation, cosmic ray bombardment, micrometeorite bombardment. These processes are likely to contribute to surface processes differently. For example, solar wind bombardment would be more important on a body closer to the Sun compared to a comet where cosmic ray bombardment might be a more significant weathering mechanism. How can we measure the effects of space weathering? A big problem is that we don't know the nature of the surface before it was weathered. We are in a new era in the study of surface processes on small bodies brought about by the availability of spatially resolved, color and spectral measurements of asteroids from Galileo and NEAR. What processes are active on which bodies? What physics controls surface processes in different regions of the solar system? How do processes differ on different bodies of different physical and chemical properties? What combinations of observable parameters best address the nature of surface processes? Are there alternative explanations for the observed parameters that have been attributed to space weathering? Should we retain the term, space weathering? How can our understanding of space weathering on the Moon help us understand it on asteroids and comets? Finally, we have to leave behind some presuppositions, one being that there is evidence of space weathering based on the fact that the optical properties of S-type asteroids differs from those of ordinary chondrites.

URL: <http://www.astro.umd.edu/~mcfadden/swAGU.html>

P52C-03 1600h

### Space Environment Measurements for Icy Surfaces in the Solar System and Beyond

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There are dozens of icy satellites orbiting the giant planets and trillions of icy comets populating the Kuiper Belt and Oort Cloud. Such objects are likely to be common throughout other planetary systems, particularly those now known to have giant planets. Interactions of the local space environment with these bodies must be taken in account for proper interpretation of photometric and spectroscopic measurements related to surface composition. Most of these bodies either are known to have, or likely have, tenuous atmospheres of volatile gases produced by internal outgassing & surface sublimation, sputtering from charged particle and UV irradiation, and diffuse dust clouds produced from meteoritic impacts. Whether or not Pluto counts as a small icy planet or a big comet, its thin and variable atmosphere also allows direct surface exposure to the space environment. Even glacial ices on the surface of (e.g., Snowball) Earth may have been exposed to much of the interplanetary solar UV flux at early times when an effective ozone shield was absent, and the Mars atmosphere is thin enough today for direct irradiation of polar cap ices by high energy cosmic ray and solar flare ions. Planetary magnetic fields reduce exposure to interplanetary charged particles but add irradiation by magnetospheric plasma and energetic particles. At Europa the intense surface irradiation from the Jovian magnetosphere might play a role via radiolytic chemistry in possible evolution of life within the putative sub-surface ocean. Although an armada of spacecraft have been measuring for many years the parameters of the solar UV, plasma, energetic particle, and dust environments of rocky bodies, large and small, in the inner solar system near Earth's orbit around the Sun, only six spacecraft with varying capabilities (Pioneer 10/11, Voyager 1/2, Galileo Orbiter, Cassini Orbiter) have yet ventured into the domain of the icy bodies near and beyond the orbit of Jupiter. The first five have collectively provided extensive measurements on the high flux plasma and energetic particle radiation environments of the jovian and saturnian magnetospheres, and the sixth will arrive to begin a long orbital tour of the Saturn system in July 2004. For the Uranus and Neptune systems we have the Voyager 2 flyby data sets. Extensive interplanetary measurements beyond Jupiter's orbit have only been provided by the Pioneer and Voyager missions into the 30 - 60 AU region of observable Kuiper Belt Objects (KBO). Of particular note are the recent report of dust in the outer solar system from interstellar grain impacts on KBOs, which may be relevant to observed color diversity of these objects, and the likely emergence of the Voyager 1 spacecraft into the heliosheath region of the heliosphere within the next eleven-year solar cycle if not in the next few years. The solar wind termination shock now sought by Voyager 1 marks the inner boundary of the heliosheath where outward solar wind flow slows down. The heliopause at a few times that shock distance in AU is the outer contact boundary for entry into the local interstellar plasma and cosmic ray environment of the outer Kuiper Belt at about 100 -