

Key Points:

- Crater mixing time scales are ~1 sol during all seasons
- Methane abundances vary by at least an order of magnitude over a diurnal cycle under all the scenarios considered
- It is difficult to reconcile measurements with the modeled transport and mixing

Correspondence to:

J. Pla-Garcia,
jpla@cab.inta-csic.es

Citation:

Pla-Garcia, J., Rafkin, S. C. R., Karatekin, Ö., & Gloesener, E. (2019). Comparing MSL Curiosity rover TLS-SAM methane measurements with Mars Regional Atmospheric Modeling System atmospheric transport experiments. *Journal of Geophysical Research: Planets*, 124. <https://doi.org/10.1029/2018JE005824>

Received 13 SEP 2018

Accepted 15 JUL 2019

Accepted article online 24 JUL 2019

Author Contributions:

Conceptualization: Jorge Pla-Garcia, Scot C. R. Rafkin

Data curation: Jorge Pla-Garcia, Scot C. R. Rafkin, Özgür Karatekin, Elodie Gloesener

Formal analysis: Jorge Pla-Garcia, Scot C. R. Rafkin

Funding acquisition: Jorge Pla-Garcia, Scot C. R. Rafkin

Investigation: Jorge Pla-Garcia, Scot C. R. Rafkin, Özgür Karatekin, Elodie Gloesener

Methodology: Jorge Pla-Garcia, Scot C. R. Rafkin

Project administration: Jorge Pla-Garcia, Scot C. R. Rafkin
(continued)

Comparing MSL Curiosity Rover TLS-SAM Methane Measurements With Mars Regional Atmospheric Modeling System Atmospheric Transport Experiments

Jorge Pla-Garcia^{1,2} , Scot C. R. Rafkin³, Özgür Karatekin⁴, and Elodie Gloesener^{4,5}

¹Centro de Astrobiología (CSIC-INTA), Torrejón de Ardoz, Spain, ²Space Science Institute, Boulder, CO, USA, ³Southwest Research Institute, Boulder, CO, USA, ⁴Royal Observatory of Belgium, Brussels, Belgium, ⁵Earth and Life Institute, Université Catholique de Louvain, Louvain-la-Neuve, Belgium

Abstract The detection of methane at Gale crater by the Tunable Laser Spectrometer–Sample Analysis at Mars instrument aboard the Curiosity rover has garnered significant attention because of the implications for the presence of Martian organisms (Webster et al., 2015, <https://doi.org/10.1126/science.1261713>). Methane's photochemical lifetime is several centuries unless there is a fast, as-yet-unknown destruction mechanism (Lefèvre and Forget, 2009, <https://doi.org/10.1038/nature08228>). This is much longer than the atmospheric mixing time scale, and thus, the gas should be well-mixed except when near a source or shortly after a release. Although most measurements report low background levels of ~0.4 parts per billion by volume, observed spikes of several parts per billion by volume or greater and a subsequent return to the background level are intriguing (Webster et al., 2015, <https://doi.org/10.1126/science.1261713>). The Mars Regional Atmospheric Modeling System is used to simulate, via passive tracers, the transport and mixing of methane released inside and outside of the crater from instantaneous and steady state releases, and to test whether the results are consistent with in situ observations made by the Mars Curiosity rover. The simulations indicate that the mixing time scale for air within the crater is approximately 1 sol. The timing of methane measurements within the crater is also important, because modeled methane abundance varies by ~1 order of magnitude over a diurnal cycle under all the scenarios considered. While the observed low background levels can be reproduced by the model under some circumstances, it is difficult to reconcile the measured peaks with the modeled transport and mixing. For periods of high methane abundance lasting longer than a few hours there must be a continuous release of methane inside the crater to counteract mixing, or there must be a large, methane-rich air mass continually transported into the crater. The few scenarios that can produce peaks are problematic, because they would result in background methane values above what is observed.

Plain Language Summary The in situ detection of methane at Gale crater by the Tunable Laser Spectrometer–Sample Analysis at Mars instrument aboard the Mars Science Laboratory Curiosity rover has garnered significant attention because of the potential implications for the presence of indigenous Martian organisms. There are many major unresolved questions regarding this detection: (1) Where is the release location? (2) How spatially extensive is the release? (3) For how long is methane released? In an effort to address the release location of methane, the spatial extensivity, and the magnitude and duration of the release, atmospheric circulation studies of Gale crater (where the Mars Science Laboratory Curiosity rover landed in 2012) were performed with the Mars Regional Atmospheric Modeling System Martian meteorological model using tracers to study transport and mixing of methane from potential source locations. The aim of this work is to test whether methane releases inside or outside of Gale crater are consistent with Tunable Laser Spectrometer–Sample Analysis at Mars observations.

1. Introduction

1.1. Review of Ground-Based and Orbiter Methane Measurements

The possibility of detecting methane in Mars' atmosphere has attracted a great deal of attention, because methane on Earth is produced primarily by biological activity (90–95%), organic waste, or fossilized matter (Atreya et al., 2007). Martian methane could be a signal of present or past life on the Red Planet. The first

©2019. The Authors.

This is an open access article under the terms of the Creative Commons Attribution License, which permits use, distribution and reproduction in any medium, provided the original work is properly cited.

Resources: Jorge Pla-Garcia, Scot C. R. Rafkin, Özgür Karatekin, Elodie Gloesener

Software: Jorge Pla-Garcia, Scot C. R. Rafkin

Supervision: Jorge Pla-Garcia, Scot C. R. Rafkin

Validation: Jorge Pla-Garcia, Scot C. R. Rafkin

Visualization: Jorge Pla-Garcia, Scot C. R. Rafkin

Writing - original draft: Jorge Pla-Garcia, Scot C. R. Rafkin

Writing - review & editing: Jorge Pla-Garcia, Scot C. R. Rafkin, Özgür Karatekin, Elodie Gloesener

reported detection of methane in the atmosphere of Mars was made with the Mariner 7 spacecraft Infrared Spectrometer, and was announced at press conference two days after the Mars flyby (Sullivan, 1969); however, shortly after, it was shown that the observed spectral signatures were actually from CO₂ ice.

Despite the Mariner 7 confusion, the search for methane on Mars continued. Over the last 20 years, there have been several reports of methane detection from Earth and from Mars orbit, although the detections are controversial. Until very recently, all the putative detections have presented uncertainties associated with weak signal, poor spectral resolution, telluric line contamination, or instrument noise or performance issues. A variety of detections of methane in the Mars atmosphere have been reported since 1999 (Table 1). The first of these detections suggested a global average value of 10 ± 3 parts per billion by volume (ppbv) using the Fourier transform spectrometer at the Canada–France–Hawaii Telescope (capturing only a portion of the Martian disk) and searching for methane in the 3.3- μ m spectral band (Krasnopolsky et al., 2004).

In the second detection, Formisano et al. (2004) co-added spectra from the Planetary Fourier Spectrometer (PFS) onboard ESA's Mars Express (MEX) spacecraft over a wide range of latitude and longitude to obtain a global average value of 10 ± 5 ppbv requiring a methane emission of 126 t per year. That global average value was later updated to 15 ± 5 ppbv (Geminale et al., 2011). Geminale et al. (2011) showed evidence of widespread temporal and spatial variability with indications of discrete localized sources (they found that methane is not uniformly distributed in the Martian atmosphere) and a summertime maximum of 45 ppbv in the north polar region. These observations suggest that the variation of methane abundance was a feature of Mars from at least 2004 to 2008. Like all the detections, there are potential issues and controversy. The PFS-MEX measurements are limited by a spectral resolution that is ~ 200 times lower than the Mars methane line widths (Webster & Mahaffy, 2011; Zahnle et al., 2011) with detection compounded by noise from mechanical vibrations (Comolli & Saggin, 2009).

Mumma et al. (2009) (hereafter M09) used the infrared high-resolution spectrometers NIRSPEC and CSHELL at the high-altitude telescope observatories Keck-2 and NASA-IRTF, respectively, to search for methane in the 3.3- μ m spectral region. The distinct spatial variability reported in M09 suggests regional source emissions (Figure 1).

These results suggest that methane is not just slowly leaking out from discrete, local regions, but that there must also be large, intermittent releases from specific regional areas. The emission flux for the largest methane plume was estimated to be ≥ 0.63 kg/s, generating a mean mixing ratio of ~ 33 ppbv close to the Syrtis Major volcanic area (approximately 8,000,000 km²) with a peak mixing ratio of ~ 45 ppbv during northern summer, and very low methane outside that area. Distributed over the whole planet, this is equivalent to a global average mixing ratio of ~ 2 ppbv. The volume of this emission could rise up to 57,000 t per year. Together with other detections, M09 estimated a global average of ~ 6 ppbv. Observations of the following Martian year found a global mixing ratio of ~ 3 ppbv from which M09 concluded that the lifetime of atmospheric methane had to be less than approximately four Earth years. Methane measurements at later seasons (Villanueva et al., 2013) were generally smaller than the values reported by M09. The temporal variability could indicate seasonal variations in the source strength, an intermittent source of methane or extremely rapid destruction of methane through nonphotochemical processes. Or, the detection could be in error.

Zahnle et al. (2011) suggested that Earth-based observations (Krasnopolsky et al., 2004 and M09) of methane lines could be confused with other Martian spectral lines or Doppler-shifted telluric lines (when Mars was blue-shifted, the methane lines observed by M09 overlapped telluric lines, while the lines did not overlap when Mars was red-shifted) and that the most favorable wavelengths of observations indicate no methane above a 3-ppbv noise floor.

Fonti and Marzo (2010), using the Thermal Emission Spectrometer (TES) onboard Mars Global Surveyor (MGS), showed evidence of widespread temporal variability with a global average value of 5 ± 2 to 33 ± 9 ppbv between 1999 and 2004 and with strong spatial variability in the methane signal intermittently present over locations where favorable geological conditions such as residual geothermal activity (Tharsis and Elysium) and strong hydration (Arabia Terra) might be expected. There is controversy about these detections (Zahnle et al., 2011), because TES lacks spectral line resolving power and requires the co-addition of nearly 3,000,000 spectra to produce a very weak signal. Also, the identification of a methane signal is

Table 1*Detections of Methane in the Mars Atmosphere Reported Since 1999 Through 2019 in ppbv*

Type (instrument)	Observation	L_s (°)	Maximum value (region)	Global average (ppbv)	Reference
EB (FTS-CFHT)	1999	88	-	10 ± 3	Krasnopolsky et al. (2004)
MO (TES-MGS)	1999	180	~68 (Tharsis), ~64 (AT), ~60 (Elysium)	33 ± 9	Fonti and Marzo (2010)
MO (TES-MGS)	1999	270	~26 (Tharsis), ~30 (AT), ~24 (Elysium)	6 ± 2	Fonti and Marzo (2010)
MO (TES-MGS)	2000	0	~34 (Tharsis), ~32 (AT), ~32 (Elysium)	17 ± 5	Fonti and Marzo (2010)
MO (TES-MGS)	2000	90	~30 (Tharsis), ~40 (AT), ~38 (Elysium)	14 ± 4	Fonti and Marzo (2010)
MO (TES-MGS)	2001	180	~56 (Tharsis), ~62 (AT), ~60 (Elysium)	18 ± 7	Fonti and Marzo (2010)
MO (TES-MGS)	2001	270	~24 (Tharsis), ~24 (AT), ~22 (Elysium)	5 ± 2	Fonti and Marzo (2010)
MO (TES-MGS)	2002	0	~32 (Tharsis), ~28 (AT), ~30 (Elysium)	10 ± 4	Fonti and Marzo (2010)
EB (CSHELL-IRTF)	2003	155	<45 (TS, NF, and SM)	6	Mumma et al. (2009)
MO (TES-MGS)	2003	180	~58 (Tharsis), ~56 (AT), ~52 (Elysium)	30 ± 8	Fonti and Marzo (2010)
MO (TES-MGS)	2003	270	~22 (Tharsis), ~20 (AT), ~20 (Elysium)	5 ± 1	Fonti and Marzo (2010)
MO (TES-MGS)	2004	0	~30 (Tharsis), ~30 (AT), ~30 (Elysium)	9 ± 3	Fonti and Marzo (2010)
MO (TES-MGS)	2004	90	~56 (Tharsis), ~60 (AT), ~40 (Elysium)	28 ± 8	Fonti and Marzo (2010)
MO (PFS-MEX)	2004	330–350	-	10 ± 5	Formisano et al. (2004)
MO (PFS-MEX)	2004	330–10	20 ± 10 (Elysium)	-	Encrenaz (2008)
EB (CSHELL-IRTF)	2006	10	-	<14	Krasnopolsky (2007)
EB (CSHELL-IRTF)	2006	10	<10 (VM, 63–93°W and 0 to 7°N); 3 outside this region	-	Krasnopolsky (2012)
EB (CSHELL-IRTF, NIRSPEC)	2006	17	4	3	Mumma et al. (2009)
EB (CSHELL-IRTF, NIRSPEC)	2006	352	-	<7.8	Villanueva et al. (2013)
MO (PFS-MEX)	2004–2008	50	21 (–40E and +70E longitude)	14 ± 5	Geminale et al. (2008)
MO (PFS-MEX)	2004–2008	160–180	<45 (north polar region)	15 ± 5	Geminale et al. (2011)
MO (PFS-MEX)	2004–2008	325	5 (–40E and +70E longitude)	15 ± 5	Geminale et al. (2008)
EB (CSHELL-IRTF, NIRSPEC)	2009	12	-	<6.6	Villanueva et al. (2013)
EB (CSHELL-IRTF)	2009	20	<8 (0–30°W)	-	Krasnopolsky (2012)
EB (CSHELL-IRTF)	2010	70	<8 (30°W to 90°E and along the central meridian)	-	Krasnopolsky (2011)
EB (CSHELL-IRTF, NIRSPEC)	2010	83	-	<7.2	Villanueva et al. (2013)
In situ (TLS-SAM)	2013–2014	336–82	7.2 ± 2.1 (Gale crater)	-	Webster et al. (2015)
EB (SOFIA~12–14 km)	2016	123	1–9 (several locations)	-	Aoki et al. (2018)
MO (PFS-MEX)	2013	336	15.5 ± 2.5 (Gale crater)	-	Giuranna et al. (2019)
MO (ACS-NOMAD-TGO)	2018	162–240	<0.05 (several locations)	-	Korablev et al. (2019)

EB, MO, AT, TS, NF, SM, and VM stands for Earth-based, Mars orbit, Arabia Terra, Terra Sabae, Nili Fossae, Syrtis Major, and Valles Marineris, respectively.

controversial due to the presence of nearby H_2O and CO_2 lines. The identification of methane depends on spatial and seasonal correlations with results from Geminale et al. (2011) and M09.

Other locations with high methane levels from subsequent observations are Valles Marineris, with an upper limit of 10 and 3 ppbv outside that region detected with CSHELL/IRTF (Krasnopolsky, 2012), and over the Elysium region with a value of 20 ± 10 ppbv detected with PFS-MEX (Encrenaz, 2008), although the methane fit of these latter data relies on a single spectral element and could be spurious.

In an effort to minimize the previous problems observed (telluric contamination of Martian spectral features, instrument noise, and very low spectral resolution data), observations were performed with the Echelon-Cross-Echelle Spectrograph onboard the Stratospheric Observatory for Infrared Astronomy (SOFIA). The high-altitude measurements of SOFIA (~12–14 km) significantly reduce the effects of the terrestrial atmosphere, allowing the use of methane lines in the 7.5- μm band. These measurements suggest an upper limit on the methane volume mixing ratio ranging from 1 to 9 ppbv (Aoki et al., 2018). The measurements were taken during summer in the northern hemisphere (L_s 123°) of Mars year 33.

