

## **Methane on Mars: Current Observations, Interpretation, and Future Plans**

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An ESA-ASI organized workshop on the Martian methane brought together nearly one hundred enthusiastic participants to ESRIN, Frascati, Italy from world over – two thirds from Europe, one-fifth from the US, and the rest from other places. The program comprised thirteen hours of oral presentations, two hours of posters, and, most noteworthy, about five hours of discussions, over a three-day period from November 25<sup>th</sup> through the 27<sup>th</sup>, 2009. The topics cast a wide net, ranging from earth-based and space observations to their implications for potential geology and biology, from models and supporting laboratory measurements to future exploration of Mars. This is a synopsis of the main themes covered at the Workshop, many of which are discussed in detail in peer-reviewed papers in this special issue.

The two principal teams involved in the detection of the Martian methane and its behavior since 2003 reported the status of their ongoing investigations. The Mars Express (MEX) Planetary Fourier Spectrometer (PFS) team led by Vittorio Formisano (IFSI/CNR) confirmed a global methane abundance of 10-15 ppbv from an analysis of current data set of several hundred thousand spectra in the three-micron range, consistent with their 2004 report based on a much smaller subset of data (Science, 306, 1758, 2004). They further presented maps of methane distribution that are characterized by large contrasts on a regional scale. The ground-based telescopic investigation team led by Michael Mumma (GSFC) confirmed the observations of spatial and temporal variations of atmospheric methane reported previously in their 2009 paper (Science, 323, 1041, 2009). At the time of the workshop, they were not yet able to report new detection of methane (or lack of it) from their most recent observations. This team expects the new observations, obtained with unprecedented spectral resolution (e.g. CRIFES on the Very Large Telescope), to be available in the near future. Sergio Fonti reported a subtle detection of methane from an analysis of MGS Thermal Emission Spectrometer observation in the 7.65 micrometers methane band.

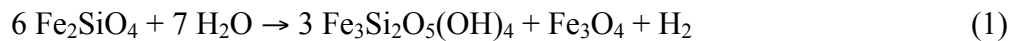
Although the MEX/PFS and the ground-based observations were generally not collected at the same time nor did they cover identical areas of Mars, the characteristics of the observed methane fields appear to be significantly different. The PFS measurements suggest maximum abundance (about 3-4 times the global value) of methane near the

north pole in mid to late summer, whereas the GSFSC observations point to a low-latitude source. Further work is warranted to reconcile the spacecraft and ground-based observations. In particular, are the differences real, do they represent temporal or spatial differences, do they point to transient flux of methane, etc? Modeling and laboratory studies presented at the workshop indicate that the reported behavior of methane, and especially its temporal and spatial variations, is difficult to reconcile with present understanding of the planet. On one hand, a strong source seems necessary to explain the reported plumes (ground-based) or maxima and minima (PFS) of methane, whereas its relatively rapid variation requires a rapid destruction mechanism, contrary to the expectations based on conventional ideas of photochemistry. New ideas – theoretical and laboratory – have been suggested, including a reactive surface, adsorption into the regolith, and seasonal release of methane from clathrates lofted into the Martian clouds, which need to be pursued further along with additional analysis of the data. Within that context, the finding of variations in methane on Mars remains an extraordinary claim, which thus “requires extraordinary evidence”. For some members of the community, the unexpected detection of methane variations could result from unexpected artifacts in the retrieval techniques, but this claim is yet to be proved. Kevin Zahnle (represented by Francois Forget; also Abstract 2456, LPSC 2010 by Zahnle et al.) challenged the detection and abundance of methane from ground-based observations since the doppler-shifted  $^{12}\text{CH}_4$  spectral feature that was attributed to Mars (Mumma et al., 2009) actually overlaps with the 20 times stronger terrestrial  $^{13}\text{CH}_4$  feature from which it cannot be distinguished. The argument does not apply to space-based Mars Express PFS data, however. The fascinating debate about the level and variability of methane started by the above observations from ground and space and their interpretation is likely to benefit from new observations in the near future, beginning with promising plans to monitor methane from the ground using heterodyne spectroscopy with an alternative methane band near 7.8 micrometers, and the tunable laser spectrometer measurements to be carried out at the surface of Mars on the 2011 Mars Science Laboratory (MSL) Mission by the Sample Analysis at Mars (SAM) instrument suite.

