

Key Points:

- Numerical simulations strongly suggest that the methane measurements by Mars Science Laboratory (MSL) result from a localized source in the northwest interior of Gale Crater
- A localized source in the area of MSL could not be easily detected by Trace Gas Orbiter (TGO), which indeed has not detected methane to date
- Localized sources must be extremely uncommon across Mars to satisfy TGO unless an unknown loss mechanism is invoked

Supporting Information:

Supporting Information may be found in the online version of this article.

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Constraints on Emission Source Locations of Methane Detected by Mars Science Laboratory

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Abstract The Sample Analysis at Mars (SAM) instrument on the Mars Science Laboratory (MSL) Curiosity rover has detected both methane spikes and variable background methane abundance in recent years in Gale Crater, Mars. While methane spikes have been attributed to a hypothetical local or regional source emission, the background measurements acquired during the nighttime were postulated to represent the global methane abundance on Mars. However, recent high-accuracy observations by instruments on the Trace Gas Orbiter (TGO) in several locations around the planet have not detected methane at all, apparently contradicting the SAM measurements. This paper analyzes the constraints that TGO and MSL impose on the hypothetical location of the emission source of methane responsible for the levels detected by SAM. The numerical simulations presented here indicate that not only the spikes but also the *background* measurements performed by MSL must result from localized emissions, specifically in the northwest interior of Gale Crater. Other simulated emission source locations at a greater distance from MSL, even if still within Gale Crater, are difficult to reconcile with current observations by MSL and TGO. Confirming previous studies, these results therefore point either to an improbable scenario, in which the rover has landed close to one of only a few localized emission sources on Mars, or to a problematic scenario, in which an unknown loss mechanism must be invoked that is able to destroy methane orders of magnitude faster than predicted by standard gas chemistry or to a weighted action between both scenarios.

Plain Language Summary The Curiosity rover has detected both a background abundance and transient spikes in methane abundance in recent years in Gale Crater on Mars. These methane spikes were suggested to be produced by a local or regional emission source. On the other hand, the background measurements were originally postulated to be the result of a global methane abundance on Mars. However, recent high accuracy observations by the Trace Gas Orbiter (TGO) have not detected any methane in the Martian atmosphere, apparently contradicting the Curiosity measurements. The numerical simulations presented here indicate that both the spikes and the background measurements performed by the Curiosity would have the same origin in localized methane emissions close to the Curiosity rover. These results simultaneously satisfy the constraints imposed by TGO and Curiosity observations, although they are improbable or problematic: either there exists a strong and unknown loss mechanism in the atmosphere that prevents the accumulation of global methane or methane emissions are extremely uncommon on Mars and the Curiosity rover has fortuitously landed next to one of them.

1. Introduction

The observation of methane in the Martian atmosphere has generated much debate in the scientific community. Methane in the Martian atmosphere was discovered in the first decade of the 2000s (Formisano et al., 2004; Krasnopolsky et al., 2004; Mumma et al., 2009). Observations from the Earth and from Mars orbit have shown strong spatial and temporal variability, including some cases of nondetection (e.g., Formisano et al., 2004; Krasnopolsky, 2012; Mumma et al., 2009; Villanueva et al., 2013). The observed variability of methane is surprising because atmospheric motions ought to homogenize methane given a predicted chemical lifetime of the order of several hundred years (e.g., Atreya et al., 2007; Lefèvre & Forget, 2009; Mumma et al., 2009; Summers et al., 2002; Zahnle et al., 2011). Thus, variability in methane abundance on Mars would, in principle, imply active sources (emissions sites) on the surface, as well as potentially requiring an unknown fast methane loss process (e.g., Hu et al., 2016; Lefèvre, 2019; Webster et al., 2018; Yung et al., 2018). The study of methane on Mars was advanced by the arrival of the Mars Science Laboratory (MSL) Curiosity rover in Gale Crater. Curiosity carried the Tunable Laser Spectrometer within the Sample Analysis at Mars (TLS-SAM) instrument suite, which

detected methane in Gale Crater (Webster et al., 2015). The TLS-SAM observations were interpreted to have an apparent repeatable seasonal variation during the nighttime with a mean abundance of 0.41 ± 0.16 ppbv, with higher methane abundances around the equinoxes and lower ones roughly at the solstices (Webster et al., 2018). However, more recent statistical analysis suggests that the data are not inconsistent with no seasonal variation (Gillen et al., 2020). Whether steady or seasonally varying, these abundances were suggested to be representative of a global background abundance of Mars atmospheric methane (Webster et al., 2015, 2018), which we hereafter refer to as the *background* MSL observations. Occasional methane spikes above the background were observed by TLS-SAM, with abundances an order of magnitude higher than the first ones. One of these spikes has been observed contemporaneously from orbit by the Planetary Fourier Spectrometer (PFS) on board Mars Express (Giuranna et al., 2019). These spikes were attributed to a local or perhaps a regional source emission strongly increasing the Gale Crater methane distribution over the background levels.

Measurement issues led some researchers to question several of the methane detections from orbiters, for example, some methane detections by the PFS (Geminale et al., 2008, 2011) or from Mars Global Surveyor Thermal Emission Spectrometer (TES) (Fonti et al., 2015). For example, Fonti et al. (2015) note that their prior methane detections inferred from Mars Global Surveyor (MGS) Thermal Emission Spectrometer (TES) data cannot be confirmed due to the TES instrument's low spectral resolution and the noise content in this area. The in situ observations made by MSL (the only platform that can measure methane on the Martian surface) have also been questioned (e.g., Lefèvre, 2019; Zahnle, 2015; Zahnle & Catling, 2019), due to instrument contamination involved in the TLS-SAM measurements. It should be noted, however, that a detailed analysis has not demonstrated that such potential issues could significantly affect the methane measurements (Webster et al., 2013, 2015, 2018, 2020). More recently, observations by the Atmospheric Chemistry Suite (ACS) and by the Nadir and Occultation for Mars Discovery (NOMAD) instruments on board the ExoMars Trace Gas Orbiter (TGO), which has a much lower detection limit than any previous instrument sent to Mars orbit, have not detected methane in the atmosphere (Knutsen et al., 2021; Korabiev et al., 2019; Montmessin et al., 2021). This raises the crucial question of whether the nondetection of methane by TGO can be reconciled with MSL observations. If near-surface atmospheric methane does indeed vary as observed by MSL, then major questions are, what process(es) are producing or emitting methane, what rapid mechanism is destroying it, and where on the planet methane is being produced or emitted.

TGO methane observations began roughly two months before the onset of the Mars Year (MY) 34/2018 Global Dust Storm (GDS) (Guzewich et al., 2018; Viúdez-Moreiras, Newman, et al., 2019), when methane continued to be measured in Gale Crater despite contemporaneous nondetection by TGO from orbit. More than two years later, the nondetection by TGO remains and indeed the detection limits have been lowered to 0.02 ppbv above the boundary layer (Montmessin et al., 2021). Two possibilities for reconciling the MSL and TGO observations are that: (a) TGO is not sensitive to the near-surface atmosphere, whereas MSL only samples the very near-surface atmosphere and (b) TGO performs observations either at sunset and sunrise (Korabiev et al., 2019), whereas the enrichment observations of methane by MSL reported in Webster et al. (2018) were measured during the nighttime. Moores et al. (2019) used an atmospheric box (0-D) model coupled with a one-dimensional regolith model to propose that the much deeper planetary boundary layer (PBL) during the daytime could be dispersing methane from hypothetical methane emissions to below the measurement threshold of TGO. In this way, the nighttime abundance of methane from a surface source, restricted from vertical mixing by the stable nighttime PBL, could be much higher than in the well-mixed daytime. Recent enrichment observations suggest that the values during daytime are below the detection limit of TGO (Webster et al., 2021). Viúdez-Moreiras, Arvidson, et al. (2020) suggested that the methane abundance detected by MSL could be significantly influenced by local meteorology and/or that methane source emissions could be highly dependent on location, areocentric solar longitude (L_s), and local time.