Based on current understanding, the total photochemical loss rate of methane in the Martian atmosphere is $2.2 \times 10^5 \text{ cm}^2/\text{s}$, and its lifetime is 340 years (Krasnopolsky et al., 2004). Since the vertical and horizontal mixing time is much shorter than the photochemical lifetime, methane should be uniformly mixed and

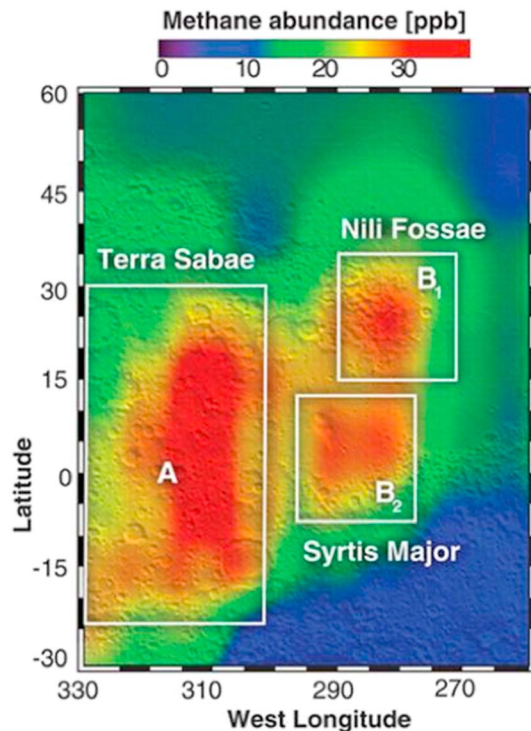


Figure 1. M09 observations of methane near the Syrtis Major volcanic district where methane appears notably localized (A, B₁, and B₂) in northern summer L_s 155°. Adapted from M09.

distributed throughout the atmosphere. However, the different methane observations (Table 1) indicate a temporal and spatial variability of methane that is inconsistent with a well-mixed atmosphere or with a long photochemical lifetime (Lefèvre & Forget, 2009) unless the observations were made shortly after methane was emitted. Further, with a long photochemical lifetime, even episodic emissions like those identified in Table 1 would result in large global methane abundance, since the abundance would build over time.

Photochemical activity might be expected to rise and fall slightly with the seasons, but the slight variations are insufficient to produce the necessary and large methane sink required to explain the observed methane variability. Of course, Martian photochemical models could be missing one or more destruction mechanisms. Heterogeneous chemistry processes and the interaction with chemicals on the surface and within the regolith (Jensen et al., 2014) are generally lacking in most models but could be important.

1.2. Tunable Laser Spectrometer– Sample Analysis at Mars Methane Detections at Gale Crater

In situ measurements provide ground truth using direct and, in principle, more reliable methods than those from Earth or Mars orbit. The Tunable Laser Spectrometer (TLS) of the Sample Analysis at Mars (SAM) instrument suite aboard the MSL Curiosity rover at Mars (Mars) was specifically designed to obtain abundance measurements of methane and its isotopologues using laser absorption spectroscopy, the latter being possible if abundance >1 ppbv were encountered. TLS-SAM determines methane abundances by

taking the difference between measurements from a cell filled with an atmospheric sample and after it has been evacuated. Using this difference technique minimizes the effect of potential contamination between measurements (Webster et al., 2015).

The two different atmospheric sampling methods used by TLS-SAM are the “direct ingest” method (hereafter DIM) and the “enrichment” method (hereafter EM), as further detailed in Webster et al. (2015). In both cases, the local time of ingest is constrained by competing rover activities and power availability. Most of the TLS-SAM measurements were acquired during nighttime (except on sols 305 and 525) due to thermal requirements of the TLS-SAM sample handling system (Table 2). Also, long periods of time usually pass between measurements.

The record of TLS-SAM methane measurements is shown in Table 2 and Figure 2. The first three measurements (sols 79, 81, and 106 after MSL Curiosity rover landing) indicated a value of <1.5 ppbv using the direct ingestion method. Another measurement taken almost 200 sols later (on sol 293, more than a half Earth year later from previous measurement) was also at the 1-ppbv level.

A spike in methane abundance was first noted on sol 305 (L_s 336°). This is a transitional time at Gale crater, when strong flushing northern winds give way to a less intense circulations typical of the rest of the year as shown by Pla-Garcia et al. (2016) and Rafkin et al. (2016) (hereafter PGR16). It is commonly assumed that methane abundance remained continuously elevated between sol 467 and 525 ($\sim L_s$ 55–81°), with a mean value of 7.2 ± 2.1 ppbv (95% confidence interval; Webster et al., 2015), but there are no data to support this assumption. The infrequency of methane measurements introduces uncertainty about variations between spikes, because it is not known precisely when the spikes began, how long they lasted, or how long it took for the values to return to background levels. It is possible that methane values return to background values in hours or sols after the peak, and the consecutively detected spikes are serendipitous, or it could be that the abundance remains elevated for nearly the entire 60-sol period. The methane concentration once again was at background levels when a measurement was taken at sol 573 using the EM. Subsequent measurements at sol 684 (both direct ingest and enrichment method) were also at the background level. Between sols 1,141

Table 2

MSL Curiosity Rover TLS-SAM Methane Measurements at Gale Crater Over an ~4.5 Earth Years (~56-Month) Period (from 26 October 2012 to 27 May 2017)

Run description	Sol	LMST	L_s deg	Mean CH ₄ value ± 1 SEM (ppbv)
DIM1	79	23:02	195.60	-0.51 ± 2.83
DIM2	81	21:21	196.77	1.43 ± 2.47
DIM3	106	03:21	211.74	0.69 ± 2.15
DIM4	293	03:50	329.16	0.56 ± 2.14
DIM5	305	13:55	336.12	5.78 ± 2.27
DIM6	314	03:21	340.83	2.13 ± 2.03
DIM7	467	03:21	55.59	5.48 ± 2.19
DIM8	475	03:21	59.20	6.88 ± 2.11
DIM9	505	02:52	72.66	6.91 ± 1.84
DIM10	525	13:26	81.84	9.34 ± 2.16
EM1	573	01:55	103.48	0.43 ± 0.09
EM2	684	01:26	158.61	0.74 ± 0.13
DIM11	684	06:28	158.73	-0.41 ± 1.89
EM3	965	23:45	331.57	0.60 ± 0.08
EM4	1086	01:26	32.81	0.23 ± 0.05
DIM12	1125	03:21	50.81	0.85 ± 1.44
DIM13	1141	02:24	58.03	2.40 ± 1.50
EM5	1169	00:28	70.57	0.22 ± 0.07
DIM14	1169	05:31	70.66	-0.15 ± 1.46
DIM15	1222	02:24	94.50	2.03 ± 1.85
EM6	1322	00:00	142.46	0.57 ± 0.11
DIM16	1322	05:02	142.57	2.57 ± 2.24
EM7	1451	01:26	216.58	0.49 ± 0.07
DIM17	1451	06:28	216.71	0.01 ± 1.97
EM8	1527	01:26	265.78	0.33 ± 0.09
DIM18	1527	06:28	265.91	5.55 ± 2.06
EM9	1579	00:00	298.76	0.23 ± 0.06
DIM19	1579	05:02	298.89	0.94 ± 1.38
EM10	1709	00:00	10.84	0.31 ± 0.09
DIM20	1709	05:16	10.95	-0.56 ± 1.73

SEM stands for standard error of the mean. DIM stands for direct ingest method. EM stands for enrichment method. Adapted from Webster et al. (2018).

and 1,322, TLS-SAM detected three minor spikes of ~2–2.5 ppbv. Another spike (5.55 ± 2.06 ppbv) in methane abundance was noted at sol 1,527 (L_s 265°). Once again, the time variation of abundance between measurements is unknown. There does not appear to be any relationship between the time of day of sample acquisition and the measured abundance, although the confidence in this is low due to the small sample size of observations outside the early morning window.

1.3. Recent Retrievals From Mars Orbit

PFS-MEX recently reported a detection of 15.5 ± 2.5 ppbv of methane above Gale crater (Giuranna et al., 2019), just one sol after the methane spike observed on sol 305 by TLS-SAM. General circulation model (GCM) modeling by Giuranna et al. (2019) suggests that the most likely source of methane is from outside Gale crater ~250 km ESE and SE of the MSL location. This is curious in that locations between the source region and the crater are not also implicated as source regions. If methane from the source region were travelling strictly along the surface, then locations near Gale crater would necessarily also be potential source regions. It could be that the methane in the GCM model is transported upward, then horizontally toward Gale crater, then down into the crater. The spatial resolution of a GCM model may not be sufficient to resolve the complex transport circulations of the crater as we will show below. Of course, a methane detector sitting on the surface has no knowledge of methane abundance aloft. It is only the surface value, not the vertically integrated value, that is relevant for TLS-SAM.

Juxtaposed against all other putative detections, NOMAD (Nadir and Occultation for MArS Discovery; Vandaele et al., 2018) and ACS (Atmospheric Chemistry Suite; Korabev et al., 2018) instruments, both onboard ESA ExoMars Trace Gas Orbiter (TGO), found no methane above their detection threshold of ~0.05 ppbv (Korabev et al., 2019). These instruments were optimized to

search for methane with higher sensitivity than ever before. However, because the theoretically optimal altitude (detection-wise) of TGO measurements is usually found between 15 and 25 km due to atmospheric opacity, being then less sensitive in the lowest atmosphere (<~3-km height), and because many of the measurements were made during the MY34 global dust storm, the nondetection is not definitive. If methane is periodically emitted at the surface of Mars, the TGO measurements strongly suggest that there must be a rapid destruction mechanism. Prior detections could be compatible with the TGO non-detection if strong destruction mechanisms are invoked. Future TGO observations will provide more, and possibly definitive, insights into the Martian methane mystery.

1.4. Methane Sources and Sinks

Assuming only known, slow destruction mechanisms, the observed methane variability implies a methane source (Figure 3). These sources (Yung et al., 2018) could include nonbiological processes such as Fischer-Tropsch type reactions for which H₂ is made available through serpentinization of olivine (Atreya et al., 2007; Oze & Sharma, 2005), geothermal production (Etiope et al., 2011), erosion of basalt with methane inclusions (McMahon et al., 2013), release of regolith-adsorbed gas (Gough et al., 2010; Meslin et al., 2011), exogenous sources including infall of interplanetary dust particles and cometary impact material (Schuerger et al., 2012), biological sources like subsurface methanogen microorganisms (Krasnopolsky et al., 2004), or release of methane from organic decay in solution (Keppler et al., 2012; Poch et al., 2014; Schuerger et al., 2012). The evidence for each of these sources is generally weak or speculative.

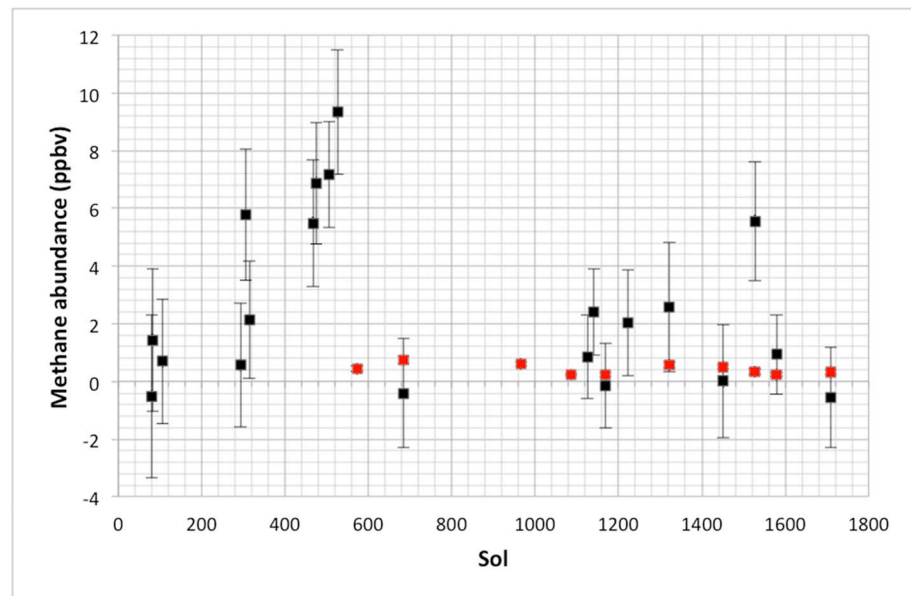


Figure 2. MSL Curiosity rover TLS-SAM methane measurements at Gale crater over an ~4.5 Earth years (~56-month) period (from 26 October 2012 to 27 May 2017) taken during the rover's journey of 16.5 km over highly varied terrain. Black squares represent DIM and red squares represent EM. The time between measurements is insufficient to determine how frequently spikes in abundance occur, how fast they decay, or how common the spikes may be as discussed in section 3.

No known fast methane destruction mechanism exists on Mars, and such a mechanism is difficult to reconcile with the known abundance of other gases that would also presumably be susceptible to the same destruction mechanism (Lefèvre & Forget, 2009), but perchlorates or the intermittent production of peroxides through electrochemical processes in dust devils and dust storms might provide an oxidation source (Atreya et al., 2007). Because there is no known source of strong oxidation for the Martian atmospheric methane other than oxygen, the latter should be depleted in the Martian atmosphere. This effect on oxygen has not been observed, although there are unexplained variations in O_2 (McConnochie et al., 2018) that might provide some clues. Photochemical removal of methane also disrupts the hydrogen chemistry and those effects would presumably be seen in other, more obvious places on Mars (e.g., water, OH, O_2 , O_3 , and CO/ CO_2 abundance) but there is little evidence of such effects. Another analysis shows that methane in the wind can react with the eroded surface quartz grains (abraded silicates) which sequester methane by forming covalent Si-CH₃ bonds and thus an enrichment of the soil with reduced carbon, offering a possible explanation for the fast and selective disappearance of methane on Mars (Jensen et al., 2014).

1.5. Previous Modeling of Methane Transport

Several Mars GCM studies have investigated the putative methane detections. Lefèvre and Forget (2009) employed a GCM with advanced photochemistry to investigate the question of methane destruction time scales. Their simulations show no correlation between the M09 reported spatial patterns of methane variability and the computed spatial patterns of CO_2 condensation. Mischna et al. (2011) concluded that a best match to M09 observations would be found if a nearly instantaneous (rather than gradual), spatially large release occurred no more than 1 to 2 sols before the time of observation (before it could be substantially diluted; Figure 4). This result is consistent with relatively fast atmospheric mixing.

Holmes et al. (2015) showed that the M09 observations could be produced by advection from localized time-dependent sources, but again, a currently unknown methane sink should be invoked in order to explain the spatial and temporal variability of methane observations. The best agreement between the existing observations is found in their simulations using a steady state release from a small source over Nili Fossae. Holmes et al. (2015) suggest that the lower levels of TLS-SAM measurements, as compared to previous results (M09 and Fonti & Marzo, 2010), can be explained by a relative lack of, or indeed complete absence of, methane source emission in the intervening period. Again, this requires a hitherto unknown large methane sink.

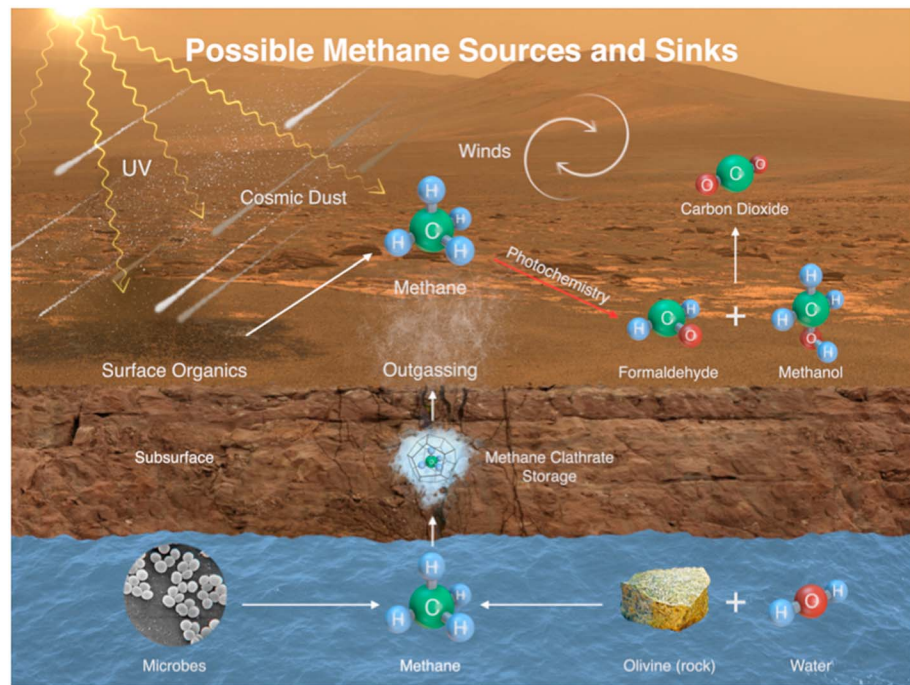


Figure 3. Possible methane sources and sinks on Mars. Image credit: NASA/JPL-Caltech/TLS-SAM-GSFC/University of Michigan.