The origin of methane on Mars remains a puzzle. There was considerable discussion, especially in talks by Vincent Chevrier, Tullis Onstott, Buford Price and Sushil Atreya, on such questions as how methane was produced on Mars and whether its origin was biological or geological. All other potential production mechanisms – exogenic (comets, meteorites), volcanic, and atmospheric – had been previously ruled out (Atreya et al., *Planet Space Sci.*, 55, 358, 2007), and no new developments have warranted their resurrection. Since 90-95% of the 1775 ppbv of methane in the Earth’s atmosphere is ultimately biological in origin either directly or indirectly, a possible biogenic origin has also been suggested for the Martian methane and was discussed extensively at the meeting, using terrestrial analogs. On the other hand, a fraction of the terrestrial methane originates from abiotic, i.e. geological, processes such as those found in the deep sea Black Smokers and Lost City vents. For either origin, biotic or abiotic, liquid water is essential. If the production of methane on Mars is current, it implies the presence of underground aquifers. If the methane was produced when Mars was supposedly warmer and liquid water flowed on the surface, it could have been produced then, stored as clathrate, and released from time to time upon destabilization of the clathrate source.

Neither the existing observations nor any modeling studies can satisfactorily address the question of the production mechanism (origin) of methane on Mars. Much new information is required, especially on the carbon and other stable gas isotopes at high precision, map and localization of methane vents, heavier hydrocarbons such as ethylene, ethane and propane, trace gases of sulfur, nitrogen and halogen including but not limited to H<sub>2</sub>S, SO<sub>2</sub>, dimethyl sulfide, N<sub>2</sub>O, HCl, presence or absence of surface organics, together with the environmental, mineralogical and geological context of these data. The importance of above data and the means of acquiring them were discussed in several presentations on the “origin” (Sushil Atreya, Tullis Onstott, Buford Price and others), “sources” (Eric Chassefiere, Vincent Chevrier, Giuseppe Etiope, and others), “biology” (John Coates, Buford Price), and future missions (Paul Mahaffy, Christopher Webster, Richard Zurek). Additional insight into the two possible production mechanisms – biology and geology – laboratory work, and future missions, is summarized below and is based on the presentations and discussion at the workshop and papers in this issue.

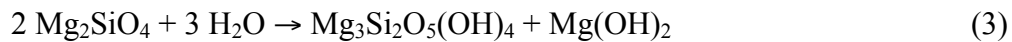
The main abiotic process for the formation of methane on Earth is hydrothermalism, which leads to the oxidoreduction (redox) reaction between iron-bearing primary silicates (olivine and pyroxene) and water to form hydrogen and FeIII-bearing phyllosilicates, a process called serpentinization. Iron-bearing olivine fayalite reacts as follows:



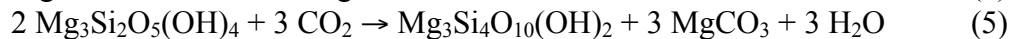
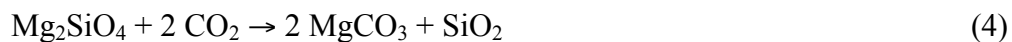
Hydrogen, a product of the reaction, can react in turn with carbon dioxide naturally present in hydrothermal systems to form methane:



Once methane is formed it can be stored as clathrate in the subsurface and released to the surface via various mechanisms, the most significant terrestrial one being mud volcanoes. (e.g. G. Etiope, this issue). Very small similar surface features (small volcanoes, fractures, etc.) could therefore be responsible for the delivery of methane to the atmosphere. The other outcome is FeIII-serpentine. This process is the main abiotic source of methane on Earth, as exemplified by production along the Mid-Atlantic Ridge. In the case of Mg-olivine (forsterite), the process also leads to serpentine (magnesian pole), but without hydrogen, due to the absence of an oxidoreduction reaction. The following reaction describes the formation of Mg-serpentine from forsterite:



Mg-olivine and Mg-serpentine can in turn react directly with CO<sub>2</sub> to form carbonates (magnesite) through a process called carbonation:



Interestingly, the recent observation of  $\text{MgCO}_3$  (magnesite) in the same region as serpentine indicates the past reactions of  $\text{CO}_2$  with the bedrock. Therefore, the iron counterparts of the silicates may have also reacted to form methane. Even if the bedrock is mostly magnesian, it always contains substitutional iron in olivine and pyroxene. For example, the most primitive Martian meteorite (ALH84001), an orthopyroxenite, has its enstatite composed of 75% Mg and 25% Fe, much more iron-rich than the terrestrial counterparts (95% Mg for the typical terrestrial olivine). The presence of serpentine in Nili Fossae (Ehlman et al., GRL, 37, L06201, 2010) indicates that serpentinization and hydrothermalism have probably occurred on early Mars. Therefore, it seems more than a coincidence that the region where methane is observed is also the region where serpentinization and carbonation have occurred. Formation of methane on early Mars could explain the overall lack of abundant carbonates on the surface, and the formation of abundant phyllosilicates like smectites instead. Methane is itself a good greenhouse gas, which could have contributed to warming the early surface and allowing the presence of liquid water.

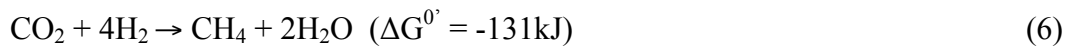
Now a major problem is to relate the observation of methane on present-day Mars to observations of serpentine and carbonates that have been dated to the oldest terrains, i.e. about 3.9 Ga old. This very long time gap complicates the connection between mineralogical and atmospheric observations. There are two possible explanations for such time gap. The first one is that the previously described processes are still active in the subsurface of Mars and could be responsible for the formation of the observed methane. This would imply a currently active Mars. However, it is difficult to imagine that Mars has been active in the same region for the past 4 billion years.

A possible alternative would be a temporary trapping of methane in the deep subsurface through clathrate formation. Methane clathrate hydrates are solid crystalline compounds (cages) composed of one mole of methane for 5.75 moles of water on average. Such compounds are mainly stable at low temperature and high pressure, compared to pure water ice. Clathrates could act as a temporal buffer, where methane might have been trapped a long time ago and be presently released to the atmosphere, due to some slow destabilization process. Clathrate particles could also be transported to the surface and then destabilized in the atmosphere (Eric Chassefiere, this issue), releasing methane. It is important to note that the concentrations of methane, even in the localized plume, are quite small. Therefore, the amount of clathrate that must be destabilized need not be large.

An important constraint on the mechanism of methane release and formation would come from future observations of potential seasonality of methane release which might reveal seasonal heating during the summer, pointing to a shallow subsurface source, or the absence of seasonality, suggesting instead a deep source. A deep source is more likely because once again it is difficult to conceive that clathrates would remain stable for 4 billion years in the shallow subsurface. Moreover, the absence of significant hydration in the shallow subsurface argues also for a deep source. Alternatively, if the source is shallow, then the clathrates could be close to their equilibration depth and this would explain why the amount of methane is so small.

A fascinating and very educational series of reports on microbial life, metabolism in water ice, and biological experiments under Martian conditions was presented by authors John Coates, Buford Price, Daniel Prieur, L. Whyte, T. Kral, R. Amils, G. Galletta, and B.K. Chastain, who considered perchlorate-reducing bacteria, microbes trapped in extreme environments, the mechanism and complexity of terrestrial methanogenesis, the extent to which cryoenvironments could be active microbial ecosystems, terrestrial analogs and methanogen energy sources.

On Earth methanogenic archaea are thought to have originated ~4 Ga, and a case has been made that microbial life, including methanogens, could have originated in an icy environment. Substrates used to provide energy and carbon include CO, CO<sub>2</sub>, formate, methanol, and acetate. Of these, the simplest is



At issue in this workshop was whether methanogens might have originated on early Mars, utilizing CO<sub>2</sub> and H<sub>2</sub> in its atmosphere and producing the bursts of methane. The Gibbs free energy for reaction (6) depends on temperature and fugacities of the reactants. Experiments have shown that  $\Delta G_r$  must be < -10 kJ/mol H<sub>2</sub> for methanogenesis via reaction (6). For a typical concentration of 15 ppm H<sub>2</sub> in the Mars atmosphere, reaction (6) can take place only if the temperature is below 0°C. This constraint has been shown to be avoided if there are H<sup>+</sup> ions in aqueous solution in the presence of Fe<sup>0</sup>, for example, from meteorites, and methanogens that utilize the H<sub>2</sub> produced.

Bacteria are known to live in liquid veins at triple junctions of grains in polycrystalline ice, where they can extract energy and essential elements from ions in the veins. They are also known to be able to live on mineral grains coated with unfrozen water layers. Montmorillonite, abundant on Mars, is favored as a habitat because it is a source of micronutrients and its surfaces retain a sub-nm coating of H<sub>2</sub>O that remains unfrozen down to the coldest Martian temperatures.