Regarding the origin of methane detected by MSL, particularly the spikes, it should be noted that they were postulated to be the result of localized emission sources in the Gale Crater region or in its vicinity (e.g., Giuranna et al., 2019; Webster et al., 2015, 2018; Yung et al., 2018). Pla-García et al. (2019) modeled source emissions inside and outside Gale Crater based on clathrate emissions in an attempt to explain such spikes. Although no good match with observations was found, simulations indicated that emissions near to MSL came closest to matching, thus reinforcing the idea of a local origin of spikes. Viúdez-Moreiras (2021a) suggested that the variability of winds in the vicinity of MSL is sufficient to produce strong methane spikes at MSL's general location

from a nearby emission source, even without invoking sol-to-sol variability in winds, variable emission fluxes or eventual emissions into the atmosphere, or microscale flows not resolved in the meteorological data. A recent modeling study by Luo et al. (2021) also favors localized emission sources in the Gale Crater region. However, the *background* MSL observations of methane are even more complex in their interpretation. As noted above, they were initially taken to represent the background (global or regional) level of Mars atmospheric methane (Pla-García et al., 2020; Webster et al., 2015, 2018). The subsequent nondetection of methane in the atmosphere of Mars by TGO led to a major problem in our understanding of the methane on Mars. It has been proposed that the methane detected by MSL could come from extensive nonlocalized areas of the planet, such as the Gale crater region and beyond, emitting methane for example, by microseepage (e.g., Moores et al., 2019), and requiring an unknown strong loss mechanism to be at work. Such a loss mechanism would be necessary to destroy methane at rates even orders of magnitude faster than the standard gas-chemistry scheme (i.e., oxidation by OH and O(¹D) in the lower troposphere and photolysis at higher altitudes), which properly predicts the methane distribution on Earth (Lefèvre & Forget, 2009). Without such a loss mechanism, emission sites would slowly build up global atmospheric methane abundances to levels that would violate the TGO-observed upper limit. The source location has strong implications concerning the requirements for a hypothetical loss mechanism, and therefore, for atmospheric chemistry on Mars. It also has implications for the potential compatibility between orbital and surface observations. In this paper, we address two questions: (a) is it possible to explain the MSL detections with an emission source that TGO could not detect it? and (b) where is the likeliest source of emissions to explain the MSL detections? Here, a systematic search for the possible emission source location of the background methane abundance detected by MSL is performed for the first time by using a three-dimensional dispersion model (DISVERMAR) (Viúdez-Moreiras, 2021a).

The paper is structured as follows. Section 2 describes the three-dimensional dispersion model (DISVERMAR) as well as the three-dimensional climate model used to provide input meteorology (MarsWRF) and the various simulation scenarios used. Section 3 presents the modeled wind patterns in the Gale crater region. Section 4 presents and analyzes several dispersion simulations that constrain the location of the source responsible for the methane detected by MSL. Section 5 presents numerical simulations that constrain the size of the hypothetical source location. Section 6 discusses a global picture of the findings presented in this paper, focusing on the problems presented by the sources that produce the best match to methane observations. Finally, Section 7 presents the conclusions.

2. Methods: Three-Dimensional Atmospheric Modeling Used in This Work

2.1. Three-Dimensional Atmospheric Dispersion Model (DISVERMAR)

Atmospheric dispersion models are widely used on Earth to predict and study the effects of local and regional emissions (e.g., Leelosy et al., 2014 and references therein). A major advantage of including tracers into weather prediction models is that dispersion models can run orders of magnitude more quickly, and only require the output from a single meteorological data set (e.g., from a single weather prediction simulation) to produce many chemical transport simulations (Grell et al., 2004; Leelosy et al., 2014). In this study, we use the three-dimensional atmospheric dispersion versatile model for Mars (DISVERMAR), input with the meteorological fields previously computed in a separate simulation by MarsWRF. DISVERMAR operates on local-to-regional scales, as is typical in dispersion models developed for Earth (e.g., Leelosy et al., 2014). A brief description of DISVERMAR is presented in this section, while the reader is referred to Viúdez-Moreiras (2021a) for further model details.

DISVERMAR solves the advection-diffusion equation in an Eulerian framework:

$$\frac{\partial N}{\partial t} - \nabla(N\mathbf{v}) + \nabla(\mathbf{D} * \nabla N) = S, \quad (1)$$

where N is the species number density, $\mathbf{v}$ the three-dimensional wind field, $\mathbf{D}$ the eddy diffusivity matrix, and S the source term. The discretization of this Partial Differential Equation (PDE) is performed by high-order central differences for the diffusive term and an upwind scheme for the advective term (e.g., Durran, 2010; Hirsch, 2007). In this study, the model integrates the PDE with the forward Euler scheme and makes use of a standard Arakawa-C grid (Arakawa & Lamb, 1977), where the u and v wind components (the orthogonal x and y direction components) are staggered half a grid point in the x and y directions, respectively, relative to the tracer

species points). In the vertical, the tracer species is staggered by one-half a vertical grid below the vertical velocity w , and the u and v wind components are at the same level as the tracer species. The model can operate in z (height) or σ - z (terrain-following) coordinates. Here, σ - z coordinates are used due to the significant topography in the model domain, that is, the topography of the Gale Crater and its vicinity including the dichotomy boundary (Neumann et al., 2003). Furthermore, the vertical grid can provide, if needed, an increase in vertical resolution as the altitude decreases to better capture transport in the near-surface atmosphere.

The eddy diffusivity coefficients are derived inside DISVERMAR from the bulk Richardson number using mixing length theory in the vertical domain and from the wind gradient in the horizontal domain (e.g., Draxler & Hess, 1998; Hong & Pan, 1996; Louis, 1979; Smagorinsky, 1963). The predicted three-dimensional meteorological fields (winds, pressure, and temperature) as a function of local time were obtained from a MarsWRF simulation (Richardson et al., 2007; Toigo et al., 2012) as described below.

2.2. Meteorological Fields From MarsWRF Simulations

MarsWRF solves the Navier Stokes equations and also includes parameterizations of radiative transfer through a dusty CO₂ atmosphere, with a prescribed 4-D dust distribution; the CO₂ condensation-sublimation cycle; sub-grid scale horizontal and vertical mixing; heat transport within the subsurface; and the exchange of heat and momentum between the surface and atmosphere.

MarsWRF simulations were performed to obtain three-dimensional winds, pressure, and temperature fields as a function of local time at $L_s = 0^\circ, 90^\circ, 180^\circ$, and 270° , which were later input to DISVERMAR simulations. MarsWRF simulations were performed using the same setup as in Newman et al. (2017) and Richardson and Newman (2018), in which smaller, higher-resolution regions were “nested” inside a global model, centered on the Gale crater. The vertical grid spacing chosen was vertical grid-A as described in those papers, which resolves the lowest layers of the PBL with three layers below 100 m. This is necessary to properly simulate the species transport with DISVERMAR in the boundary layer. The resolution of each nest is increased compared to its parent domain, with the second nest at ~ 13 km resolution, the third nest covering an area roughly 10 times larger than the crater at ~ 4.4 km horizontal resolution, and the fourth covering just the crater at ~ 1.4 km resolution. The vertical mixing scheme used in the MarsWRF simulations is described in Hong and Pan (1996) and was selected for consistency with the DISVERMAR simulations. The prescribed atmospheric dust distribution was obtained from the Mars Climate Database (MCD; Forget et al., 1999; Millour et al., 2018), while the surface properties (albedo, emissivity, and thermal inertia) were obtained from Mars Global Surveyor (MGS) and Thermal Emission Spectrometer (TES). The topography was retrieved from the MGS Mars Orbiter Laser Altimeter (Neumann et al., 2003).