Viscardy et al. (2016) explored the three-dimensional dispersion of methane throughout the atmosphere after a surface release. Their simulations show that surface emissions of methane result in a nonuniform vertical distribution, including the formation of elevated methane layers shortly after the release. As expected, the distant destination of the released methane is determined by the global circulation pattern at the time of the release, and the methane can be transported to locations over the planet that are far away from the emission source. It typically takes several weeks for the methane to become uniformly mixed, implying that the detection of vertical layers of methane can be an indicator of a recent surface emission. Their finding shows that abundances of methane higher in the atmosphere can be much larger than those measured at the surface where the rover Curiosity is located.

It is challenging to investigate the very localized TLS-SAM methane measurements (both spikes and background levels) using global-scale models. As shown by PGR16, the circulation in and around the ~150-km-diameter Gale crater is highly complex, with strong seasonal and diurnal variations. The expectation is that the distribution of methane in and around the crater will be strongly influenced by these complex circulations. The use of a GCM to represent the large-scale release and dispersion of methane is appropriate, but a GCM cannot capture the transport in and around Gale crater. The diurnal cycle of mixing and flushing of the crater air mass could have an impact on the concentration of methane (and other gases or aerosols) within the crater. The mesoscale circulations driven by the complex topography at the scale of the crater can only be simulated by a model with significantly greater spatial and temporal resolution. The work herein extends PGR16 to investigate the transport and dispersion of methane by regional and crater circulations resolved by a mesoscale model.

The behavior of highly localized releases (on the scale of Gale crater or smaller) or the transport of a larger release by the complex circulations in Gale crater has yet to be fully explored. The Mars Regional Atmospheric Modeling System (hereafter MRAMS; Rafkin et al., 2001, 2002; Rafkin, 2009) is used to simulate, via passive tracers, the transport, dispersion, and mixing of methane released inside or outside of Gale crater from instantaneous or steady state releases, and to test whether the results are consistent with in situ observations made by TLS-SAM. Section 2 provides the reader with a description of the MRAMS configuration and the different methane experiments scenarios performed. This is followed by summary and conclusions in section 3.

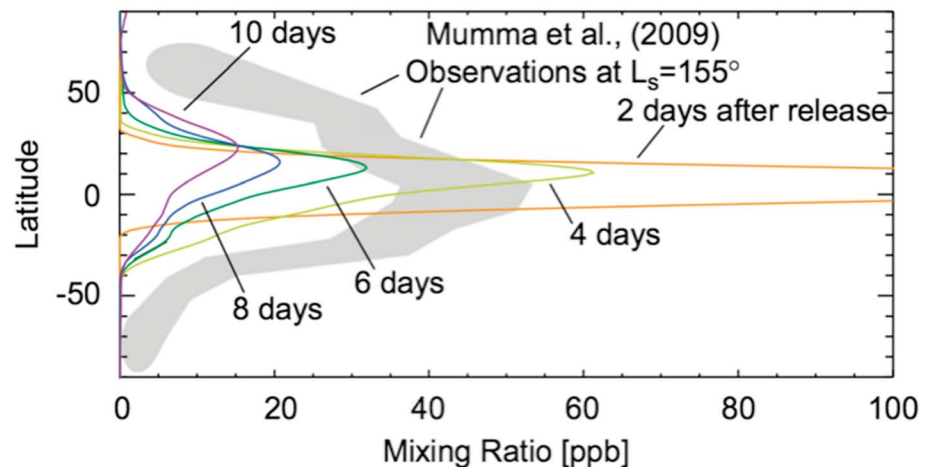


Figure 4. Methane mixing ratio distribution as a function of time for central longitude of 315°W, and $16^\circ \times 10^\circ$ (longitude, latitude) smoothing after an instantaneous (quick episode) release. From Mischna et al. (2011).

2. Mars Methane Modeling Experiments

2.1. MRAMS Configuration

A full description of the MRAMS model configuration for Gale crater (4.5°S, 137.4°E) is included in Figure 1 of PGR16. The horizontal grid spacing at the center of the five grids used in the experiments are 240, 80, 26.7, 8.9, and 2.96 km, respectively, with the innermost grid centered on Gale crater landing site very near (~435 m) where TLS-SAM first detected the methane spikes. The lowest thermodynamic level in all the grids is ~14.5 m above the ground. Ideally, the first vertical level would be located at the height of TLS-SAM (~1 m above the ground), but this is not computationally practical, as described in PGR16. This height difference has little effect into this study because, although the very high stability at night tends to inhibit vertical mixing and might tend to confine methane very near the surface, there is nontrivial mixing and transport by mesoscale circulations, especially in Gale crater, even if the boundary layer eddies are small or ineffective. Slope winds, katabatic flows, mountain waves, and the overall mean flow continuously stir the atmosphere. Any gases close to the crater surface during the night are quickly transported away, and vertical mixing occurs as a consequence of the transport.

The model was run for at least 10 sols for each of the scenarios described in section 2.3. Although the circulation patterns are highly repeatable from sol to sol beginning within a few hours of initialization, the first sol may be regarded as “spin-up.” In order to characterize seasonal mixing changes throughout the Martian year, simulations were conducted centered at L_s 270° and L_s 90°. The selection of these two seasons is based on PGR16. L_s 270° is anomalous in that it is a very windy season with large amplitude breaking mountain waves, and rapid mixing with air external to the crater. The regional northwest winds from the northern lowlands scour the very bottom of the crater floor, and the canonical crater circulation is pushed and extended dramatically to the south. Basically, the internal crater air masses are replaced by air from the northwest. Based on PGR16, L_s 270° was inferred to be the fastest exchange period between air inside and outside the crater. Outside of the L_s 270° season, mixing between the crater air mass and the external crater air was interpreted to be more subdued. L_s 90° was selected as being representative of most of the year for mixing experiments. In addition to these baseline seasons, we conducted another single simulation at L_s 336° motivated by the recent results of Giuranna et al. (2019) that were published during the manuscript review process.

Using the above model configurations, PGR16 demonstrated that the model was able to reasonably reproduce the meteorological observations obtained by the Mars Science Laboratory (hereafter MSL) Curiosity rover REMS instrument (Gómez-Elvira et al., 2012) in Gale crater. The observational biases inherent in REMS winds during many periods, when only winds from certain directions could be measured (Newman et al., 2017), make the comparison with modeled winds in PGR16 challenging. Validating the model wind is difficult because of a lack of reliable wind data overnight and for many times of sol in some seasons. Hence, we do rely to some degree on faith in the model’s ability to represent the essence of the circulation

in the absence of sufficient data validation. The faith is reasonably justified, however, and is not meaningfully different from most Mars modeling studies that similarly lack sufficient data for validation. The REMS pressures and temperatures are well reproduced by the model, and the pressure cycle in particular encodes significant information about the local crater circulation and the mean column temperature (Richardson & Newman, 2018). There are also prior successful studies with MRAMS from which the circulation can be more directly inferred from dust (Rafkin et al., 2002) and clouds (Michaels et al., 2006) and compared to the model results. MRAMS results do compare favorably to the REMS data in periods when the most reliable observational wind data are available (PGR16), and we can always trust fundamental physics to provide further constraints, especially when atmospheric dust loading and surface albedo, thermal inertia, and topography are known.

MRAMS has the capability to simulate the transport of gases as tracers, and this capability is used to represent the transport and mixing of methane. Atmospheric tracers are inert and are transported by advection and eddy diffusion (i.e., subgrid turbulent mixing). Since the photochemical lifetime of methane is thought to be very long (Lefèvre & Forget, 2009) compared to the duration of the simulation, no sinks are imposed on the tracers; there is no destruction mechanism in the model. Tracers in the MRAMS model can be placed anywhere, and may be released instantaneously or at a user-specified, time-dependent rate. Tracers are not radiatively active and do not contribute to the tendency of any model prognostic variables. Like all prognostic variables in MRAMS, tracers are implemented via two-way nesting; values are communicated between the parent and child grids in both directions (Walko et al., 1995). Tracers for the L_s 270° and L_s 90° experiments are strategically placed after the spin-up sol into the highest-resolution domain (grid #5) shown in Figure 1 of PGR16. Tracers for the L_s 336° experiment are strategically placed after the spin-up sol into the grid #4 shown in Figure 1 of PGR16, to better represent the modeling experiment of Giuranna et al. (2019). Tracers released from the same location but with different emission fluxes will evolve identically with abundances in proportion to their source fluxes. In other words, the source flux may be scaled after numerical integration in order to get a proportional answer. The solution for any desired flux magnitude can be obtained from the MRAMS results, regardless of the actual mechanism or initially specified flux rate. Atmospheric transport depends on winds, and these on how the atmosphere is differentially heated driven by topography, albedo, thermal inertia, and physical parameterizations like subgrid-scale mixing and energy exchange with the surface. Also, there is always a subscale which is not resolved by the model. Therefore, results should be interpreted as being one realization of transport which might be expected to differ slightly with slightly different representations of topography and other parameters in the model.

2.2. Martian Clathrate Subsurface Model

The methane emission rate from the subsurface in MRAMS is agnostic of the source mechanism, but since a flux needs to be imposed in the simulations, it is beneficial to utilize an emission rate that is representative of at least one plausible or proposed Martian emission mechanism. Methane clathrates are selected for this purpose out of convenience. Clathrates are a potential reservoir of methane, but they are not a chemical production mechanism. Clathrate hydrates are crystalline compounds composed of cages formed by hydrogen-bonded water molecules inside of which guest gas molecules are trapped. An increase in temperature or a decrease in pressure can lead to the dissociation of clathrates, which results in the release of the trapped gas. Under colder conditions of an earlier climate period (e.g., resulting from obliquity cycles), a cryosphere might trap methane, as clathrates, in a stable form at depth. Under current climate conditions, those same clathrates could become unstable and result in a sporadic release. Previous studies (Chastain & Chevrier, 2007) indicate that the present-day conditions in the Martian subsurface are favorable for the presence of clathrates.

Methane clathrates are widespread on Earth and might be stable also at other places in the solar system including Mars, Titan, and comets. It is very important to emphasize that, despite the numerical modeling and theory supporting their possible existence (Miller & Smythe, 1970), there have been no conclusive observations of CO₂ or methane clathrates on Mars. Since they are not stable at the surface conditions of Mars, only subsurface investigations are likely to provide definitive evidence of their existence.

Karatekin et al. (2016), Karatekin et al. (2017), and Gloesener et al. (2017) (hereafter KG) produced maps of methane-rich clathrate stability zones (Figure 5) obtained by coupling the stability conditions of methane clathrate with a subsurface model. Ancient clathrates may exist at depth where the geothermal gradient

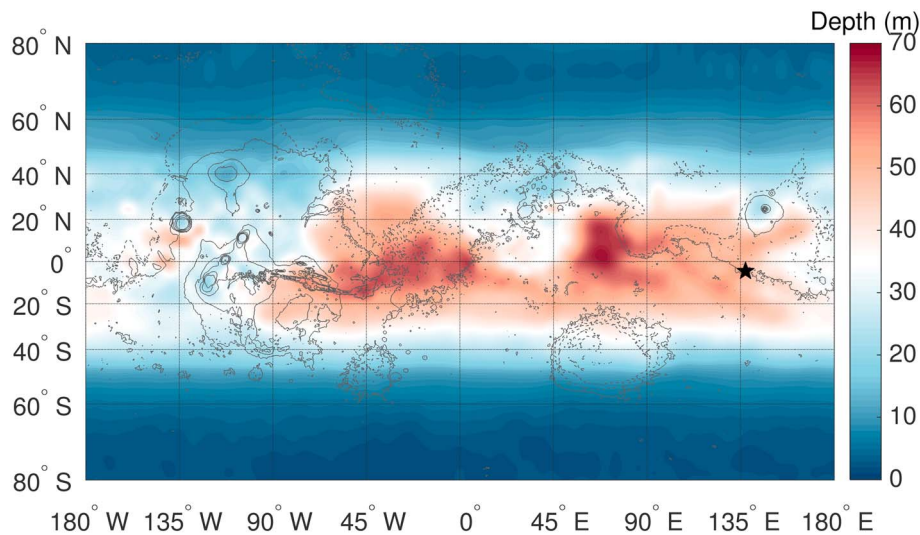


Figure 5. Depth (m) of the beginning of hydrate stability zone in the present-day Martian subsurface for clathrates formed from a gas phase with 90% fraction of methane. Black star is the MSL location. Adapted from Gloesener et al. (2017) and Karatekin et al. (2017).

causes them to decompose over time (Stevens et al., 2017). The regolith properties directly control the subsurface thermal conditions and therefore the depth of clathrate stability; assuming homogeneous values with depth, the lower the thermal inertia at the surface is, the less stable the clathrates will be. This map, based on the mean annual temperature and TES-derived thermal inertia, among other variables, does not show local-scale variations. In addition, some metastable clathrate reservoirs could be locally closer to the surface than would be inferred from this map. Indeed, at the time period of Gale crater formation, the methane clathrate stability zone was probably different with stability conditions possibly met shallower in the subsurface. After the crater formation, clathrates may have formed in the sediments of the lake and below depending on methane availability. With the evolution of Mars and changes in surface conditions, clathrate hydrates at the surface and those excavated would have been dissociated while those remaining isolated from the atmosphere could have persisted until the present day. Therefore, some clathrates at Gale crater could be outside their current stability zone, that is to say, at depths shallower than 45–50 m deep, and in a metastable state.

Methane clathrates can be stable very near the surface at high latitudes, and can be as close as 20 m to the surface in the tropics under today's climate. In the cases where a surface flux of methane is specified in the MRAMS simulations, the flux is assumed to come from subsurface methane clathrate emplaced in earlier geological times, which has been destabilized due to changes in the regolith energy balance. Obliquity changes should have dissociated a large part of methane clathrates; however, Root and Madden (2012) have shown that some of these clathrates may be preserved as metastable reservoirs over geologic time scales due to slow dissociation and diffusion rates. These reservoirs could provide a long-term release of methane in the atmosphere without any current addition of methane in the reservoir. On the other hand, if methane is produced continuously in the present subsurface (via serpentinization followed by Fischer-Tropsch reactions, for example), the base of the Martian cryosphere could be gradually enriched in small amounts of methane clathrate hydrate and the methane diffusion through the overlying ice could expand the reservoir with time. Therefore, methane clathrates could eventually form near the surface at the top of their stability zone, although formation rates at these shallow depths would be significantly slower than those at the base of the hydrate stability zone (Gainey & Madden, 2012).

KG calculated the surface methane flux by modeling methane gas transport through the regolith to the surface via molecular and Knudsen diffusion and assuming an atmospheric pressure profile in the subsurface. Gas adsorption processes are ignored in determining the methane flux used in the MRAMS experiments. Including adsorption reduces the methane flux by a factor of ~ 30 (Meslin et al., 2011), although it increases the emission time by the same amount, amplifying seasonal variations of background methane through

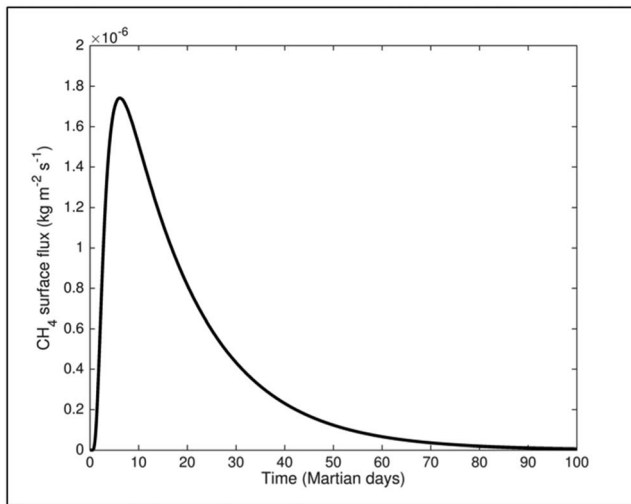


Figure 6. Methane flux for clathrates formed from a gas phase with 90% of methane derived from Gloesener et al. (2017) subsurface diffusive model that includes molecular and Knudsen diffusion.