As a result of ongoing gardening by micrometeorites, microbes now at depths up to 2 or 3 m below the surface were at one time on the surface, where they are unlikely to have survived exposure to solar UV and highly oxidizing chemical compounds in the regolith. Some future missions to Mars will carry drills to recover soil samples from below the gardened layer.

Microbes immobilized in terrestrial ice have been shown to metabolize at a rate proportional to  $\exp(-Q/RT)$  that is six orders of magnitude lower than that for exponential growth and unlimited movement. This rate is just sufficient to repair macromolecular damage due to spontaneous amino acid racemization and DNA depurination. At sub-zero temperatures it is so slow that microbial lifetimes are constrained not by the supply of nutrients but by the rate of double strand breaks of DNA in cell nuclei penetrated by alpha particles from Th and U impurities within a distance ~20 μm in soil or rock.

Microbes encased in low-radioactivity ice would largely avoid this radiation damage and may have survived to the present day.

In order to account for the rapidly time-varying methane flux on Mars, methanogens could live on surfaces of extended cracks that seal up with ice in winter, and their ongoing metabolic methane could reach the surface by sublimation of that ice during summer. If the methane bursts are due to methanogens, their present concentration in subsurface regolith is estimated from their temperature-dependent metabolic rate to be as high as 0.1 to 1 cell per cm<sup>3</sup>, depending on the temperature of their habitat and the thickness of the inhabited zone. A future Mars lander equipped with a 2- to 3-m drill and a PCR analyzer might be able to detect subsurface methanogens if their DNA structure was similar to those of terrestrial bacteria. An instrument capable of measuring  $\delta^{13}\text{C}$  with sufficiently high precision could test whether the methane is consistent with methanogenesis.

If conditions were once favorable for methanogens to have originated on Mars, other bacteria or archaea might also have arisen, in which case gaseous participants in microbial metabolism such as CO, methanol (CH<sub>3</sub>OH), N<sub>2</sub>O, and ammonia (NH<sub>3</sub>) might be detectable in spatial and temporal coincidence with CH<sub>4</sub> bursts. Concentrations of 0.4-0.6 wt% perchlorates have been detected in Mars soil by the Phoenix Lander at polar latitudes. Methanogens have been shown to be able to produce methane in the presence of up to 1 wt% perchlorate. If perchlorate-reducing bacteria exist on Mars, they might lead to biological oxidation of some of the methane. Liquid solutions of perchlorates and water with eutectic temperatures as low as -67°C could provide a microbial habitat in liquid veins in permafrost. Numerous other compounds have sufficiently low eutectic temperatures to provide aqueous habitats for suitably adapted microbes.

Some sedimentary rocks on Mars may have formed in acidic lakes, probably in areas where there was prior volcanic activity. The discovery of methanogens in Rio Tinto, a terrestrial river with pH ~2.3, supports the possibility that methane bursts in Martian sedimentary rocks have a biological origin.

The Lost Hammer Spring is an Arctic hypersaline spring in 600 m of permafrost with winter temperature as low as -50°C. Methane seeps are found in the spring with average flux 6000 L/yr, and underlying wet sediment contains ~6 x 10<sup>5</sup> cells/g, of which a few percent comprise methanogens and anaerobic methane-oxidizing bacteria. The site provides a model of how a methane seep can form in thick permafrost and a mechanism that might contribute to plumes of methane on Mars.

Experiments in environmental chambers are being used to study the limits of microbial life under Martian conditions. *Bacillus* strains survive from minutes to a few hours. Endospores survive even in vacuum, and they typically survive ultraviolet doses ~10<sup>2</sup> times higher than are tolerated by vegetative cell.

Laboratory measurements relevant to the study of Mars methane were also presented, covering a wide range of studies of Earth and Martian studies within the disciplines of

chemistry, photochemistry, heterogeneous chemistry, spectroscopy, astrobiology, microbiology, geophysics, and thermodynamics.