The wind patterns for the aforementioned L_s are described in Section 3. Winds from this MarsWRF simulation also agree moderately well, in terms of the time of day variation in wind speed and especially wind direction, with MSL Rover Environmental Monitoring Station (REMS) near-surface winds measured during a special wind monitoring campaign conducted at $L_s \sim 70^\circ$ in the Bagnold dune field (Newman et al., 2017). Unfortunately, due to damage to the REMS wind sensor on landing, winds in most other seasons are complex in their interpretation and need special retrieval algorithms to overcome the wind sensor damage (Viúdez-Moreiras et al., 2019a, 2019b), making it difficult to quantitatively compare any model predictions with wind observations. However, MarsWRF winds have also been used to predict aeolian activity inside the Gale crater as a function of season, location, and time of day, and these predictions also compare well with observations except in cases with local topography at scales not captured by the model (e.g., Baker et al., 2018). This provides confidence in the use of MarsWRF wind predictions for this study.

2.3. DISVERMAR Simulations

Numerical simulations for a mesh with source locations in the region around Gale Crater were performed to evaluate the correspondence between MSL and TGO measurements for each case (Figure 1). Given the strong complexity of the wind field in the Gale Crater region as observed by MSL from the surface (Newman et al., 2017; Viúdez-Moreiras et al., 2019a, 2019b), an additional set of numerical simulations with higher resolution, both in the DISVERMAR and in MarsWRF meteorological fields, covering Gale Crater (domain-B) were also performed (Figure 1 bottom), thus resolving the Gale Crater dynamics. The grid comprised of 50×50 horizontal

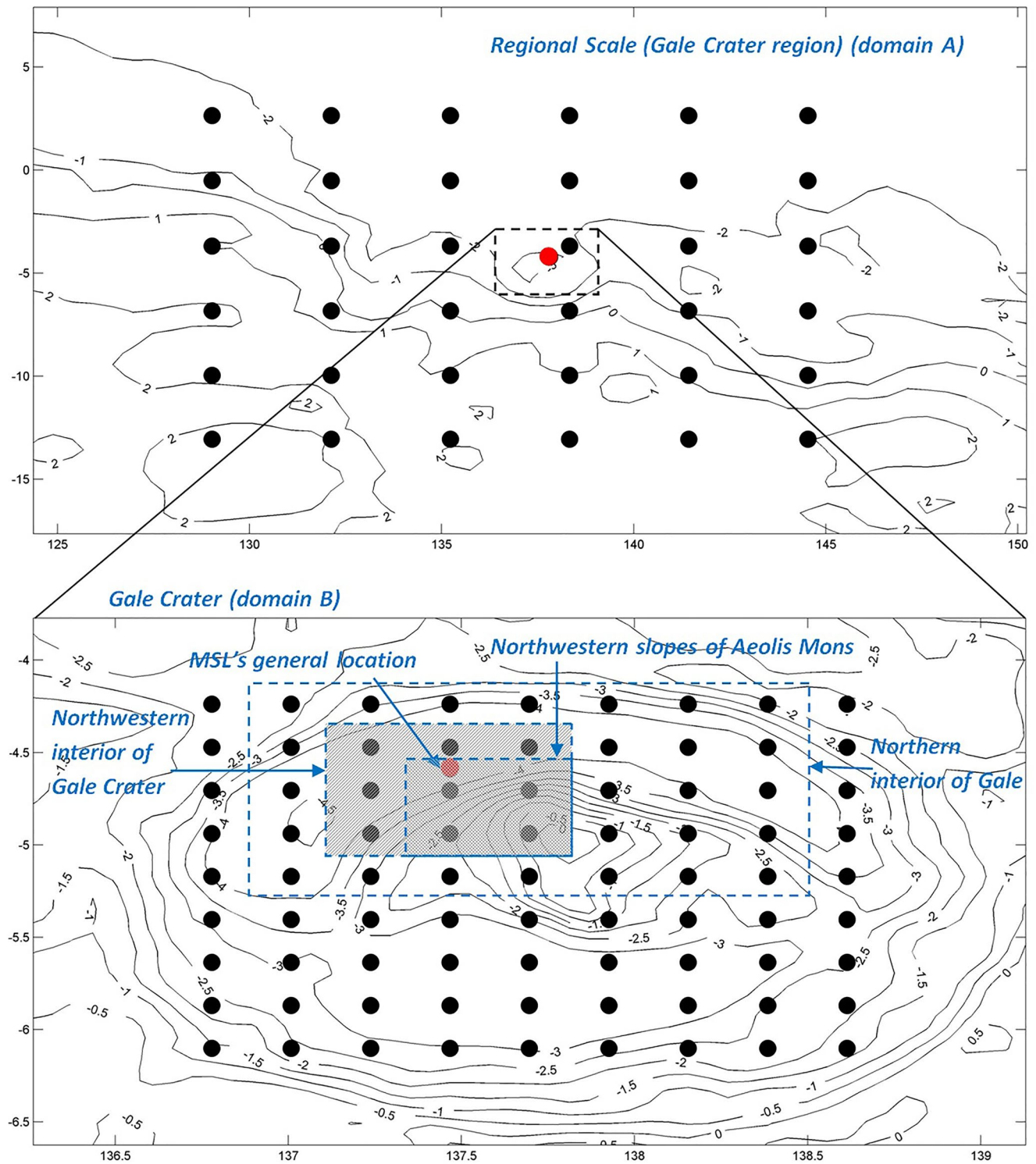


Figure 1. Locations where a methane source was tested (in black dots) in the region around Gale Crater (domain-A) (top) and in the refined mesh around MSL (domain-B) (bottom). Note that the numerical grid has a higher resolution than the mesh of simulated emission sources (see text). The general location of MSL (in red dots) is also explored as a possible source location. The contour lines depict the surface elevation relative to MOLA aeroid in kilometers.

nodes, spaced at ~ 30 km for domain-A and ~ 3 km for domain B, and 25 vertical levels. The following surface areas related to the possible location of the methane source responsible for the methane detected by MSL are defined in Figure 1: (a) the regional scale or Gale Crater region, which covers domain-A of the DISVERMAR simulations, within the tropical region of the planet, (b) the Gale Crater (domain-B); (c) the northern interior of the crater, (d) the northwestern interior of the crater, which envelopes the northwest crater rim and crater floor and the northern slopes of Aeolis Mons, (e) the northwestern slopes of Aeolis Mons, and (f) the MSL's general location, that is, the general location of MSL within the northwest slopes of Aeolis Mons.

Dirichlet boundary conditions were used for the spatial domain, except on the surface, where a Neumann boundary condition represents an emission flux of methane from the surface in a particular location. Thus, it is assumed that methane abundance is negligible at the lateral boundary of the physical domain, which is a good approximation if the boundaries are far away from the source (e.g., Leelosy et al., 2014). The effect of the choice of domain boundaries for the methane magnitudes derived in this work is presented in Supporting Information S1. As shown in Table S1 of Supporting Information S1, boundaries do not significantly affect the results, even in the worst case that the emission source is closer to them than the domain center (Figure S1 in Supporting Information S1).

The background MSL's observations to date (Webster et al., 2018) suggest that methane emission responsible for such levels of methane detected at Gale crater is not episodic. Here, the methane emission is continuous along the time domain in a particular location at the surface for each simulation. A constant emission flux is assumed for simplicity. Continuous emissions with variable emission fluxes, such as those proposed by Etiope and Oehler (2019) and Viúdez-Moreiras, Arvidson, et al. (2020), dependent on pressure fluctuations and/or winds, would produce variability in the diurnal and seasonal cycles. However, it is assumed in this study that they would not have a significant influence on mean emission fluxes, neither on the detection by TGO.