Arrhenius dependency. When adsorption of methane is taken into account, the transport equation depends on the adsorption and desorption rate constants, and the resulting rate constant can be estimated following the Arrhenius equation. Accordingly, the amount of methane that will be adsorbed or desorbed will change following temperature. When temperature increases, the adsorption process is less important and the surface flux of methane is slightly larger. When temperature decreases, more methane is adsorbed and the surface flux declines in response (Gough et al., 2010; Meslin et al., 2011; Moores et al., 2019). KG calculated the relative abundances of methane in mixed CO_2 - CH_4 - N_2 -Ar clathrates by considering several initial abundances of methane in the gas phase. For example, the dissociation of 1 m^3 of clathrates formed from a gas phase containing 90% methane, ~9% CO_2 , ~1% N_2 , and 0.16% Ar at a depth of 45 m in Gale crater (assuming a mean thermal inertia of ~365 for the few first meters and increasing with depth due to compaction) produces up to $\sim 1.8 \times 10^{-6} \text{ kg m}^{-2} \text{ s}^{-1}$ of methane at $L_s 285^\circ$ (Figure 6) during the first few sols. The determination of these surface fluxes takes into account the diurnal temperature cycle. It is important to note that, although the methane flux should be higher during warmer seasons

due to the dependence of the diffusion coefficient with temperature (and with the kinetic constant if adsorption is taken into account), a constant value is used for all the seasons modeled with MRAMS ($L_s 90^\circ$, $L_s 270^\circ$, and $L_s 336^\circ$), and so the methane flux in our simulations might be overestimated for $L_s 90^\circ$, because Gale crater at this time of the year is still relatively cool compared to the specified temperature. Also, and in contrast to Figure 6, the same constant value of $\sim 1.8 \times 10^{-6} \text{ kg m}^{-2} \text{ s}^{-1}$ over a period of 12 sols is used in our MRAMS experiments. Also, it is important to note that there are no reasonable values of adjustable parameters within the KG clathrate model that could increase the flux by 2 orders of magnitude. KG results are sufficiently robust that they fall within an order of magnitude.

2.3. MRAMS Methane Experiment Scenarios

Different MRAMS tracer scenarios were constructed for the three different seasons ($L_s 90^\circ$, $L_s 270^\circ$, and $L_s 336^\circ$), as shown in Table 3. Instantaneous release scenarios are designed to quantify the rate of mixing within the crater and between the crater and air external to the crater. The release scenario is instantaneous, because the tracer(s) appear instantaneously within a specified region of the model and no additional sources or sinks are provided. In contrast, steady state release scenarios explore the transport of methane under specific constant flux scenarios, locations, and areal extents of the emission. All the scenarios, both the instantaneous and steady state releases, invoke a release at 0500 Local Mean Solar Time (LMST) by default, but two additional instantaneous scenarios with methane release inside Gale crater ($L_s 90^\circ$ and $L_s 270^\circ$) were also conducted with a release at 1700 LMST to test the influence of the release time. It is important to note that the rover is slowly moving and drove less than 3 km (roughly a point in the grid #5 of the model) between spikes of sols 305 ($L_s 336^\circ$) and 525 ($L_s 81^\circ$). For this reason and for simplicity, methane in all MRAMS scenarios was arbitrarily sampled in the model at the MSL location for sol 305 (4.63°S , 137.4°E), when a spike in methane abundance was first noted.

Before exploring the details of the simulations, it is instructive to view the animations provided in the supporting information in order to gain an appreciation for the complexity of the transport and diffusion of methane emitted at various locations. The animations reveal a highly dynamic atmosphere that transports and mixes methane in complicated ways in all three dimensions. A primary take away from these animations should be that the methane concentration is strongly dependent on where you are, when you observe, and the location from where methane is being released.

2.3.1. Instantaneous Methane Release Scenarios

The goal of the instantaneous methane release experiments is to quantify how different air masses within and outside the crater mix. There are two types of crater mixing experiments in which four tracers are strategically placed after 1 sol into the highest-resolution domain showed in Figure 1 of PGR16 (grid 5): a first

Table 3
MRAMS Methane Instantaneous and Steady State Release Scenarios for Inside and Outside Gale Crater Release Locations at L_s 90°, L_s 270°, and L_s 336°

Methane release	Area of emission and location	Type of emission/season simulated	
		Instantaneous emission	Steady state emission
Inside Gale crater	Small size (~150 km ²) emission(s) close to MSL	L_s 90° and L_s 270°	L_s 90° and L_s 270°
Outside Gale crater	Medium size (~6,400 km ²) emission NW from Gale crater	L_s 90° and L_s 270°	L_s 90° and L_s 270°
	Medium size (~6,400 km ²) emissions NE-SW-SE from Gale crater		L_s 90° and L_s 270°
	Large size (~55,000 km ²) emissions ESE-SE from Gale crater		L_s 336°
	XL size (~2,000,000 km ²) emission at M09 (SM)		L_s 270°
	XXL size (~8,000,000 km ²) emission at M09 (TS + NF + SM)		L_s 90° and L_s 270°

All the methane abundances were sampled at the grid point corresponding to the location of MSL for sol 305 (4.63°S, 137.4°E). All the scenarios, instantaneous and steady state release, invoke a release at 0500 Local Mean Solar Time (LMST) by default, but two additional instantaneous scenarios with inside Gale crater methane release (L_s 90° and L_s 270°) were also conducted with a release at 1700 LMST in order to explore whether the timing of the release makes any difference. TS, NF, and SM stand for Terra Sabae, Nili Fossae, and Syrtis Major.

type with an instantaneous methane release inside Gale crater (less than 3 km) west of the MSL landing location and a second type instantaneous methane release ~100 km northwest—upstream of the MSL landing site outside the crater. There are no additional sources (i.e., no flux) or sinks of tracers. The tracer configuration for both instantaneous release scenarios is shown in Figure 7.

The amount of mixing can be diagnosed by evaluating the fraction of a given tracer mixing ratio compared to the total as a function of time for the tracer configuration shown in Figure 7. For example, 100% of the tracers in the bottom (<200 m high) of the crater are tracer #1 when the tracers are first instantiated, because there has yet to be any mixing. If at some later time it is found that 50% of the tracers in the bottom of the crater are tracer #1, then half of that original air mass has been mixed away. Likewise, if 40% of the total tracer mixing ratio at the bottom of the crater is tracer #4, then it can be inferred that 40% of that air mass originated from outside the crater. By looking at the fraction of other tracers, the amount of mixing with each of the different air masses can be determined.

In the first type of instantaneous methane release scenario (Figures 8 and 9), L_s 270° reveals that the air within the crater is replaced by strong, flushing, northerly flow assisted by large-amplitude breaking mountain waves that bring methane-poor air aloft down toward the surface. In the animations found in the supporting information, the flushing is visually similar in appearance to a front sweeping through the crater. At L_s 90°, external air is gradually mixed laterally and vertically into the crater. The tracer #1 fraction is reduced to a few percent or less 5 hr after release at both L_s 270° and L_s 90° (Figures 8 and 9). At 15 hr after the release, tracer #1 is diluted by 5 orders of magnitude from the initial concentration at L_s 90° and by 10 orders of magnitude at L_s 270° (Figure 10). The fraction of external crater air (tracer #4) at the bottom of the crater (replacing internal crater) at 15 hr after the release is 80% at L_s 90° and 100% at L_s 270°. Thus, the mixing of the crater air with the external environment is slower at L_s 90° (and at the other representative seasons, see PGR16) compared to the anomalous L_s 270° flushing season.

The time scale of crater ventilation could be influenced by the local time of tracer emplacement. For example, diurnal upslope winds develop shortly after sunrise, and these might tend to transport air in the crater up the rims and Mount Sharp. Mountain wave activity is also thought to vary over a diurnal cycle (PGR16). A simulation with tracers instantiated at 1700 LMST instead of 0500 LMST tests for this effect. As expected, the details of the tracer fields are different and, although the overall time scale of mixing is similar, it is slightly slower than the 0500 LMST scenario (Table 4). Nighttime downslope flow patterns through crater rims and Mount Sharp might hinder the advection of the internal crater air masses.

Regardless of the season or the time of the tracer instantiation, the simulations indicate that the air mass of the northern crater basin is replaced by external crater air in 1 sol or less (Figures 8–10 and Table 4). These new results are an important update to PGR16; the crater does not appear to be strongly isolated at any time of year. The conflict between the past PGR16 and current results presented here is due to a qualitative estimation of mixing based on potential temperature and wind field patterns in PGR16 versus the quantitative and more definitive approach in this work.

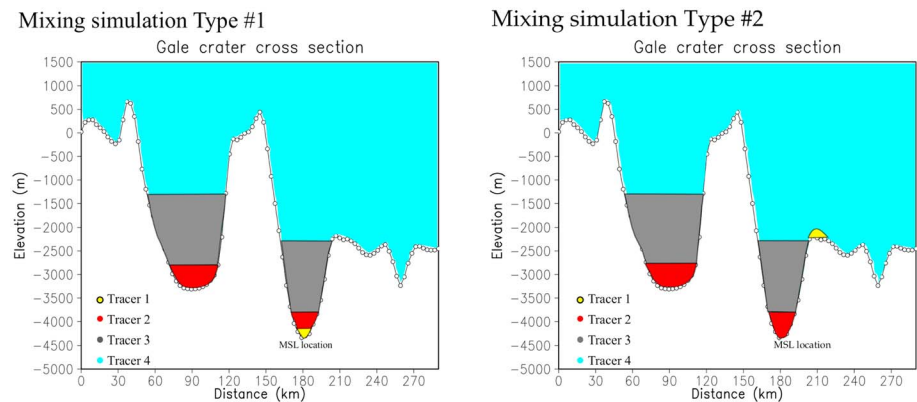


Figure 7. Cross sections through Gale crater show the two types of instantaneous methane release scenarios designed to diagnose crater mixing. The y axis is elevation in meters relative to the MOLA datum. Tracer #1 is designed to represent a hypothetical methane-enriched air mass near the surface (<200 m above the surface). In the first type of mixing experiment (left), tracer #1 covers an area of ~ 150 km² in the very bottom of the northern basin and is placed one grid point (less than 3 km) west of the MSL landing site. Tracer #2 is placed between 200 to 500 m above the lowest point of basin (hereafter ALPOB) at the north basin and between surface and 500 m ALPOB at the south basin. Tracer #3 is placed inside Gale crater from 500 to 2,000 m ALPOB at both basins and tracer #4 elsewhere else in the domain (outside and above Gale crater). In the second instantaneous release scenario (right), tracer #1 is placed outside of Gale crater, covering an area of $\sim 6,400$ km², and is located outside the crater ~ 100 km northwest—upstream of the MSL landing site. Tracer #2 is placed between surface and 500 m ALPOB at both basins and tracers #3 and #4 are as before in the first mixing experiments.

There are only a few methane release scenarios that are consistent with a rapid mixing time scale and a short-period spike in measured methane abundance. The first possibility is a methane burst that serendipitously occurs very close (within hours) to the time of measurement. This cannot be ruled out but, from a statistical perspective, it would require such releases to be fairly common. Of the 30 measurements in Table 2 and Figure 2, one third of them are at or above 2 ppb. Six out of 30 are above 5 ppb. If it is assumed that methane is detectable at or above the level of 2 ppbv or greater for up to 6 hr after a release, and if a release occurred on average once a sol at random local times, then a 25% detection rate would be expected. A 33% detection rate (10 out of the 30) would imply an average frequency of release greater than once every sol. All the additional simulations described in next sections place further constraints on this scenario.

The second scenario that could be consistent with measurements and rapid mixing is that methane originates from outside the crater and is mixed into the crater where it can be measured. This is reasonable to explore since it appears that external crater air reaches the bottom of the crater in less than a day. This second type of instantaneous methane release experiment with tracer #1 outside of Gale crater provides additional insight into the potential transport of methane into the crater from a nearby release. In this scenario, tracer #1 does enter the crater, but at 15 hr after release the concentration is diluted by 6 orders of magnitude from the initial release concentration regardless of the season (Figure 11). The transport is in the right direction, as expected from the prevailing northwesterly surface wind (PGR16), but the tracer is rapidly mixed both vertically and horizontally. The air in the crater is being rapidly replaced by outside air; it appears that a broad, three-dimensional volume of external air is replacing the internal crater air. To achieve a value of 1 ppbv at the rover location, an upwind release of methane on the order of parts per thousand would be required, which is likely unreasonable over the scale of the release area. As with the previous experiment, the release would also have to be fairly frequent to match the observational frequency of observed methane spikes.

If the observed methane peaks are long lived, that is, if the values are elevated for a period longer than the ~ 1 -sol crater mixing time scale, then either a continuous release of methane is required to counteract the mixing and rapid ventilation of the low-level crater air or there must be a release much larger in scale than the crater itself so that mixing time scale between the crater and the outside environment is less relevant. These possibilities are also tested with steady state methane release scenarios described below.

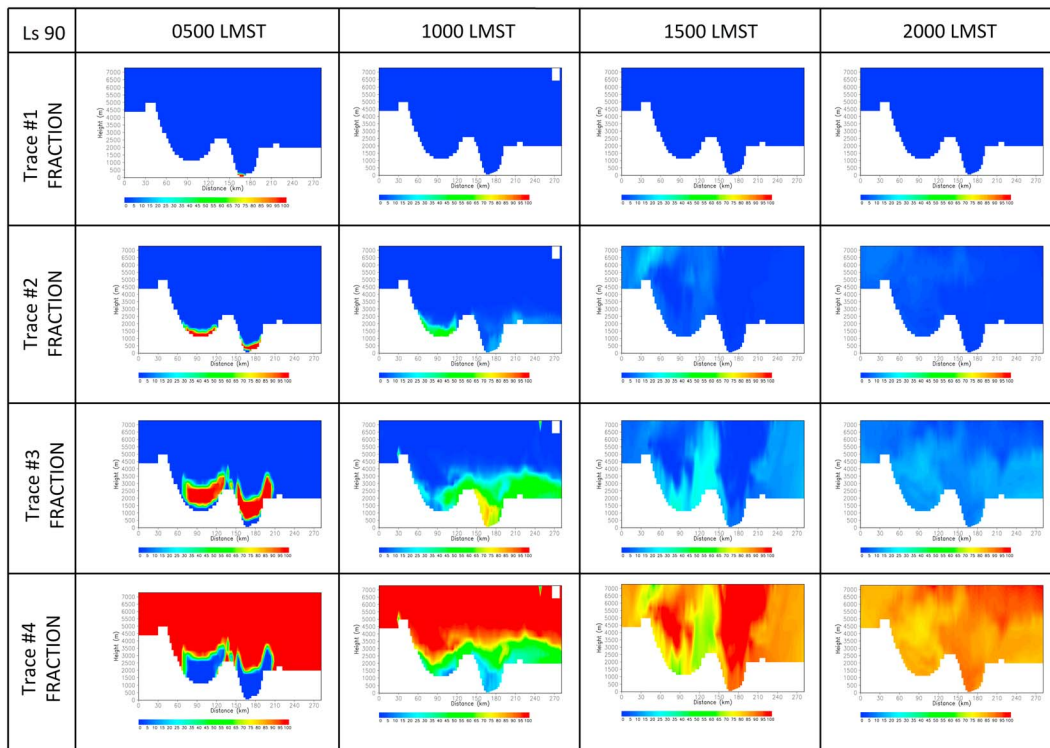


Figure 8. Fraction of the four tracers at four different times (0500, 1000, 1500, and 2000 LMST) at $L_s 90^\circ$ in a north-south cross-section view of the crater for the first type (inside Gale crater) instantaneous methane release scenario.