Vincent Chevrier presented an analysis of phyllosilicate thermodynamics at Mars conditions that underscored the importance of serpentinization, but could not reconcile the need for either a still-active process such as hydrothermalism or a long-lived buffer effect by deep clathrates. Insight into the possible Martian process was given by Tullis Onstott in his comparison with methane release in Earth's Arctic region, how stable isotope data could unravel parts of the mystery, and his report on first experiments on methanogens under Mars conditions. Laboratory investigation of the possible role of heterogeneous processing at the surface was presented by Raina Gough and coauthors. Although many groups consider the surface oxidation of methane (e.g. by hydrogen peroxide) a viable sink, the laboratory measurements of Gough et al. concluded that this process was likely too slow to account for the observed short lifetimes. Several suggestions were made that could test these results through future experimentation. For example, Sushil Atreya pointed out that although hydrogen peroxide itself is known to not destroy methane directly (e.g. Wong et al., JGR 108 (E4), 7-1, 2003), the hydroxyl (OH) and hydroperoxy (HO<sub>2</sub>) radicals as well as superoxides (such as O<sub>2</sub><sup>-</sup>), that are likely to form in the mineralogical processing of hydrogen peroxide in the regolith, can destroy the gas efficiently through oxidation. In the atmosphere this destruction mechanism is ineffective as the lifetime of peroxide is very short (<1d) in contrast to hundreds to several millions of years in the regolith. The long lifetime is crucial for mineralogical processing of oxidants such as hydrogen peroxide to even more reactive forms since such processes are relatively slow. The resulting radicals, especially hydroxyl, would play another critical role, that of recycling excess CO back into CO<sub>2</sub>, if the excess peroxide was produced by electrochemistry triggered by triboelectric processes in convective storms. Another potentially important oxidant on Mars was detected by the Phoenix Polar Lander. Perchlorates in the amount of 0.4-0.6 wt% including Mg(ClO<sub>4</sub>)<sub>2</sub>, NaClO<sub>4</sub> and smaller quantities of Ca(ClO<sub>4</sub>)<sub>2</sub> and KClO<sub>4</sub> were measured by the Wet Chemistry Lab at the high latitude landing site of this mission (Hecht et al., Science 325, 64, 2009). Although perchlorates are highly stable as oxidants, their processing in the Martian regolith could also render them into highly reactive forms, which may contribute to the destruction of surface organics and methane. Similar to peroxide, the long residence time in the soil is key to such processing. Mineralogical processing of oxidants in the Martian soil, which could somehow simulate the long processing time and the changing nature of the soil in the interim, would be valuable for understanding the destruction of Martian organics including methane. Concerning the presence of perchlorates at non-polar latitudes, no direct measurements are yet available; however, reanalysis of the Viking GCMS data suggests small quantities of perchlorates at ≤0.1 wt% level as well as organics, methyl chloride and dimethyl chloride, of the Martian origin (Navarro-Gonzalez, et al., JGR, 2010, in press) may have been present at the mid-latitude landing sites of the two Viking landers. This is quite promising for the upcoming MSL Mission, Curiosity, whose landing site will be in a non-polar region of Mars (next section).

Nearing completion for a 2011 launch, NASA's Mars Science Laboratory will investigate habitability on Mars over the next few years by assessing the biological potential of at

least one target environment (past or present), characterizing the geology and geochemistry of the landing site, and investigating planetary processes that influence habitability. Paul Mahaffy and Chris Webster described the Sample Analysis at Mars (SAM) experiment suite which includes a Quadrupole Mass Spectrometer (QMS, GSFC), Gas Chromatograph (GC from University of Paris, France) and a Tunable Laser Spectrometer (TLS, JPL). This powerful suite that also includes a gas processing system and sample manipulation system will focus on five overarching goals: (i) To explore sources and destruction paths for carbon compounds, (ii) to search for organic compounds of biotic and prebiotic relevance including methane, (iii) to reveal chemical and isotopic state of other light elements that are important for life as we know it on Earth, (iv) to study habitability of Mars by atmosphere-surface interactions expressed in compositions of trace species, and (v) to understand atmosphere and climate evolution through isotope measurements of noble gases and light elements. The TLS measurements of CH<sub>4</sub> to a few part-per-trillion capability, and of <sup>13</sup>C/<sup>12</sup>C and D/H in methane will be made in context with numerous other measurements by SAM of related trace gases, and isotope ratios in a variety of gases including water vapor and carbon dioxide. These measurements will be made for comparison with values found in Martian soil and rocks. Complementary measurements to be made by MOMA on the 2018 ExoMars mission were also presented (H. Steininger, L. Becker).