Simulations were initialized without methane in the domain, and spun-up in order to allow a proper distribution of methane (Viúdez-Moreiras, 2021a). The spin-up period was less than 4 sols for these simulations. Then, the well-established diurnal variation of the methane abundance was obtained. A timestep of 30 s was used, with the wind field being updated every 5 min. The first prognosed vertical level is at ~ 10 m from the surface, with a model top at 20 km.

An arbitrary initial emission flux of methane was injected in the domain for each of the localized emission sources shown in dots in Figure 1, totaling 37 simulations for domain-A and 82 simulations for domain-B. Then, the simulated methane abundance field was scaled to match the background MSL observations in the near-surface at MSL's general location at midnight, applying the same factor to the initial arbitrary flux for consistency, following the procedure detailed in Viúdez-Moreiras (2021a). Methane was injected in a region of ~ 90 km² in domain B (~ 900 km² in domain A), that is, the single source is considered over this area. Also, an additional simulation involving a nonlocalized emission source with a uniform emission flux across domain-B ($\sim 20,000$ km²) was considered in order to compare localized and nonlocalized scenarios.

2.4. Evaluation of the Emission Source Responsible for the Methane Detected by MSL

The source location responsible for the detected abundance by MSL at Gale Crater should simultaneously satisfy the MSL observations and the TGO constraints. The resulting 4D species mixing ratio fields for each emission source k presented in Figure 1 were thus assessed as follows.

Regarding the near-surface abundance levels detected by MSL, each emission source should satisfy sustainable midnight values of ~ 0.4 ppbv (Webster et al., 2018). Recently, Webster et al. (2021) reported the first enrichment observations performed during the daytime. Two daytime measurements close to midday of average value 0.05 ppbv ± 0.22 ppbv (95% confidence interval) were reported between the southern winter solstice and the equinox ($L_s = 121^\circ$ and 134°). These two measurements are not analyzed in this manuscript; however, DISVERMAR simulations presented in Viúdez-Moreiras (2021a) confirm previous modeling efforts by Moores et al. (2019) and Pla-García et al. (2019) concerning low methane abundance expected in the near-surface during daytime by a nearby emission source. Further modeling research will be performed once more daytime measurements are acquired.

Regarding TGO's constraints, the emission source should satisfy the upper limit of methane found by TGO around sunrise and sunset (Solar Zenith Angle, SZA, $70^\circ < \text{SZA} < 110^\circ$) in the Gale Crater region and above the

PBL, before methane is dispersed in the atmosphere, that is to say, the methane plume should not be observed by TGO. The nondetection of methane by TGO to date has an upper limit of ~ 0.02 ppbv in a global scale (Montmessin et al., 2021). The best upper limit produced by TGO-ACS near Gale Crater to date is 0.1 ppbv at $L_s = 126^\circ$ on MY34, at a distance of 328 km from the crater center and at an altitude of 11 km (Montmessin et al., 2021). Thus, a conservative upper limit, $X_{CH_4}^{UL} = 0.05$ ppbv above $z_T = 10$ km was considered to analyze whether the direct emission of methane by a source located in any location represented in Figure 1 could be detected by TGO before methane is dispersed in the atmosphere. We define the maximum volume mixing ratio achieved during the daytime above z_T , $X_{CH_4}^k = \max_{i \in D, \forall z > z_T} X_i^k$, where X_i^k is the mixing ratio of methane at any location i in the domain D for a particular emission source k . For consistency with observations, $X_{CH_4}^k$ should be lower than the detection limit of TGO, $X_{CH_4}^{UL}$, to enable the compatibility of emission source k . In any case, we present in Section 4 the $X_{CH_4}^k$ values for each emission location k in order to easily check the effects of deviations in the upper limit taken as a reference.

TGO may as well constrain the emission source in terms of the required loss mechanism to destroy methane on the Mars atmosphere, that is, the required chemical lifetime of methane (τ_{CH_4}) able to maintain global average levels below the TGO threshold, even if the plume responsible for the methane detected by MSL is not directly detected anywhere in the Gale Crater region before being dispersed in the atmosphere. Generally speaking, if the emission source is far away from MSL, the required emission flux should be higher than if the flux necessary to match MSL's detected abundance is in a closer emission flux. Actually, it will also depend on the three-dimensional transport, for example, favorable air masses coming to MSL from the emission source. Therefore, the chemical lifetime of methane was computed and analyzed for each source location ($\tau_{CH_4}^k$). $\tau_{CH_4}^k$ was derived assuming steady state, that is, the emission rate of methane, ζ_p , equals its loss rate, ζ_l , using the following equation:

$$\tau_{CH_4}^k = \frac{X_m m_T}{\zeta_e^k}, \quad (2)$$

where $\tau_{CH_4}^k$ is the required chemical lifetime of methane for the source location k , X_m is the global average mass mixing ratio of methane that equals the threshold derived from TGO (conservatively 0.01 ppbv; note that this is in the planetary-scale, thus lower than the reference detection limit considered for the Gale Crater region as stated above; note as well that Equation 2 requires mass mixing ratio), m_T is the total mass of air in the Martian atmosphere and ζ_e^k is the emission rate of methane for the location k . $\tau_{CH_4}^k$ only considers the aforementioned emission source as the unique source of methane in the atmosphere. τ_{CH_4} should be computed accordingly if more sources emitting simultaneously are considered. For N MSL-like emission sources, $\tau_{CH_4} = \frac{\tau_{CH_4}^k}{N}$. In fact, this is another trouble related to methane on Mars: even in case the source responsible for MSL's observations would not constrain the $\tau_{CH_4}^k$, a question arises concerning how many emission sources are active on the planet. Although global considerations of methane on Mars are out of the scope of the present paper, we briefly discussed them in Section 6.

3. Wind Patterns in the Gale Crater Region

The transport of methane from the hypothetical emission source to MSL's general location is mostly driven by advection. This section presents the near-surface wind patterns in the region around Gale Crater, which are computed by MarsWRF and used in the DISVERMAR simulations.

Models and available data suggest that the large-scale flow at the region around Gale Crater (Figure 1a) is mostly dominated by the Hadley circulation and thermal tides, which are strongly modified at the near surface by topography. In addition, the dichotomy boundary and other relevant features in the region produce regional flows that are superimposed to the aforementioned circulation, making complex interactions during the sol, which are also dependent on season (e.g., Banfield et al., 2020; Newman et al., 2017; Rafkin et al., 2016; Spiga et al., 2018; Tyler & Barnes, 2013; Viúdez-Moreiras, Newman, et al., 2020).