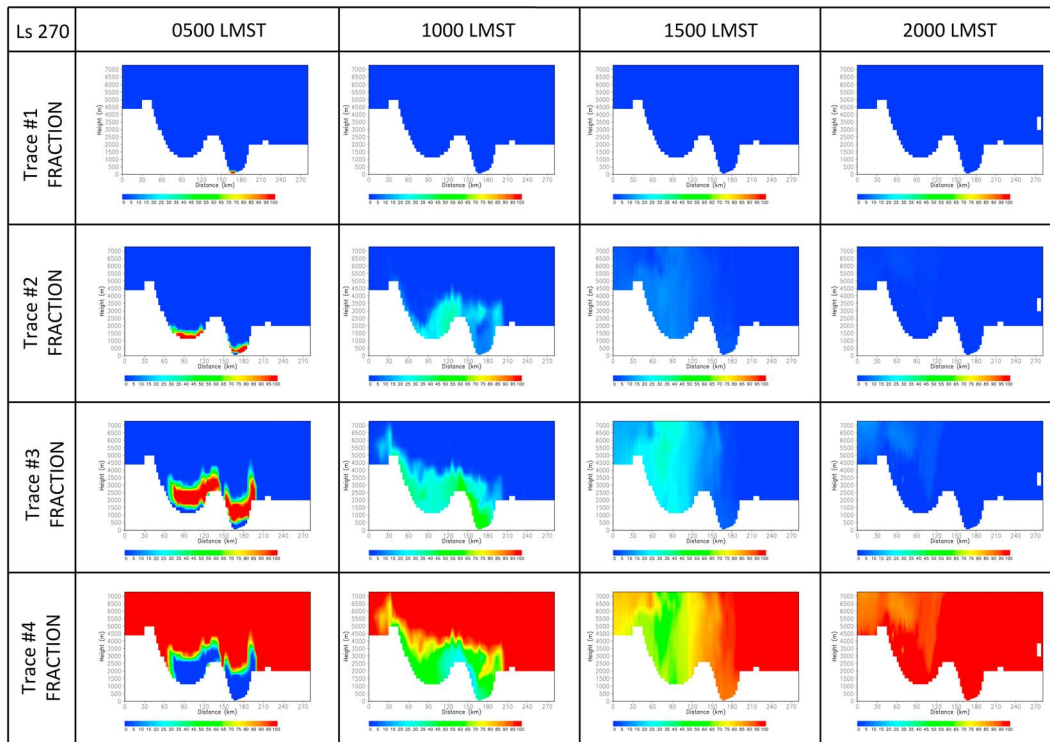


Figure 9. Same as Figure 8 but for $L_s 270^\circ$.

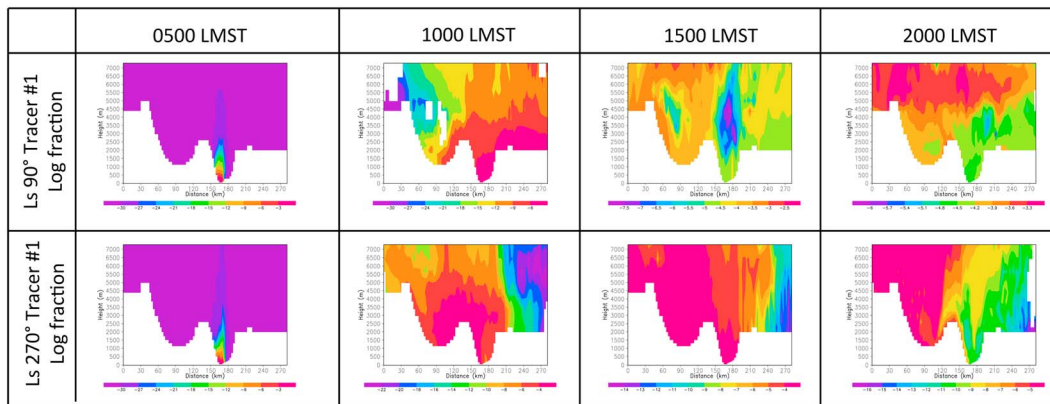


Figure 10. Same as Figures 8 and 9 but for log10(fraction) at L_s 270° and L_s 90° for tracer #1 only.

2.3.2. Steady State Methane Release Scenarios

Although instantaneous releases might be consistent with observations after invoking extraordinary conditions, a continuous release of methane might provide a better explanation. A continuous source of methane could, in theory, counteract dilution by mixing. A continuous release is modeled with a steady state (continuous and constant flux emission over time) flux of $\sim 1.8 \times 10^{-6} \text{ kg m}^{-2} \text{ s}^{-1}$ starting after the 1-sol model spin-up. As previously discussed, any desired flux rate can be achieved by simply scaling the resulting tracer distribution. There are 10 independent methane steady state release sources (tracers A to J; Figure 12). Multiple tracer configurations can be studied simultaneously in a single simulation, because the tracers do not interact with each other.

In the steady state methane release scenarios outside of Gale crater at L_s 270° and L_s 90° ($\sim 10 \text{ km}$ NW, NE, SW, and SE outside Gale crater), modeled abundances at MSL location are ~ 100 times lower compared to TLS-SAM spikes, as shown in Figure 13. Thus, to match the observations, the steady state fluxes would need to be increased by 2 orders of magnitude. A source to the SW (tracer C) at L_s 90° produces the maximum values ($\sim 0.08 \text{ ppbv}$) while effectively no tracer from the southeast (tracer D) makes it to MSL at L_s 270°. At L_s 90°, regardless of the tracer, there is a pronounced diurnal signal with values that are higher during nighttime and lower during the day.

If a continuous methane flux 2 orders of magnitude higher than modeled is plausible, this could provide an explanation consistent with observations. The methane microseepage flux on Earth is generally on the order of 10^{-8} to $10^{-6} \text{ kg m}^{-2} \text{ s}^{-1}$. Increasing this by 2 orders of magnitudes would place it well outside the range of terrestrial microseepage observations. There is no obvious reason to expect that Mars would be nearly as active as Earth. Further, there are no reasonable values of adjustable parameters within the KG clathrate model or other similar models that follow those physics that could increase the flux by 2 orders of magnitude. Other potential subsurface methane sources besides clathrates still require diffusion through the regolith and are subject to the same physics. Thus, the localized release of methane upwind and just outside the crater seems unlikely to explain the peak methane observations.

Recent results by Giuranna et al. (2019) point to a preferred methane source just to the SE/ESE of Gale crater, $\sim 250 \text{ km}$ from the MSL location, at around L_s 336°, which is almost coincident (1 sol after) with one of the TLS-SAM spikes on sol 305. Our results for the L_s 336° season with tracers E and F mimicking release areas E8 and ESE shown in Figure 3 of Giuranna et al. (2019) show that some methane from the E and F emission locations can reach Gale crater, but the abundance predicted by MRAMS is far below ($< 0.1 \text{ ppbv}$) the observed values (Figure 14). It is not surprising that some methane makes it to the crater; a continuous, large areal release will eventually distribute methane everywhere given sufficient time for transport and mixing.

Table 4
Fraction of Tracer at MSL Location 15 hr After the Release

Release time	Tracer/Season			
	L_s 90°		L_s 270°	
	Tracer #1	Tracer #4	Tracer #1	Tracer #4
0500 LMST	10^{-5}	90%	10^{-10}	100%
1700 LMST	10^{-4}	75%	10^{-4}	85%

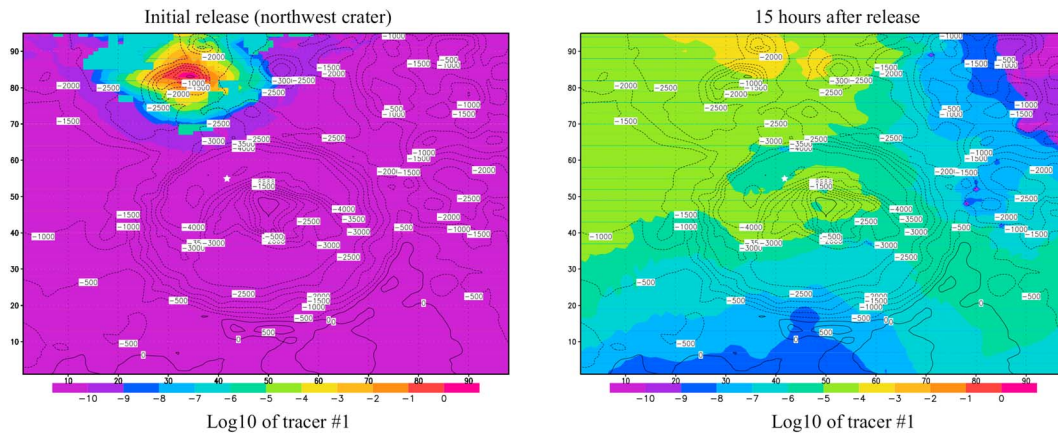


Figure 11. Plan view of tracer #1 (methane) fraction at ~14.5 m high at L_s 90° for the instantaneous release just outside and upstream of the crater (left) for the second type (outside Gale crater) instantaneous methane release scenario. When the tracer reaches the rover location at the surface ~15 hr later (right), the concentration is diluted by approximately 6 orders of magnitude regardless of the season. Similar behavior is observed at L_s 270°. The x-y axis labels stand for grid point (~3 km/grid point). White star is the MSL Curiosity rover location.

Looking to the wind field (Figure 15), the horizontal winds are from the north-northeast most of the time, except in the morning (0600–1200 LMST) when they are toward the west, which is in general agreement with of the Giuranna et al. (2019) modeling results. It is during the 0600–1200 period during which one might expect methane from the release area to be advected directly into the crater. There is a tendency for that to occur, but the appropriate wind direction is not sustained for a long enough period, and whatever methane is horizontally advected is simultaneously being transported vertically and diffused.

Further analyzing the three-dimensional behavior of the atmospheric circulation (supporting information) reveals that the more typical northerly surface winds advect the methane to the south, where it is then transported vertically into a layer of easterly winds between 7 and 15 km above ground level. It is aloft where methane is most easily transported to Gale crater, and indeed, some amount of methane does so.

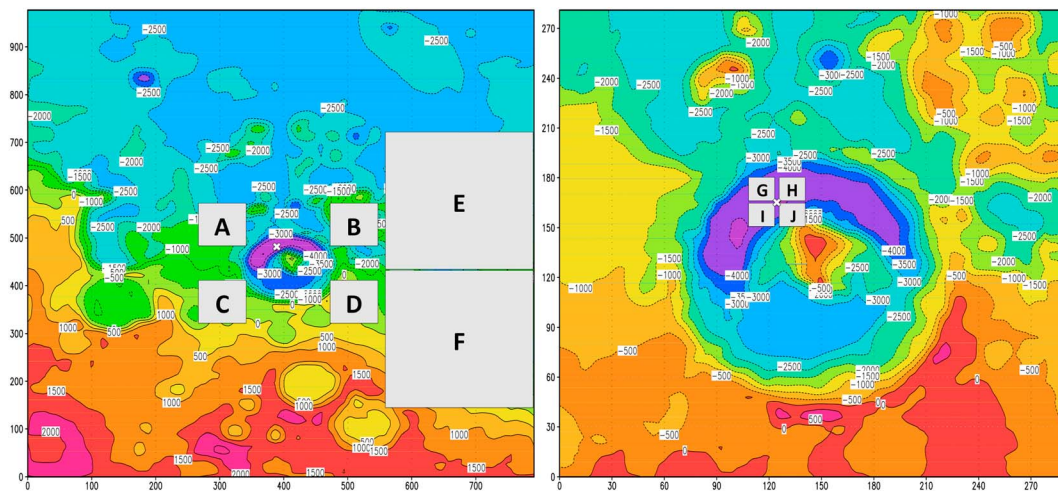


Figure 12. Steady state methane release scenarios outside (A to F, left) and inside (G to J, right) Gale crater. Topography is shown as color-coded elevation (m) from the Mars Orbiter Laser Altimeter (MOLA). White cross is the MSL Curiosity rover location. The x-y axis labels distance in km. All tracer release areas are square-shaped. Four of the release areas (tracers A to D, seasons L_s 270° and L_s 90°) are located outside the crater ~10 km NW, NE, SW, and SE of the crater rims, each with an area of ~6,400 km². Two of the release areas (tracers E and F, season L_s 336°) are located outside the crater ~250 km ESE and SE of the rover location, each with an area of ~55,000 km² in order to represent the Giuranna et al. (2019) experiment. Four of the release areas (tracers G to J, seasons L_s 270° and L_s 90°) are located inside the crater ~1 grid point NW, NE, SW, and SE of the rover location, each with an area of ~150 km². Tracer G is similar in size and in release location to tracer #1 in the first mixing experiment scenario.

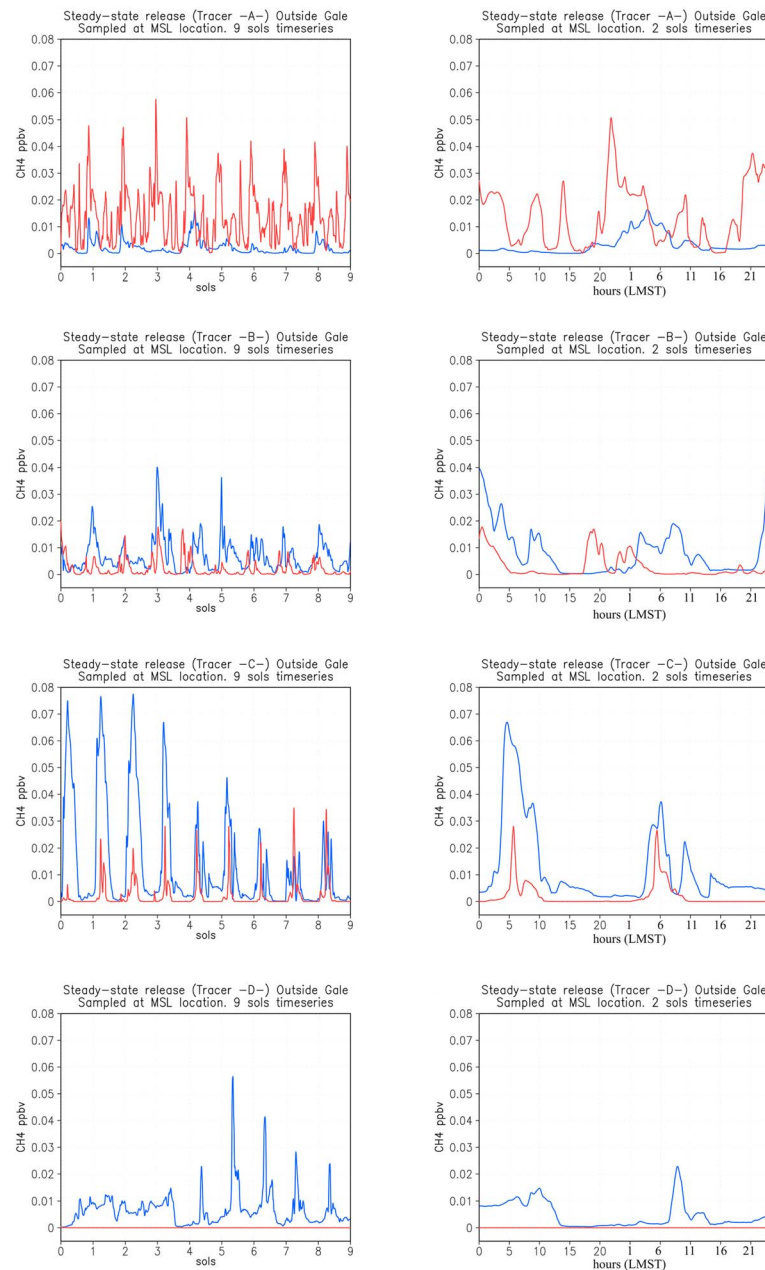


Figure 13. (left panels) Nine-sol and (right panels) two-sol (spanning sols 3–4 of the left panels) time series of MRAMS methane abundances sampled at MSL location for four steady state release locations outside Gale crater (tracers A to D) described in Figure 12. Blue is L_s 90° and red is L_s 270°. The abundance of tracers is shown shortly after the flux is turned on.