The session on future missions continued with a presentation by Richard Zurek and Augustin Chicarro on the report of a Joint Instrument Definition Team studying a proposed ESA-NASA mission to be launched in 2016. This would be the first of a series of joint missions between ESA and NASA in a possible program outlined by Marcello Coradini in the opening session of the workshop.

The 2016 mission would focus on: (1) sensitive detection of a suite of trace gases in the Mars atmosphere, to better understand the nature of the exchange of trace gases between the solid planet and the atmosphere and their possible origin (2) characterization of processes affecting atmospheric lifetimes and of possible residence in temporary reservoirs, such characterization would include the role of dust and ice aerosols in trace gas chemistry, and (3) localization of surface sources, assuming they exist and can be identified at a scale appropriate to exploration by a future lander or aerobot. The mission concept is to fly a capable payload into a low, inclined Mars orbit to facilitate observations at various local times in various seasons. This was felt necessary to capture the essence of expected photochemical variability and to understand the nature of the variations in methane concentration reported thus far. The payload would have the advantage of avoiding the Earth's atmosphere, with which the ground-based observers must contend, but would also need significantly improved spectral resolution and wavelength coverage, beyond that now available from current and past Mars missions. Given that the release of the payload Announcement of Opportunity for 2016 was imminent (and in fact happened on January 15, 2010, and payload selected in August 2010), there was great expectation that a mission could be deployed to follow up on the recent discoveries. Several presentations and follow-up discussions focused on how such a payload and observations generally of trace gases might be optimized. Presentations in the oral and poster sessions examined recent technical applications and possible new



developments, such as the use of acousto-optic spectrometers (SOIR; Drummond et al.) and laser heterodyne techniques (R. Passmore et al.). The importance of understanding not only dust concentration, but also dust properties such as shape and area, was discussed in light of possible heterogeneous chemical processes on the aerosols (C. Muller and others); examples of instrumentation (e.g., the SPEX spectropolarimeter) that could provide the needed information were presented.

More futuristic approaches to the problem were considered, including measurements from airborne platforms such as airplanes (J. Levine) and various classes of balloon-borne payloads (S. Pahari). Discussion (M. Mumma and others) raised the novel possibility of a satellite docked at a Mars Lagrange point to provide global imaging of atmospheric composition with very high spectral resolution.

A key point of the discussion on trace gas measurements was continuation of the present ground-based program and coordination of those efforts with observations from space such as those with Mars Express PFS. As mentioned previously, both earth-based spectrometers and PFS observe striking variations in methane concentration, but the patterns reported differ in important respects. Coordinated measurements might help resolve the matter, though final resolution of present differences and unassailable evidence of any variability will still require the new measurements planned on future missions to Mars.

### **Acknowledgments**

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Methane on Mars. Quite the same Wikipedia. Just better. Since 2004, trace amounts of methane (range from 60 ppb to under detection limit ( $< 0.05$  ppb)) have been reported in various missions and observational studies.[9][10][11][12][13] The source of methane on Mars and the explanation for the enormous discrepancy in the observed methane concentrations are still unknown and. The enigma of methane on Mars Methane, an indicator of life? Methane ( $\text{CH}_4$ ) is an organic molecule present in gaseous form in the Earth's atmosphere. More than 90% of methane on... For further progress to be made in unveiling the origin of methane on Mars, future space missions with new technologies devised to better characterise the Martian environment and its subsurface will be necessary. Confirming the presence of methane on Mars, a goal of the ExoMars programme. Observations from the Planetary Fourier Spectrometer (PFS) on ESA's Mars Express and from very high spectral resolution spectrometers on ground-based telescopes, have detected variable amounts of methane in the atmosphere of Mars. Could this be evidence for life on Mars? The greenhouse gas, methane, is produced by both natural processes and human activities. While there has been much attention paid to curbing anthropogenic emissions, a changing climate will likely increase the production of natural methane. In an open access article recently published in Reviews of Geophysics, Dean et al. [2018] describe the ways in which biological, geochemical, and physical systems influence methane concentrations and explore how methane levels in natural systems may alter in a warming climate. Here the authors answer some questions about the sources and significance of meth... PDF | Recent measurements of methane ( $\text{CH}_4$ ) by the Mars Science Laboratory (MSL) now confront us with robust data that demand interpretation. The | Find, read and cite all the research you need on ResearchGate. have been observed. These observations pose fundamental challenges to our. current understanding of Mars. First, according to known. atmospheric chemistry, the gas-phase lifetime of  $\text{CH}_4$ .