In order to analyze the seasonal effect in the transport of methane from a source (Figure 1) to MSL's location, four scenarios around the Martian year can be considered from a climatology perspective: the equinoxes and the solstices ($L_s = 0^\circ, 90^\circ, 180^\circ$, and 270°). The remaining areocentric solar longitudes could be considered, roughly speaking, as intermediate scenarios from the former. Figures 2a–2d presents the diurnal average wind fields at the Gale Crater region as predicted by MarsWRF, both for the equinoxes and solstices. As can be seen, it is

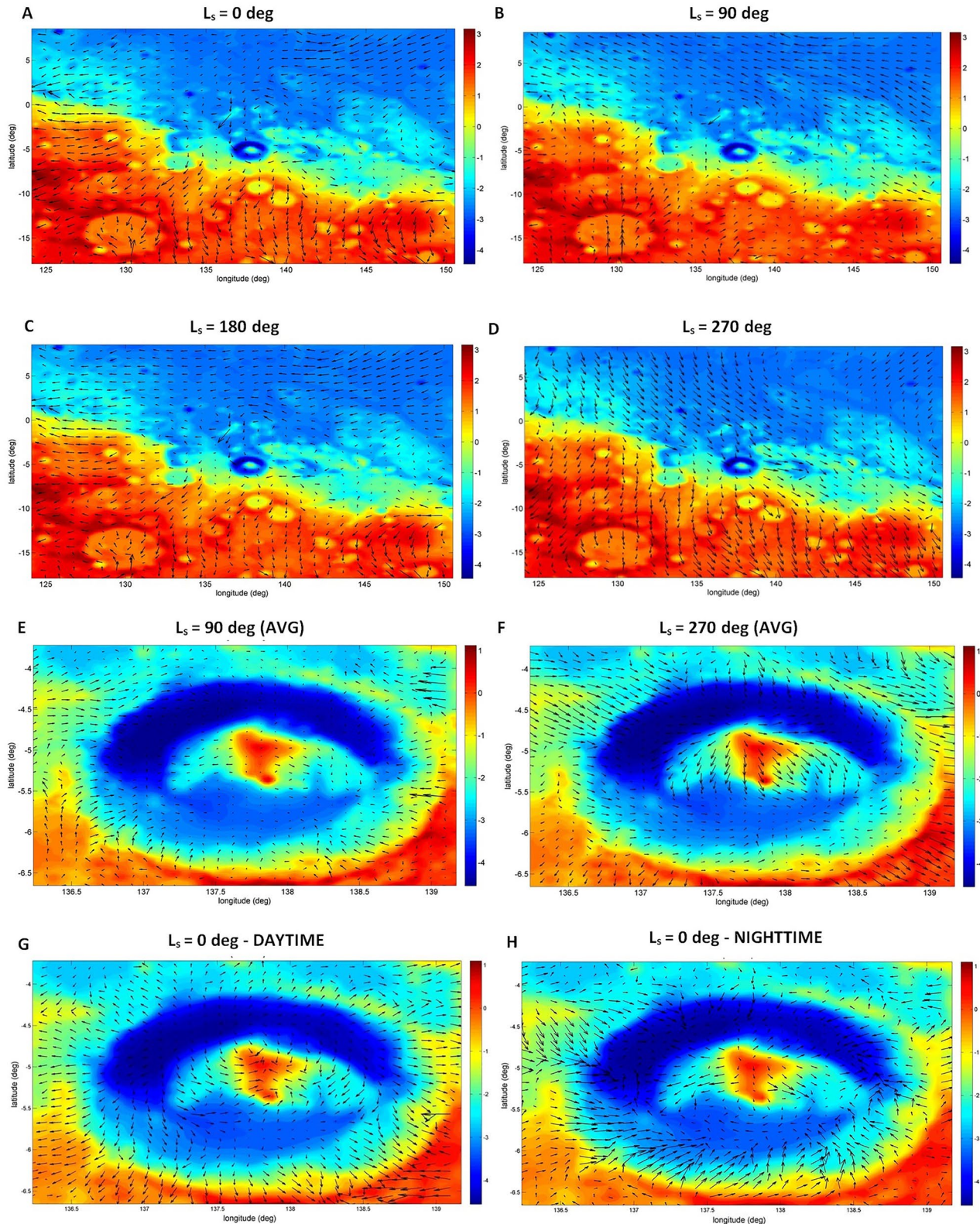


Figure 2. Near-surface modeled wind patterns at the Gale Crater region, as predicted by MarsWRF. (a–d) Diurnal average wind fields at the Gale crater region (Figure 1—domain A) for $L_s = 0^\circ$, 90° , 180° , and 270° . (e and f) Diurnal average wind fields at Gale Crater (Figure 1—domain B) for $L_s = 90^\circ$ and 270° . (g–h) Daytime (12:00 LTST) and nighttime (0:00 LTST) wind fields at Gale Crater for $L_s = 0^\circ$. Topography is shown as color-coded elevation (km) from the Mars Orbiter Laser Altimeter (MOLA).

possible to extract two conclusions relevant for this study: (a) the differences in the solstitial Hadley cell between southern winter ($L_s = 90^\circ$, Figure 2b, producing zonal-mean southerly flows in the near surface) and southern summer ($L_s = 270^\circ$, Figure 2d, producing zonal-mean northerly flows in the near surface), and the asymmetry in the magnitude of the cell due to the orbital eccentricity of Mars (which is stronger during the southern summer) has a marked impact in the resulting near-surface winds, in accordance with previous model results and (b) the dual-Hadley cell, weaker equinoctial circulation, can be considered in this region, roughly speaking, to be similar between both equinoxes ($L_s = 0^\circ$, Figure 2a and $L_s = 180^\circ$, Figure 2c), which allows us to consider in a first approximation one equinox (e.g., $L_s = 0^\circ$) as the representative for the equinoctial scenario.

At Gale Crater, where the MSL is located (Figure 1b), the situation is more complex, given that a strong local circulation with a marked diurnal cycle usually dominates over the regional and global circulations in the near surface (Newman et al., 2017; Rafkin et al., 2016; Tyler & Barnes, 2013; Viúdez-Moreiras et al., 2019a). Significant differences can be observed in the diurnal-mean winds, as in the Gale Crater region described above. Figures 2e and 2f present the diurnal-mean near-surface winds simulated in the domain B (Figure 1b) by MarsWRF. Convergent flows can be observed in the northwest interior of Gale Crater, being stronger in $L_s = 270^\circ$, with the Aeolis Mons flows dominating over the crater rim flows at MSL's general location. In addition, the strong solstitial Hadley circulation at $L_s = 270^\circ$ produces mostly northerly winds in the southern slopes of Aeolis Mons. A northerly meridional component is typical in most of winds at Gale Crater at this season, except in the northern slopes of Aeolis Mons. The equinoxes (not shown for simplicity) present an intermediate scenario between the solstitial circulations, although closer to $L_s = 270^\circ$ than $L_s = 90^\circ$. The local slope flows in Aeolis Mons and the crater rims produce a diurnal perturbation superimposed on the mean flow. The diurnal variation in Gale Crater can be simplified in two regimes: a daytime regime, where upslope anabatic flows are at work (which tend to produce a northerly meridional component at MSL's general location on the northwest slopes of Aeolis Mons), and the nighttime regime, where downslope katabatic flows take place (which tend to produce a southerly meridional component at MSL's general location), and with two transition periods within them. Figures 2g and 2h present the near-surface winds predicted by MarsWRF for daytime (12:00 LTST) and nighttime (00:00 LTST), at the southern autumn equinox ($L_s = 0^\circ$). As can be seen, the local flows dominate at the near-surface, producing a very rich environment with complex interactions between the large-scale, regional, and local circulation, but with established divergent flows during the daytime in the crater rims and the slopes of Aeolis Mons (Figure 2g) and convergent flows during the nighttime (Figure 2h) in the near-surface atmosphere.

4. Localizing the Emission Source Responsible for the Levels of Methane Detected by MSL

This section presents and analyzes several dispersion simulations that constrain the location of the source responsible for the background methane detected by the MSL. Several parameters were considered to evaluate the possible location of the methane levels detected by the MSL, in accordance with Section 2.4.

4.1. Detection of the Methane Plume Emitted by the Emission Source Responsible for MSL's Observations

There are not enough data from TGO to constrain the hypothetical plume of methane at Gale Crater at every season (Montmessin et al., 2021). It is assumed here that $X_{\text{CH}_4}^{\text{UL}}$ is applicable in the L_s -range 0–180°. Thus, $X_{\text{CH}_4}^{\text{k}}$ (above z_{T}) was computed both for the equinox and for the southern winter solstice. These L_s are selected because they are overall illustrative for most of the wind patterns throughout this L_s -range (Section 3).