However, during the transit from the source region to a location above Gale crater, the air undergoes substantial mixing and diffusion such that the net abundance of methane at the surface of the crater ends up being quite low (Figures 14 and 15). Interestingly, it is during the first sol (i.e., spin-up) when the highest methane abundances are modeled in the crater. Later sols have abundances even lower than 0.1 ppbv (Figure 14). As suspected, methane does not move only along the surface from one location to another. Correctly representing the mesoscale circulations in and around the crater is likely critical to correctly modeling the complex transport and diffusion scenario. Therefore, it is necessary to utilize a model that resolves the topography and the associated circulations.

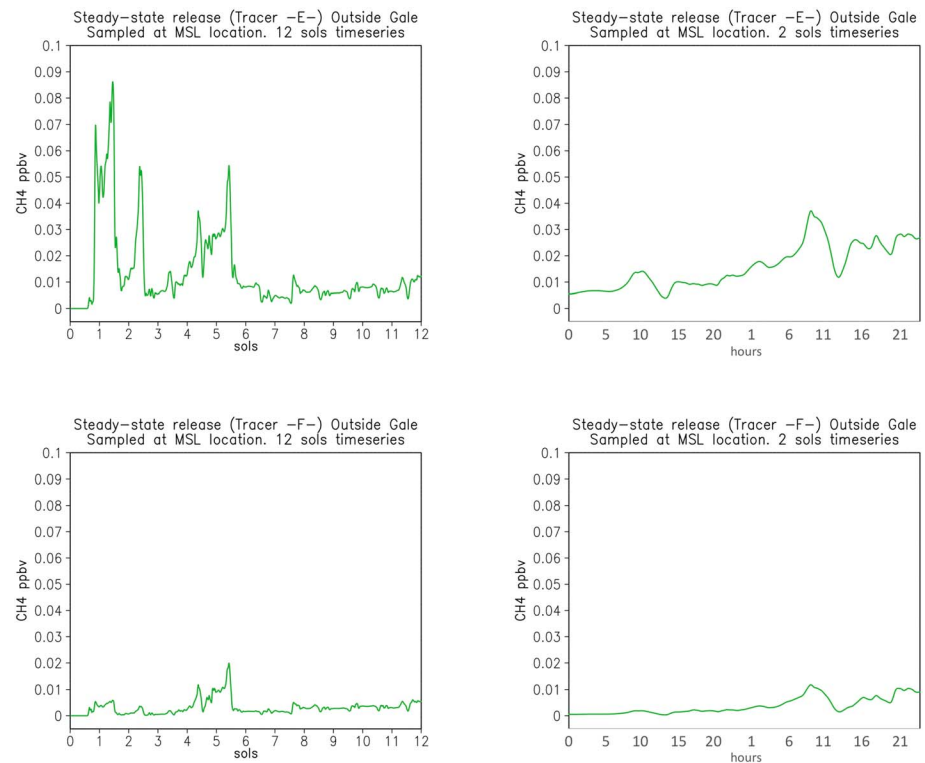


Figure 14. (left panels) Twelve-sols and (right panels) two-sols (spanning sols 3–4 of the left panels) of MRAMS methane abundances sampled at the MSL location for two steady state release locations outside Gale crater (tracers E and F) at L_s 336°, as configured in Figure 12.

In the steady state release experiment inside Gale crater, the methane values at the source location fluctuate from 0.1 to ~1 ppbv (Figure 16; tracer H during L_s 270°). This is comparable to the TLS-SAM low background methane abundances, but is still ~1 order of magnitude lower than the methane spikes. Values are smaller at neighboring grid points. Peak values are modeled during the early morning hours just before sunrise at L_s 90°, and during the evening at L_s 270°. Most of the TLS-SAM sampling times are typically in the early morning, which is coincidentally beneficial for detection except during the L_s 270° wind season. Yet, one of the highest peaks (9.34 ppbv) was measured at L_s 81° during one of the rare afternoon ingests, which is definitely not consistent with the modeled diurnal cycle at any season.

To match with spike observations, the emission flux rate for the continuous release inside the crater would need to be increased by almost an order of magnitude. As previously noted, the imposed emission rate is already near a physical maximum, but it might not be entirely reasonable to reject a five to tenfold increase in flux given the unknowns. The scenario would still require methane to be venting continuously approximately one third of the time in order to reproduce the observed frequency of detection, and the limited observations outside of the early morning window do not match with the modeled diurnal cycle. Consistency between the model and observations also requires that the release be very near the rover. Moving another grid point away (~3 km) reduces the peak model values to levels entirely inconsistent with the observed peaks. Thus, either the moving rover is extremely fortunate to operate within a few kilometers of an active vent in Gale crater, or there must be many active vents such that the rover is reasonably likely to be near one at any given time. The first option is extremely difficult from a statistical standpoint. The second option has implications for the contribution of Gale crater to the overall global methane budget.

Gale crater is a special place, but it is unlikely to be unique. If Gale crater is emitting methane at many locations, there are likely to be other places on Mars doing the same. If the surface fraction of the planet

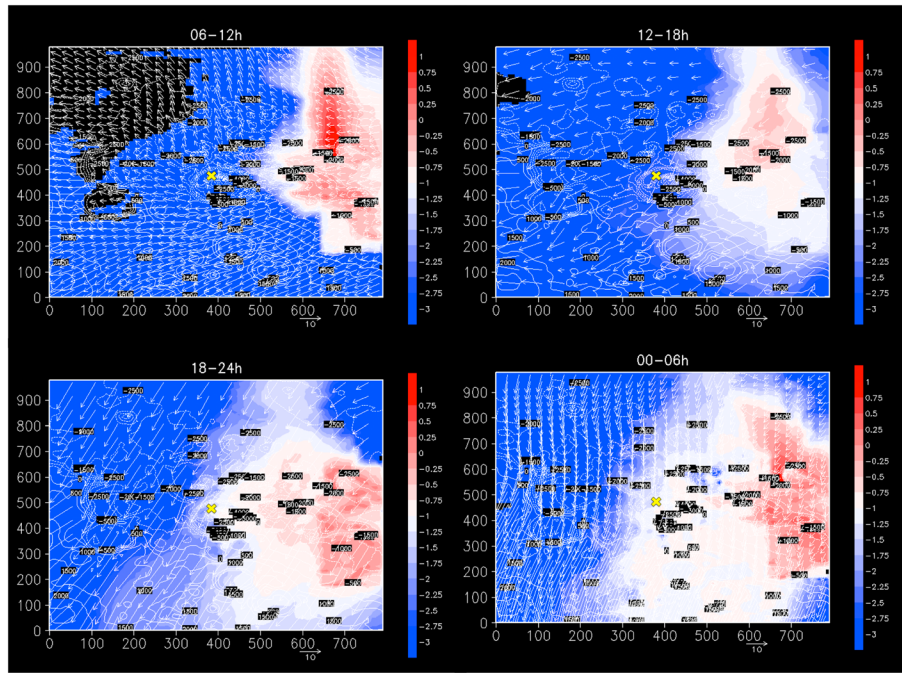


Figure 15. Plan view of tracers E + F (log10 scale bar) and horizontal winds at $L_s 336^\circ$ for four periods of time: 6–12, 12–18, and 18–24 hr of sol 0 and 0–6 hr of sol 1 shown in Figure 14. Tracers are shown at ~ 14.5 -m height, since it is the surface abundance that matters for TLS-SAM. Black empty areas represent values below the lowest color scale. Winds are averaged, as in Giuranna et al. (2019), below 5 km in order to characterize the general boundary layer advective flow direction. For clearness, wind vectors are plotted at every third model grid point. Topography is contoured (m). The x-y axis labels show distance in km. Yellow cross is the MSL location.

equivalent to Gale crater, A, is emitting methane at the rate F, then the rate of increase in global molar fraction of methane, q , is

$$\dot{q} = \frac{M_{\text{CO}_2} F A g}{M_{\text{CH}_4} p_s} \quad (1)$$

where g is the gravitational acceleration, p_s is the global average surface pressure (~ 600 Pa), and M is the molecular weight of either methane or the bulk atmosphere (assumed to be pure CO_2 for this calculation). Using $F = 10^{-6} \text{ kg m}^{-2} \text{ s}^{-1}$ and $A = 0.0005$ in equation (1) evaluates to $\sim 0.85 \times 10^{-2} \text{ ppbv/s}$. Thus, within 600 s and in the absence of a rapid sink, the emission would be the equivalent of ~ 5 -ppbv increase in global methane abundance.

Clearly, any scenario that invokes a widespread release of methane in Gale crater would necessarily contribute to a detectable increase in global background methane concentration in the absence of a strong sink. This necessary increase in background methane has not been observed by MSL, and is incompatible with the nondetection of methane by TGO. Therefore, the model results for a continuous release nearby MSL are compatible with the TLS-SAM low background methane detections only if the MSL landing location within Gale crater is truly special and MSL was fortunate to land there and not even a few kilometers away.

In the steady state methane release inside Gale crater scenarios, methane increases during the evening and night, and decreases during the daytime (Figures 16 and 17). This can be explained by the diurnal, mostly horizontal, slope flow patterns. During nighttime, the downslope winds from crater rims and Mount Sharp converge and constrain methane to the very bottom of the crater, causing it to persist and become trapped for a longer period close to the point where it is released, whereas during daytime the upslope winds transport and vent the methane away (Figure 17).

Figure 17 shows that mixing is strongly influenced by horizontal motions that can transport air into and out of the crater regardless of boundary layer depth. In contrast, Fonseca et al. (2018) indicated that the depth of the PBL is the likely driver of changes in the local dust content of Gale crater, and is also likely to play an

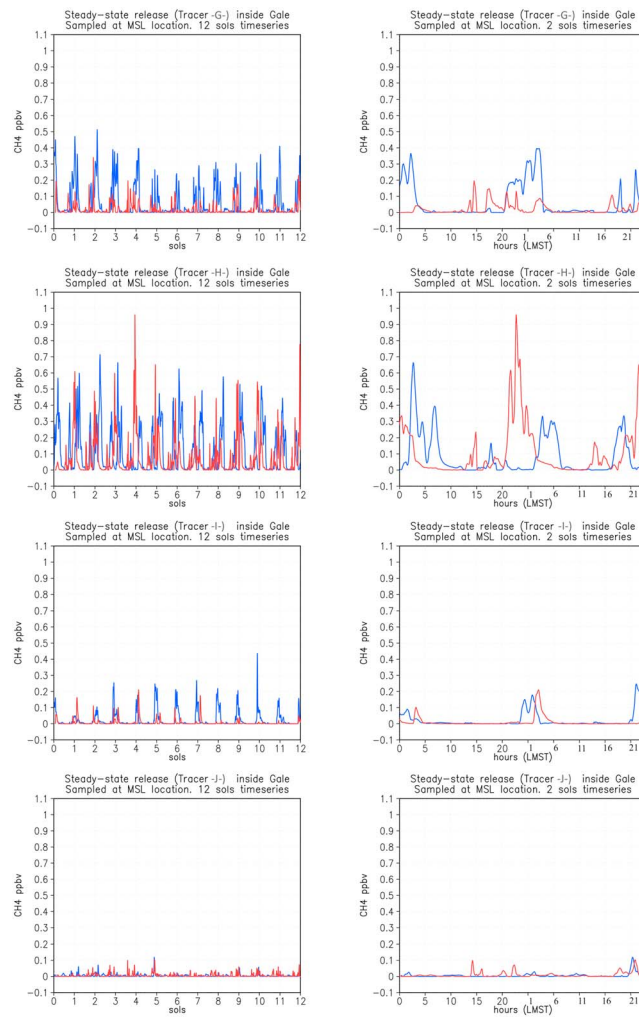


Figure 16. (left panels) Twelve-sols and (right panels) two-sols (spanning sols 3–4 of the left panels) time series of MRAMS methane abundances sampled at the MSL location for four steady state release locations inside Gale crater (tracers G to J) described in Figure 12. Blue is L_s 90° and red is L_s 270°. The abundance of tracers is shown shortly after the flux is turned on.

equally important role in trace gas abundance. Our results show that the growth and collapse of the PBL is not in and of itself necessarily sufficient to explain the diurnal behavior of methane (or dust or water for that matter), and the depth of the PBL is highly variable over the complex topography of the crater. In a one-dimensional view, there is little doubt that the convective motions during the day will tend to mix methane deeper into the lower atmosphere while keeping the gas more confined at night. Yet as shown in PGR16, even with a very shallow and stable boundary layer, horizontal winds are capable of sweeping through the crater and scouring the low-level air. Regardless of what the boundary layer is doing, there is a fairly rapid horizontal exchange and deep vertical exchange above the PBL associated with topographic venting (Rafkin, 2012), and that external air will carry its own methane, water, dust, and other trace gases. Based on modeling results, the atmospheric circulation at Gale crater is strongly 3-D, not just 1-D or 2-D, and any scenario describing the transport of methane must recognize this dimensionality. The source location of methane emission cannot be determined by 2-D dispersion, and variations of methane concentration cannot be determined by simply considering 1-D vertical mixing based on PBL height. Further, because of the complexity of the circulation, the local horizontal (2-D) wind speed and direction at the rover location may not be representative of the larger prevailing wind field. Consequently, trying to determine the source location of methane based only on REMS horizontal wind estimates at the time of

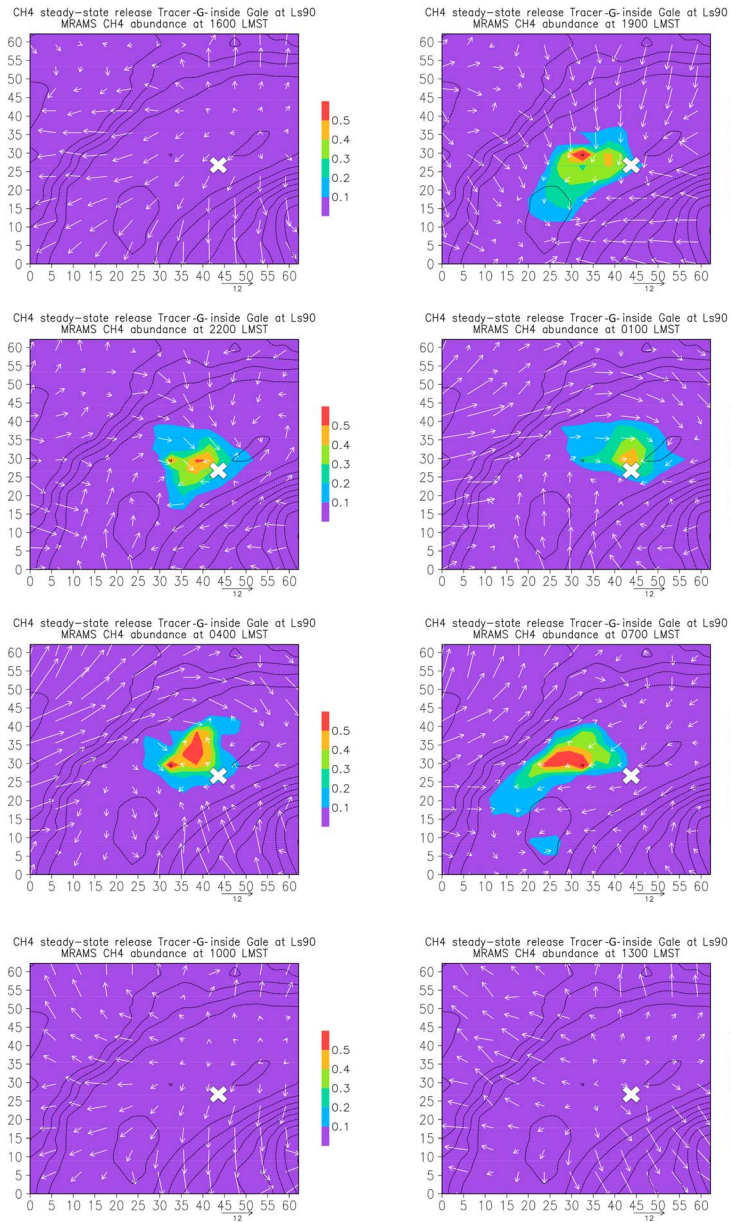


Figure 17. Plan view of methane mixing ratio in the lowest model layer for a (clockwise from 1600 LMST to 1300 LMST) 21-hr time series steady state methane release at $L_s 90^\circ \sim 1$ grid point northwest of the MSL location on sol 305. White arrows represent wind speed and direction. Black contours represent topography. The methane steady state release began at 0500 LMST the sol before. The x-y axis labels distance in km. White cross is the MSL location for sol 305.

the TLS-SAM measurements or trying to explain putative suppressed mixing with a PBL suppression (1-D) is a dubious proposition. It is even conceivable that methane spikes could be due to the downward vertical transport of material from an elevated methane layer produced far away, which would have little to do with the local surface winds.