Figure 3 presents the response to the surface sources showing the aforementioned values of $X_{\text{CH}_4}^{\text{k}}$ for southern autumn equinox ($L_s = 0^\circ$, left column) and winter solstice ($L_s = 90^\circ$, right column). The figure should be read by looking up the latitude and longitude to find the location of the source, and then reading off the maximum value of $X_{\text{CH}_4}^{\text{k}}$ corresponding to this source location. Note that by definition, all simulations have been scaled so that the simulated value of methane at the MSL site in the near surface is consistent with nighttime TLS-SAM observations. In particular, it is important not to interpret Figure 3 as representing maps of the instantaneous atmospheric distribution of methane for any given time. Nor does the maximum value $X_{\text{CH}_4}^{\text{k}}$ necessarily occur at the latitude-longitude value shown (the value should be read as the maximum value of $X_{\text{CH}_4}^{\text{k}}$ anywhere in the domain above z_{T} if the source was at that latitude and longitude). Figure S2 in Supporting Information S1 presents, for

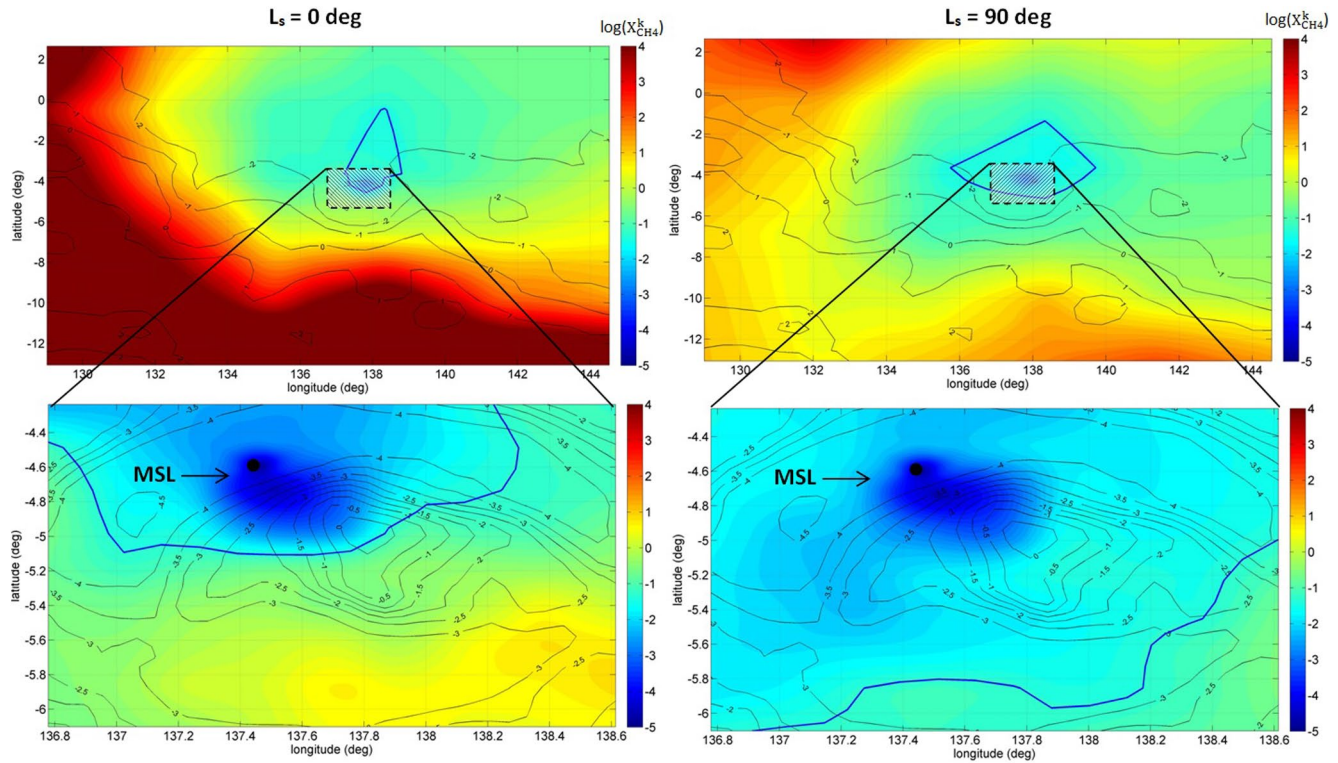


Figure 3. Values of maximum methane mixing ratio $X_{CH_4}^k$ (ppbv, in logarithmic scale) above z_T achieved during the sunrise/sunset (the observable timeslot by TGO), as a function of the location (latitude and longitude) of the emission source, for two areocentric solar longitudes ($L_s = 0^\circ$ and 90° , left and right column, respectively). The mesh of source locations were generated with model simulations from each source showed in Figure 1. The resolution in the Gale Crater region is enhanced in the lower panels by means of high-resolution simulations around MSL (Domain-B, Figure 1 bottom). The blue contour lines highlight the TGO upper limit $X_{CH_4}^{UL}$; therefore, $X_{CH_4}^k$ should be lower than this threshold to prevent the TGO detection from orbit. Topography is shown as black contour lines in km.

completeness, the results for $L_s = 270^\circ$, which show similar results than $L_s = 0^\circ$, but even more constrained than it, particularly at Gale Crater.

It can be seen in Figure 3 that the sustainability of the potential emission location depends on season, as wind patterns, the main driver to transport the chemical species from the emission location to MSL's location, are strongly dependent on this factor. Southern emission sources outside equatorial latitudes would involve $X_{CH_4}^k$ mixing ratios of the order of 100 ppmv or higher above z_T to produce the near-surface abundance of methane detected at MSL's general location close to the equinox, which would be easily detected by TGO. Source locations close to the Gale Crater produce $X_{CH_4}^k$ values above $X_{CH_4}^{UL}$ ($X_{CH_4}^k$ values even increase to 1–10 ppbv at 5 km of altitude for such emission sources), which could possibly be detected by TGO at both seasons, invalidating as well such source locations as responsible for the detected methane by MSL. Only emission sources in $\pm 2^\circ$ centered at Gale Crater can produce levels of methane that are not detectable by TGO (Figure 3 top). Even source locations in the south of Gale Crater would produce emissions that could be detected by TGO (Figure 3 bottom).

However, a source in the northern interior of Gale Crater would produce methane mixing ratios of the order of 10^{-2} to 10^{-5} ppbv above z_T , which are far below the detection limit of TGO. The closer the source is to MSL, the less methane is observed at higher altitudes. The simulated methane abundance above z_T drops to $\sim 10^{-5}$ ppbv, far below the detection limit of TGO, if the methane is emitted at the northwestern slopes of Aeolis Mons. There are even other regions within Gale Crater that would be unable to produce enough methane at MSL's general location to meet the near-surface abundance without being detected by TGO, such as the south of Gale Crater. Thus, regarding the parameter evaluated in this subsection (the direct visibility of the methane plume), and strictly speaking, the simulations presented here suggest that the available TGO data effectively constrain the source responsible for the methane detected by MSL to the northern crater extending the region north of Gale Crater as far as latitude $\sim 2^\circ$ S, within longitude ranging between 136° and 139° . Otherwise, the plume should have been detected. It represents a hard constraint and invalidates the possibility that methane comes from more distant

regions around the planet, without an event that could strongly affect the meteorology, such as a dust storm. Furthermore, in accordance to Figure 3, it is more probable that the source is in the northwest interior of Gale Crater and, in particular, in the northwest slopes of Aeolis Mons, thus not emitting methane just close to the detection limit of TGO.