The last set of experiments to consider assumes a large-scale release similar in area and magnitude to M09. Rather than small, nearby releases as presented in the previous simulations, perhaps regional releases are compatible with TLS-SAM background measurements. Two experiments were performed based on the M09 detections: (1) A “full” M09 release area ($\sim 8,000,000 \text{ km}^2$) source at Terra Sabae (A in Figure 1), Nili Fossae (B_1 in Figure 1), and Syrtis Major (B_2 in Figure 1) and (2) a “partial” M09 release source only from the Nili Fossae area ($\sim 2,000,000 \text{ km}^2$).

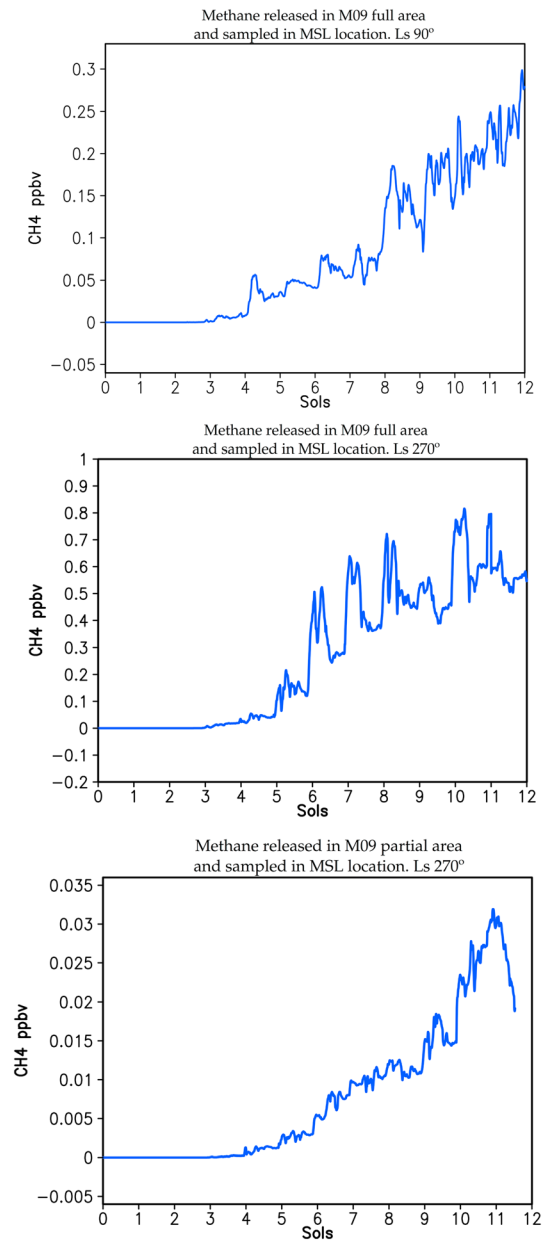


Figure 18. Twelve-sol time series of MRAMS methane abundances sampled at the MSL Curiosity rover location for the steady state methane emission scenario corresponding to the M09 detection at (top) L_s 90°, (middle) L_s 270°, and (bottom) over the M09 limited area (B_2 , Syrtis Major) for L_s 270° only.

Figure 18 shows the results for the steady state release comparable in areal extent to the M09 observations. The most obvious signal in the time series is the near-monotonic increase in abundance as the methane is advected and dispersed from the source region. The trend is not entirely unexpected; continually pumping large quantities of methane into a model in the absence of a sink will lead to increasing values (equation (1)). Still, even an area as large as the putative M09 release is insufficient to reproduce the sporadic higher spikes of methane measured by TLS-SAM and is barely able to reproduce the low background values. At L_s 270°, the methane values from the model at the rover location fluctuate from 0.1 to 0.8 ppbv. These values are compatible with TLS-SAM low background methane abundances but are ~12 times lower than TLS-SAM direct ingest detections (high spikes). Thus, a XXL size, M09-like scenario could explain the background values if it were to go on nearly continuously for ~10 sols. An emission of this magnitude, however, would cause an

extremely large and easily detectable increase in background levels over time. Again, observations do not support such activity, and, in the absence of a rapid destruction mechanism, the global background methane values would climb ever higher with each release.

When comparing the M09 full area ($\sim 8,000,000 \text{ km}^2$) release scenario with the M09 partial area ($\sim 2,000,000 \text{ km}^2$) release scenario at $L_s 270^\circ$, results show 30 times higher methane values with the larger release area. Not surprisingly, the areal extent of the source has an impact on the methane, as might be expected, but this provides no improved correlation between the model and the observations. A larger release area produces a large, transient increase in methane values that do not scale linearly with area.

3. Summary and Conclusions

The circulations in Gale crater are extremely complex and local meteorology plays a major role in the transport, dispersion, and distribution of methane emanating from the surface. Global-scale photochemical models are not ideal for studying the complex local transport problem. The tracer capabilities of MRAMS are employed to address the local and regional transport problem in greater detail.

The instantaneous methane release experiments showed rapid mixing and transport of external air into the crater and a rapid replacement of low-level crater air. The time scale of this exchange is less than a sol with some variation as a function of season. $L_s 270^\circ$ had more rapid exchange than $L_s 90^\circ$. This result updates the results of PGR16.

For a limited area with an instantaneous release to the northwest just outside and upwind of Gale crater, methane is diluted by 6 orders of magnitude in less than a sol, again regardless of season. This solution demonstrates that external air is indeed quickly mixed or transported into the crater, but there is also substantial dilution along the trajectory of an air parcel originating just outside the crater and traveling toward MSL. A local release of methane originating outside the crater is highly unlikely to generate even the low background values observed by MSL. Dispersion of the upwind methane plume is simply too great to achieve measurable concentrations within the crater even if the source region is highly concentrated in methane. A local instantaneous release just outside and upwind of the crater can be excluded as a reasonable explanation for the observed methane spikes and very likely for even the lower background values.

The steady state release experiments outside (NW, NE, SW, and SE) Gale crater follow up on the possibility that a nearby continuous, rather than instantaneous, release might work to counteract mixing and maintain a background value of methane, possibly with periodic spikes in concentration. None of the small external release locations were able to produce methane values anywhere close to even the background values (Figure 13). To reach background values, the magnitude of the methane flux would need to be increased by 2 orders of magnitude. This is unlikely for a clathrate source, and unlikely for most any other possible sources (Oehler & Etiope, 2017).

The nearby instantaneous release experiment did produce pronounced and largely repeatable diurnal cycles of methane variation at the MSL location. For the more quiescent seasons representative of $L_s 90^\circ$, the nighttime, convergent downslope flow from the rims and Mount Sharp tend to confine the methane inside the crater. During the day, the divergent upslope flow advects the methane toward and up the crater wall and Mount Sharp, thereby mixing and transporting the methane out of the crater. With an external northwest source release at $L_s 270^\circ$, the convergent/divergent methane signal is overwhelmed by the strong, flushing winds and the signal becomes more erratic. Whatever the origin of the observed methane, the local time of the sampling is likely to be important.

A continuous release inside the crater and near MSL came the closest to achieving the observed peak methane concentration, but values were too low by a factor of 7 or more. Nevertheless, the deficit is small enough to consider a larger flux than modeled, even if it does stretch the constraints on maximum emission rates. In the steady state methane release inside Gale crater scenario, methane increases during the evening and night and decreases during the daytime (Figures 16 and 17). This is again explained by the diurnal slope flow patterns. Also, as previously mentioned, whatever methane is in the crater at night would tend to become vertically confined by the very cold, dense nocturnal inversion, but the PBL depth alone is not sufficient to explain diurnal variations.

There are complications to invoking a nearby continuous release as an explanation. As one moves away from the source, even by a few kilometers (the equivalent of model grid point), the tracer concentration drops dramatically. Clearly, the source cannot follow the rover, so that would mean that each of the observed spikes represents individual, very large magnitude local releases very near the rover. It is not inconceivable that the rover serendipitously happens to be just in the right spot at the right time to measure these brief and large-amplitude events, but that serendipity has additional implications.

Given how infrequent the methane measurements are, nearby release events would have to be fairly common in both space and time for the mobile rover to happen upon even a handful. Every so often (approximately one third of the time), or every few kilometers, a methane release event would need to occur. From the perspective of the model, a methane source would need to be located at every grid point along the rover's trajectory. In other words, Gale crater, or the portion of the crater where the rover is located, would need to be frequently spewing methane. Integrated over time and space, this is likely to add up to a significant amount of methane that would keep the background levels above the observed value even with external mixing of the air. Simple calculations (equation (1)) show that the emission over a fractional area roughly the size of Gale crater for a period of less than an hour would produce a noticeable increase in global background values after mixing. For this reason alone, the continuous emission sources near the rover are excluded as an explanation for the TLS-SAM observations.

Based on the modeled time series of abundances for many of the tracer experiments, the timing of TLS-SAM sample ingestion is likely to be very important. The steady state methane release inside Gale crater shows diurnal methane variations spanning an order of magnitude, increasing during the evening and night, and decreasing during the daytime. Again, most of the TLS-SAM measurements were acquired during nighttime (except on sols 305 13:55 LMST and 525 13:26 LMST) due to thermal requirements of the TLS-SAM sample handling system. Also, in some cases, ingestion takes place over several hours and so the measurements will reflect the variations of methane during the ingestion period. If the rover were parked very near a large-amplitude local release, it may not see the release at all, depending on the time of day. Even for a release outside of the crater, there is considerable variability of the methane abundance over a sol. It is incapable that diurnal time variability is convolved with time and spatial variability of the source, and there is no easy way of unambiguously separating out the independent effects. Are the spikes due to a sudden nearby release or the result of the circulation within the crater acting upon a release? It is impossible to tell. Could there be a large nearby release while the rover measures low values simply because of the time of day or because the wind is blowing from the wrong direction? Absolutely.

The larger regional release simulations inspired by M09 also do not provide explanatory relief. A large methane-enriched air mass upwind of the crater is unable to produce methane concentrations much above 1 ppb. The large release does provide abundances sufficient to quickly produce observed background levels, but that is only until the methane is globally mixed and dispersed. Furthermore, another large release like M09 has yet to be observed (Aoki et al., 2018; Krasnopolsky, 2011; Krasnopolsky, 2012; Villanueva et al., 2013; Webster et al., 2013; Webster et al., 2015; Webster et al., 2018).

Based on the numerous tracer experiments conducted under this study, there is no scenario consistent with any TLS-SAM observations in the absence of a special times or special places argument. This suggests either that the models are wrong or are missing an important physical process, or that the measurements themselves are in error.

Dynamically, the MRAMS model (and other models) appears to be in reasonable agreement with available meteorological observations. There is no reason to suspect that the transport by the models is substantially in error. There is no doubt that the real transport (if it is ever to be measured) will differ in some details, but the overall mixing time scales and dispersion provided by the model is likely to be representative of reality.

It is difficult to reconcile the measured peaks from TLS-SAM and recent PFS-MEX measurements with MRAMS scenarios unless invoking an unknown rapid destruction mechanism from the lower atmosphere before it spreads globally. If such a process exists, then the local release hypothesis becomes much more viable. Short-term spikes in methane abundance could result from nearby and perhaps even ubiquitous releases, but the fast destruction mechanism could keep the global methane abundance within the observed range. An increase in the methane flux by an order of magnitude above what is modeled would still need to be invoked, however. It is possible to implement an ad hoc fast sink to tracers within the model, and this may

be explored in future work. Our attempt at reproducing the Giuranna et al. (2019) results shows that some methane can reach Gale crater, but the abundance predicted by MRAMS is far below the observed values. Correctly representing the mesoscale circulations in and around the crater is likely critical to correctly modeling the complex transport and diffusion scenario. A low-resolution GCM is incapable of resolving the topography and the associated complex transport circulations of the crater, and this could be a primary source of the disagreement. The modeling results are consistent with TGO if no methane is being released. The modeling results are inconsistent with TGO if methane is being released, particularly given the very low detection threshold. Ongoing measurements by TGO should provide definitive constraints on the frequency and magnitude of methane releases, particularly at the global scale. Should TLS-SAM continue to identify periodic methane abundance spikes while TGO finds nothing, very strong and rapid destruction mechanisms or other to-be-determined exotic processes would have to be operating for both observations to remain simultaneously valid.

Acknowledgments

The data used are listed in the references, tables, and figures. To gain appreciation for the complexity of methane emissions, it is recommended to view the atmospheric circulation animations provided in the supporting information. As with all large space missions, the ability to conduct science depends upon the dedication of hundreds of scientists, engineers, and managers. The authors are grateful for the hard work of the MSL team, without whose dedication none of this work could be accomplished. The authors would also like to explicitly thank the MSL science team for their efforts. Additionally, this manuscript benefited greatly from comments and discussions provided by Ashwin R. Vasavada, Christopher R. Webster from NASA Jet Propulsion Laboratory, Timothy Michaels (SETI), Aymeric Spiga (Sorbonne Université, LMD), Alberto G. Fairén (Cornell/Centro de Astrobiología), Lorie Bruhwiler (NOAA), and Cesar Menor-Salván (Georgia Tech). The final manuscript greatly benefited from the important comments of Michael Mischna and one anonymous reviewer. Their questions and suggestions motivated the substantive improvement of the original submission. We are grateful for their insight and the time and care that they took to provide it. This work was supported by the Spanish Ministry of Economy and Competitiveness under contract ESP2016-79612-C3-1-R and by JPL under subcontract 1356597. S. Rafkin is extremely grateful for the support provided by the MSL Project through JPL contract NNN12AA01C. We acknowledge the support of the Belgian Science Policy Office (BELSPO) through the BRAIN-be Program SCOOP and the ESA/PRODEX Program. This work has received funding from the Fonds de la Recherche Scientifique (FNRS) under grant EOS-30442502.