4.2. The Emission Source Responsible for the MSL's Methane Observations and Its Effects on the Methane Chemical Lifetime on Mars

TGO may as well constrain the emission source location in terms of the required loss mechanism able to destroy methane, after the methane plume is dispersed in the atmosphere (Section 2). The admissible emission flux to be compatible with TGO, while avoiding a rapid destruction mechanism ($\tau_{\text{CH}_4}^k \sim 300$ Earth-years), is ~ 0.8 kg/sol (Equation 2). Figure 4 shows the computed values of $\tau_{\text{CH}_4}^k$ for each emission source k , using a similar representation as in Section 4.1 (as a function of latitude and longitude), considering in a first approximation that the emission flux required for the southern autumn equinox is operating during the whole year. Thus, for continuous emission along the sol, an emission source outside the northwest slopes of Aeolis Mons imposes constraints on a hypothetical methane chemical loss mechanism; that is, even if methane is *only* produced on Mars in the northern interior of the crater but outside this region, τ_{CH_4} should be less than 1 Earth year (ζ_e^k greater than 10^2 kg/sol) (Figure 4 bottom). $\tau_{\text{CH}_4}^k$ decreases to $\sim 10^{-2}$ Earth years or even less if the emission source is in the south of Gale Crater ($\zeta_e^k > 10^4$ kg/sol) and to $\sim 10^{-5}$ Earth years (~ 5 min) or less if the emission source responsible for methane abundance detected by MSL is a few hundred km from Gale Crater (Figure 4 top). The latter results significantly differ from the predicted standard gas-chemistry schemes in the Martian atmosphere, whose chemical lifetime is of the order of hundreds of years (Lefèvre & Forget, 2009). If chemistry would have been included in the simulations, the rapid chemical loss of the emitted methane from those locations before arriving at MSL's general location would require even much higher emission fluxes from those sources to match the background MSL observations, increasing the methane abundance in the domain as well, thus delving into the incompatibility of those emission areas.

However, if the methane detected by MSL is produced within Gale Crater, and particularly in the northwest slopes of Aeolis Mons (Figure 4 bottom), the short air parcel trajectory to MSL's general location would allow for lower emission fluxes ($\zeta_e^k < 0.8$ kg/sol) and therefore, $\tau_{\text{CH}_4}^k$ of the order of centuries or greater (Equation 2), in accordance with the standard gas-chemistry lifetime. Other seasons yield a similar order of magnitude for $\tau_{\text{CH}_4}^k$. For MSL's general location, $\tau_{\text{CH}_4}^k$ increases to a nonlimited value of ~ 700 Earth-years ($\zeta_e^k \sim 0.4$ kg/sol and the estimated flux per area equals to $\sim 5 \times 10^{-14}$ kg m $^{-2}$ s $^{-1}$).

Thus, the model simulations presented in Figure 4 show that the source responsible for the methane detected by MSL would not influence the required chemical lifetime of methane on Mars only if the source is located in the northwestern slopes of Aeolis Mons, where the MSL is located. Otherwise, an unknown rapid destruction mechanism should exist even in the improbable case that there are no additional sources of methane at other locations on Mars. This scenario, which will be detailed in Section 6, is improbable simply on the basis of the likelihood of MSL having landed at a unique or exceedingly rare site, but reinforces the possibility that the source responsible for the methane detected by MSL is close to the rover, probably in the northern crater rim, the crater floor, and/or the slopes of Aeolis Mons.

5. Constraining the Size of the Emission Source Responsible for the Levels of Methane Detected by MSL

In order to gain a better insight into the surface area of the emission source responsible for the methane abundance detected by MSL, an additional simulation was conducted in which the whole of domain-B emitted with a uniform flux. This represents a more dispersed source location than localized sources in the prior sections (Figure 1). Again, the model simulation was scaled to match levels of methane at MSL's general location. Table 1 presents the comparison between both scenarios. As can be seen, $\tau_{\text{CH}_4}^k$ reduces by more than two orders of magnitude if the whole of domain-B is emitting methane instead of a localized emission source in the vicinity of MSL. This means that an enhanced loss mechanism is necessary in the nonlocalized emission scenario even in case that only Gale Crater is emitting methane on Mars. By contrast, the localized emission scenario is compatible with the current understanding of atmospheric chemistry on Mars. This difference is due to the fact that the specific

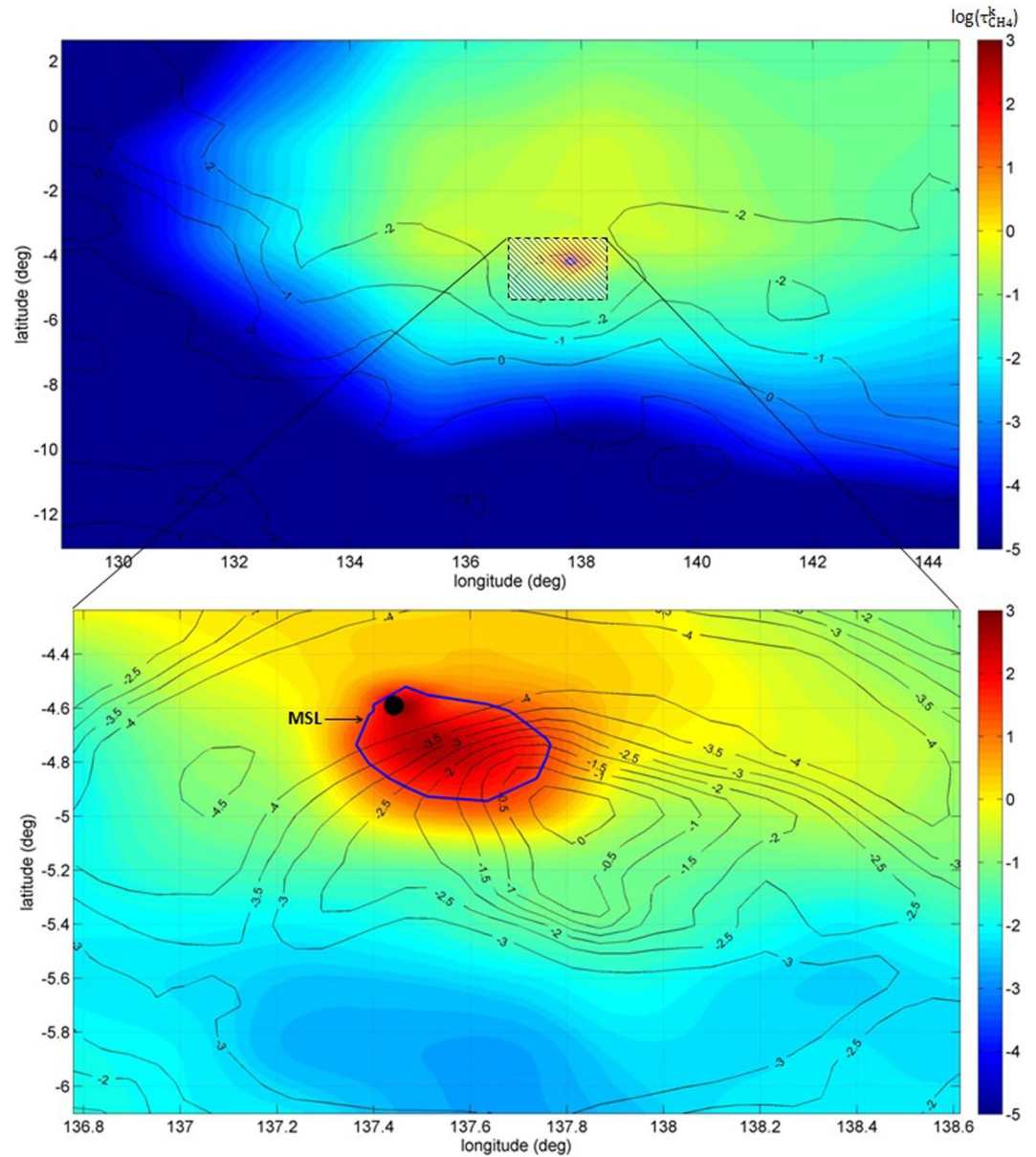


Figure 4. The required methane chemical loss time constant ($\tau_{\text{CH}_4}^k$) in the atmosphere (Earth years, in logarithmic scale), as a function of the location of the emission source of methane, based on the MSL's observations to date. The surface values were generated from the model simulations for each source location shown in Figure 1. The need for a fast loss mechanism at any source location implies a required τ_{CH_4} less of the order of a century (the blue contour line highlighted in the bottom panel), as observed outside the vicinity of MSL's general location, even without additional sources of methane at other locations on Mars. The axis has a lower limit of 10^{-5} to better observe the range in Gale Crater. Topography is shown as black contour lines in km.

fluxes per emission area resulted similar between both scenarios ($\sim 5 \times 10^{-14} \text{ kg m}^{-2}\text{s}^{-1}$), therefore most of the domain-B emissions had little influence on the levels detectable by MSL, but injected much more total methane into the global atmosphere.