References

- Aoki, S., Richter, M. J., DeWitt, C., Boogert, A., Encrenaz, T., Sagawa, H., et al. (2018). Stringent upper limit of CH₄ on Mars based on SOFIA/EXES observations. *Astronomy & Astrophysics*, 610, A78. <https://doi.org/10.1051/0004-6361/201730903>
- Atreya, S. K., Mahaffy, P. R., & Wong, A. S. (2007). Methane and related trace species on Mars: Origin, loss, implications for life, and habitability. *Planetary and Space Science*, 55, 358–369. <https://doi.org/10.1016/j.pss.2006.02.005>
- Chastain, B. K., & Chevrier, V. (2007). Methane clathrate hydrates as a potential source for martian atmospheric methane. *Planetary and Space Science*, 55(10), 1246–1256. <https://doi.org/10.1016/j.pss.2007.02.003>
- Comolli, L., & Saggin, B. (2009). Mechanical vibrations onboard Mars Express Orbiter detected by the Fourier spectrometer “PFS”, in XX AIDAA Congress (pp. 1–11). AIDAA.
- Encrenaz, T. (2008). Search for methane on Mars: Observations, interpretation and future work. *Advances in Space Research*, 42(1), 1–5. <https://doi.org/10.1016/j.asr.2007.01.069>
- Etiopie, G., Oehler, D. Z., & Allen, C. C. (2011). Methane emissions from Earth's degassing: Implications for Mars. *Planetary and Space Science*, 59(2–3), 182–195. <https://doi.org/10.1016/j.pss.2010.06.003>
- Fonseca, R. M., Zorzano-Mier, M.-P., & Martín-Torres, J. (2018). Planetary boundary layer and circulation dynamics at Gale crater, Mars. *Icarus*, 302, 537–559. <https://doi.org/10.1016/j.icarus.2017.11.036>
- Fonti, S., & Marzo, G. A. (2010). Mapping the methane on Mars. *Astronomy & Astrophysics*, 512, A51. <https://doi.org/10.1051/0004-6361/200913178>
- Formisano, V., Atreya, S., Encrenaz, T., Ignatiev, N., & Giuranna, M. (2004). Detection of methane in the atmosphere of Mars. *Science*, 306(5702), 1758–1761. <https://doi.org/10.1126/science.1101732>
- Gainey, S. R., & Madsen, M. E. (2012). Kinetics of methane clathrate formation and dissociation under Mars relevant conditions. *Icarus*, 218(1), 513–524. <https://doi.org/10.1016/j.icarus.2011.12.019>
- Geminale, A., Formisano, V., & Giuranna, M. (2008). Methane in Martian atmosphere: Average spatial, diurnal, and seasonal behaviour. *Planetary and Space Science*, 56(9), 1194–1203. <https://doi.org/10.1016/j.pss.2008.03.004>
- Geminale, A., Formisano, V., & Sindoni, G. (2011). Mapping methane in Martian atmosphere with PFS-MEX data. *Planetary and Space Science*, 59(2–3), 137–148. <https://doi.org/10.1016/j.pss.2010.07.011>
- Giuranna, M., Viscardi, S., Daerden, F., Neary, L., Etiopie, G., Oehler, D., et al. (2019). Independent confirmation of a methane spike on Mars and a source region east of Gale Crater. *Nature Geoscience*, 12(5), 326. <https://doi.org/10.1038/s41561-019-0331-9>
- Gloesener, E., Karatekin, O., & Dehant, V. (2017). CH₄-rich clathrate hydrate stability zone in the present Martian subsurface. In F. Forget, M. A. Lopez-Valverde, S. Amiri, M.-C. Desjean, F. Gonzalez-Galindo, J. Hollingsworth, B. Jakosky, S. R. Lewis, D. McCleese, E. Millour, H. Svedhem, D. Titov, & M. Wolff (Eds.), *The Sixth International Workshop on the Mars Atmosphere: Modelling and observation* (p. 4209). Granada, Spain.
- Gómez-Elvira, J., Armiens, C., Castañer, L., Domínguez, M., Genzer, M., Gómez, F., et al. (2012). REMS: The environmental sensor suite for the Mars Science Laboratory rover. *Space Science Reviews*, 170(1–4), 583–640. <https://doi.org/10.1007/s11214-012-9921-1>
- Gough, R. V., Tolbert, M. A., McKay, C. P., & Toon, O. B. (2010). Methane adsorption on a martian soil analog: An abiogenic explanation for methane variability in the Martian atmosphere. *Icarus*, 207(1), 165–174. <https://doi.org/10.1016/j.icarus.2009.11.030>
- Holmes, J. A., Lewis, S. R., & Patel, M. R. (2015). Analysing the consistency of Martian methane observations by investigation of global methane transport. *Icarus*, 257, 23–32. <https://doi.org/10.1016/j.icarus.2015.04.027>
- Jensen, S. J. K., Skibsted, J., Jakobsen, H. J., Inge, L., Gunnlaugsson, H. P., Merrison, J. P., et al. (2014). A sink for methane on Mars? The answer is blowing in the wind. *Icarus*, 236, 24–27. <https://doi.org/10.1016/j.icarus.2014.03.036>
- Karatekin, O., Gloesener, E., Dehant, V. M. A., & Temel, O. (2016). Methane clathrate stability zone variations and gas transport in the Martian subsurface, in AGU 2016 Fall Meeting Abstracts. P21B2088K
- Karatekin, Ö., Gloesener, E., & Temel, O. (2017). Clathrate hydrates as possible source of episodic methane releases on Mars, in EGU General Assembly Conference Abstracts (Vol. 19, p. 11000).
- Keppeler, F., Vignani, I., McLeod, A., Ott, U., Früchtl, M., & Röckmann, T. (2012). Ultraviolet-radiation-induced methane emissions from meteorites and the Martian atmosphere. *Nature*, 486(7401), 93–96. <https://doi.org/10.1038/nature11203>
- Korablev, O., Montmessin, F., Trokhimovskiy, A., Fedorova, A. A., Shakun, A. V., Grigoriev, A. V., et al. (2018). The Atmospheric Chemistry Suite (ACS) of three spectrometers for the ExoMars 2016 Trace Gas Orbiter. *Space Science Reviews*, 214(1), 7. <https://doi.org/10.1007/s11214-017-0437-6>
- Korablev, O., Vandaele, A. C., Montmessin, F., Fedorova, A. A., Trokhimovskiy, A., Forget, F., et al. (2019). No detection of methane on Mars from early ExoMars Trace Gas Orbiter observations. *Nature*, 568(7753), 517. <https://doi.org/10.1038/s41586-019-1096-4>
- Krasnopolsky, V. A. (2007). Long-term spectroscopic observations of Mars using IRTF/CSHELL: Mapping of O₂ dayglow, CO, and search for CH₄. *Icarus*, 190(1), 93–102. <https://doi.org/10.1016/j.icarus.2007.02.014>
- Krasnopolsky, V. A. (2011). *A sensitive search for methane and ethane on Mars, EPSC-DPS Joint Meeting 2011* (p. 49). Washington, DC: Department of Physics, Catholic University of America.

- Krasnopolsky, V. A. (2012). Search for methane and upper limits to ethane and SO₂ on Mars. *Icarus*, 217(1), 144–152. <https://doi.org/10.1016/j.icarus.2011.10.019>
- Krasnopolsky, V. A., Maillard, J. P., & Owen, T. C. (2004). Detection of methane in the Martian atmosphere: Evidence for life? *Icarus*, 172(2), 537–547. <https://doi.org/10.1016/j.icarus.2004.07.004>
- Lefèvre, F., & Forget, F. (2009). Observed variations of methane on Mars unexplained by known atmospheric chemistry and physics. *Nature*, 460(7256), 720–723. <https://doi.org/10.1038/nature08228>
- McConnochie, T. H., Smith, M. D., Wolff, M. J., Bender, S., Lemmon, M., Wiens, R. C., et al. (2018). Retrieval of water vapor column abundance and aerosol properties from ChemCam passive sky spectroscopy. *Icarus*, 307, 294–326. <https://doi.org/10.1016/j.icarus.2017.10.043>
- McMahon, S., Parnell, J., & Blamey, N. J. (2013). Sampling methane in basalt on Earth and Mars. *International Journal of Astrobiology*, 12(2), 113–122. <https://doi.org/10.1017/S1473550412000481>
- Meslin, P. Y., Gough, R., Lefèvre, F., & Forget, F. (2011). Little variability of methane on Mars induced by adsorption in the regolith. *Planetary and Space Science*, 59(2–3), 247–258. <https://doi.org/10.1016/j.pss.2010.09.022>
- Michaels, T. I., Colaprete, A., & Rafkin, S. C. R. (2006). Significant vertical water transport by mountain-induced circulations on Mars. *Geophysical Research Letters*, 33, L16201. <https://doi.org/10.1029/2006GL026562>
- Miller, S. L., & Smythe, W. D. (1970). Carbon dioxide clathrate in the Martian ice cap. *Science*, 170(3957), 531–533. <https://doi.org/10.1126/science.170.3957.531>
- Mischina, M. A., Allen, M., Richardson, M. I., Newman, C. E., & Toigo, A. D. (2011). Atmospheric modeling of Mars methane surface releases. *Planetary and Space Science*, 59(2–3), 227–237. <https://doi.org/10.1016/j.pss.2010.07.005>
- Moore, J. E., Gough, R. V., Martinez, G. M., Meslin, P. Y., Smith, C. L., Atreya, S. K., et al. (2019). Methane seasonal cycle at Gale Crater on Mars consistent with regolith adsorption and diffusion. *Nature Geoscience*, 12(5), 321. <https://doi.org/10.1038/s41561-019-0313-y>
- Mumma, M. J., Villanueva, G. L., Novak, R. E., Hewagama, T., Bonev, B. P., DiSanti, M. A., et al. (2009). Strong release of methane on Mars in northern summer 2003. *Science*, 323(5917), 1041–1045. <https://doi.org/10.1126/science.1165243>
- Newman, C. E., Gómez-Elvira, J., Marin, M., Navarro, S., Torres, J., Richardson, M. I., et al. (2017). Winds measured by the Rover Environmental Monitoring Station (REMS) during the Mars Science Laboratory (MSL) rover's Bagnold Dunes Campaign and comparison with numerical modeling using MarsWRF. *Icarus*, 291, 203–231. <https://doi.org/10.1016/j.icarus.2016.12.016>
- Oehler, D. Z., & Etiope, G. (2017). Methane seepage on Mars: Where to look and why. *Astrobiology*, 17(12), 1233–1264. <https://doi.org/10.1089/ast.2017.1657>
- Oze, C., & Sharma, M. (2005). Have olivine, will gas: Serpentinization and the abiogenic production of methane on Mars. *Geophysical Research Letters*, 32, L10203. <https://doi.org/10.1029/2005GL022691>
- Pla-García, J., Rafkin, S. C. R., Kahre, M., Gomez-Elvira, J., Hamilton, V. E., Navarro, S., et al. (2016). The meteorology of Gale crater as determined from rover environmental monitoring station observations and numerical modeling. Part I: Comparison of model simulations with observations. *Icarus*, 280, 103–113. <https://doi.org/10.1016/j.icarus.2016.03.013>
- Poch, O., Kaci, S., Stalport, F., Szopa, C., & Coll, P. (2014). Laboratory insights into the chemical and kinetic evolution of several organic molecules under simulated Mars surface UV radiation conditions. *Icarus*, 242, 50–63. <https://doi.org/10.1016/j.icarus.2014.07.014>
- Rafkin, S. C. (2012). The potential importance of non-local, deep transport on the energetics, momentum, chemistry, and aerosol distributions in the atmospheres of Earth, Mars, and Titan. *Planetary and Space Science*, 60(1), 147–154. <https://doi.org/10.1016/j.pss.2011.07.015>
- Rafkin, S. C. R. (2009). A positive radiative-dynamic feedback mechanism for the maintenance and growth of Martian dust storms. *Journal of Geophysical Research*, 114, E01009. <https://doi.org/10.1029/2008JE003217>
- Rafkin, S. C. R., Haberle, R. M., & Michaels, T. I. (2001). The Mars Regional Atmospheric Modeling System (MRAMS): Model description and selected simulations. *Icarus*, 151(2), 228–256. <https://doi.org/10.1006/icar.2001.6605>
- Rafkin, S. C. R., Pla-García, J., Kahre, M., Gomez-Elvira, J., Hamilton, V. E., Marin, M., et al. (2016). The meteorology of Gale crater as determined from Rover Environmental Monitoring Station observations and numerical modeling. Part II: Interpretation. *Icarus*, 280, 114–138. <https://doi.org/10.1016/j.icarus.2016.01.031>
- Rafkin, S. C. R., Sta. Maria, M. R. V., & Michaels, T. I. (2002). Simulation of the atmospheric thermal circulation of a Martian volcano using a mesoscale numerical model. *Nature*, 419(6908), 697–699. <https://doi.org/10.1038/nature01114>
- Richardson, M. I., & Newman, C. E. (2018). On the relationship between surface pressure, terrain elevation, and air temperature. Part I: The large diurnal surface pressure range at Gale crater, Mars and its origin due to lateral hydrostatic adjustment. *Planetary and Space Science*, 164, 132–157. <https://doi.org/10.1016/j.pss.2018.07.003>
- Root, M. J., & Madden, M. E. E. (2012). Potential effects of obliquity change on gas hydrate stability zones on Mars. *Icarus*, 218(1), 534–544. <https://doi.org/10.1016/j.icarus.2011.12.024>
- Schuerger, A. C., Moore, J. E., Clausen, C. A., Barlow, N. G., & Britt, D. T. (2012). Methane from UV irradiated carbonaceous chondrites under simulated Martian conditions. *Journal of Geophysical Research*, 117, E08007. <https://doi.org/10.1029/2011JE004023>
- Stevens, A. H., Patel, M. R., & Lewis, S. R. (2017). Modelled isotopic fractionation and transient diffusive release of methane from potential subsurface sources on Mars. *Icarus*, 281, 240–247. <https://doi.org/10.1016/j.icarus.2016.08.023>
- Sullivan, W. (1969). Two gases associated with life found on Mars near polar cap, New York Times August 8, 1
- Vandaele, A. C., Lopez-Moreno, J. J., Patel, M. R., Bellucci, G., Daerden, F., Ristic, B., et al. (2018). NOMAD, an integrated suite of three spectrometers for the ExoMars trace gas mission: Technical description, science objectives and expected performance. *Space Science Reviews*, 214(5), 80. <https://doi.org/10.1007/s11214-018-0517-2>
- Villanueva, G. L., Mumma, M. J., Novak, R. E., Radeva, Y. L., Käufel, H. U., Smette, A., et al. (2013). A sensitive search for organics (CH₄, CH₃ OH, H₂ CO, C₂ H₆, C₂ H₂, C₂ H₄), hydroperoxyl (HO₂), nitrogen compounds (N₂O, NH₃, HCN) and chlorine species (HCl, CH₃ Cl) on Mars using ground-based high-resolution infrared spectroscopy. *Icarus*, 223(1), 11–27. <https://doi.org/10.1016/j.icarus.2012.11.013>
- Viscardy, S., Daerden, F., & Neary, L. (2016). Formation of layers of methane in the atmosphere of Mars after surface release. *Geophysical Research Letters*, 43, 1868–1875. <https://doi.org/10.1002/2015GL067443>
- Walko, R. L., Cotton, W. R., Meyers, M. P., & Harrington, J. Y. (1995). New RAMS cloud microphysics parameterization part I: the single-moment scheme. *Atmospheric Research*, 38(1–4), 29–62. [https://doi.org/10.1016/0169-8095\(94\)00087-T](https://doi.org/10.1016/0169-8095(94)00087-T)
- Webster, C. R., & Mahaffy, P. R. (2011). Determining the local abundance of Martian methane and its 13 C/12 C and D/H isotopic ratios for comparison with related gas and soil analysis on the 2011 Mars Science Laboratory (MSL) mission. *Planetary and Space Science*, 59(2–3), 271–283. <https://doi.org/10.1016/j.pss.2010.08.021>
- Webster, C. R., Mahaffy, P. R., Atreya, S. K., Flesch, G. J., & Farley, K. A. (2013). Low upper limit to methane abundance on Mars. *Science*, 342(6156), 355–357. <https://doi.org/10.1126/science.1242902>

- Webster, C. R., Mahaffy, P. R., Atreya, S. K., Flesch, G. J., Mischna, M. A., Meslin, P. Y., et al., & the MSL Science Team (2015). Mars methane detection and variability at Gale crater. *Science*, 347(6220), 415–417. <https://doi.org/10.1126/science.1261713>
- Webster, C. R., Mahaffy, P. R., Atreya, S. K., Moores, J. E., Flesch, G. J., Malespin, C., et al. (2018). Background levels of methane in Mars' atmosphere show strong seasonal variations. *Science*, 360(6393), 1093–1096. <https://doi.org/10.1126/science.aag0131>
- Yung, Y. L., Chen, P., Nealson, K., Atreya, S., Beckett, P., Blank, J. G., et al. (2018). Methane on Mars and habitability: Challenges and responses.
- Zahnle, K., Freedman, R. S., & Catling, D. C. (2011). Is there methane on Mars? *Icarus*, 212(2), 493–503. <https://doi.org/10.1016/j.icarus.2010.11.027>