Both scenarios also presented significant differences in the amount of methane that reached high altitudes. Thus, a localized emission at the northwest interior of Gale Crater achieved $X_{\text{CH}_4}^k$ values lower than 10^{-3} and 10^{-4} ppbv at 5 and 10 km, respectively (Table 1 and Figure 3), much lower than the detection limit of TGO ($X_{\text{CH}_4}^{\text{UL}}$). Simulations for nonlocalized emissions were more than an order of magnitude higher, roughly in its detection threshold. This is because methane emitted into the atmosphere from a localized emission is quickly dispersed horizontally

Table 1

Comparison Between a Scenario With a Localized Emission at MSL's General Location (Figure 1), and Nonlocalized Methane Emissions (Modeled as a Uniform Flux Emission in the Domain-B), Responsible for the Observed Methane Abundance at Gale Crater

	$\tau_{\text{CH}_4}^k$ (Earth years)	$X_{\text{CH}_4}^k$ (ppbv)	
		> 5 km	> 10 km
Localized emission at MSL's general location	Nonlimiting (~ 700) ^a	$< 10^{-3}$	$< 10^{-4}$
Full-domain (domain-B) methane emission	~ 4 ^b	~ 0.02	~ 0.002

Note. The required methane chemical loss time constant ($\tau_{\text{CH}_4}^k$) is presented for each case, in addition to the maximum volume mixing ratio reached during the daytime ($X_{\text{CH}_4}^k$) above 5 and 10 km of altitude.

^aNonlimiting value corresponds with $\tau_{\text{CH}_4}^k$, that is, it only considers an emission source at the northwest interior of Gale Crater. ^bThis value corresponds to full-domain methane emissions covering domain-B as a unique emission source on the planet.

contributing to a decrease in the levels achieved at higher altitudes, and that other regions of the crater favored methane reached higher altitudes due to their local dynamics. Although the methane plume emitted in both scenarios cannot be easily detected from the orbit based on the simulations of emissions at Gale Crater responsible for the detected levels of methane by MSL, other source emissions around the planet, if they exist, may impose higher abundances at such altitudes. In any case, both scenarios imply great difficulty for methane detection from orbit, which is particularly great for a small localized emission source. As a result, the simulations suggest that a localized emission at MSL's general location is more plausible than a nonlocalized emission source in terms of nondetection, from orbit, for the emission source responsible for methane detected by MSL.

6. Discussion of Global Considerations

The results presented in Sections 4 and 5 strongly suggest that the emission source responsible for the detected levels of methane by the MSL, both the spikes and the background observations, is localized in a relatively small region in the northwest interior of Gale Crater, probably in the northwest slopes of Aeolis Mons. This scenario is compatible with nondetection by TGO of the methane abundance that is being contemporaneously observed by the MSL in Gale Crater.

Although a detailed analysis of the planetary-scale effects of the emission source responsible for the detected methane by the MSL is out of the scope of the present paper, it is appropriate to establish a global picture of the findings presented in this study. First, it seems very unlikely that only one surface source of methane is operating on Mars. We thus also need to consider the implications of MSL-like sources at other locations. For the emission flux required to match the MSL observations and also not violate TGO detection limits, there exists a trade-off relationship between the fractional area of the planet emitting methane and the required chemical lifetime of methane. Specifically, for the lifetime based on standard gas-chemistry (~ 300 years), source locations emitting at $5 \times 10^{-14} \text{ kg m}^{-2}\text{s}^{-1}$ cannot represent more than a total emission area of $\sim 200 \text{ km}^2$, which represent 1.4×10^{-6} of the planetary surface. Limited special regions might exist, such as those involving fractured media (Oehler & Etiope, 2017; Viúdez-Moreiras, Arvidson, et al., 2020), which would reduce the effective planetary surface able to emit methane, thus increasing to some extent the aforementioned surface ratio. Nevertheless, our current knowledge argues against considering natural sources emitting methane from just a few localized emission sources. Furthermore, it simply seems improbable that the MSL just happened to land close to one of them, which could point to a potential bias in the current measurements.

However, if both methane MSL and TGO observational constraints are considered, but the chemical lifetime of methane is somehow able to be lower than the standard gas-chemistry value to the scale of a few years (see e.g., Yung et al., 2018), hundreds of localized emission sources of methane could be emitting methane around the planet at similar rates to the source responsible for the methane abundance detected by MSL. Nonetheless, this solution raises a significant problem of how much more rapid destruction of methane can operate within the general scheme of atmospheric chemistry without significantly affecting the remaining species in the atmosphere, which are correctly predicted by current chemical schemes (e.g., Lefèvre & Forget, 2009; Viúdez-Moreiras, Saiz-Lopez, et al., 2020; Yung et al., 2018). As a larger emission surface area or a larger number of emission sites

(N , each with MSL-like's emission fluxes ζ_c^k) is considered, the faster the destruction mechanism is required to be, leading to a larger challenge to the current understanding of Martian atmospheric chemistry. Furthermore, a high number of locations or larger emission surface, could lead to an increasing probability of detection by TGO of methane above the lowest atmospheric layers.

7. Conclusions

A systematic study of emission sources on Mars is performed for the first time, given the capabilities of DISVER-MAR in computing three-dimensional dispersion simulations. The model results presented in this study show that the ExoMars Trace Gas Orbiter (TGO) detection limit and the standard gas-chemistry lifetime of methane on Mars strongly constrain the scenarios that could explain the MSL observations. Several scenarios have been considered concerning hypothetical source emissions responsible for the background levels of methane detected by MSL. The model simulations strongly suggest that, taking into account the recent TGO constraints and current knowledge of methane transport and chemistry, the main emission source responsible for the background MSL observations, must be located very close to MSL in the northwest interior of Gale Crater, that is, in the northern crater rim, the crater floor and/or the slopes of Aeolis Mons, and probably in the northwestern slopes of Aeolis Mons; thus updating previous considerations about the origin of background measurements. A source location in the northwest slopes of Aeolis Mons requires emission fluxes of the order of $10^{-13} \text{ kg m}^{-2}\text{s}^{-1}$ to match MSL observations. This does not mean that other hypothetical source locations cannot also eventually affect the region, but rather that the location of the dominant source is suggested to be local.

Simulations also suggest that very small area (i.e., localized) emissions at the northwest interior of Gale Crater are more plausible than a nonlocalized emission source (e.g., from the whole Gale Crater) in terms of the TGO detection limit in the Gale Crater region and the requirement of chemical lifetime of methane. Thus, a nonlocalized emission source in the Gale crater could be more easily detected by TGO ($X_{\text{CH}_4}^k = 0.02 \text{ ppbv}$ at 5 km and $X_{\text{CH}_4}^k = 2 \times 10^{-3} \text{ ppbv}$ at 10 km vs. $X_{\text{CH}_4}^k < 10^{-3} \text{ ppbv}$ at 5 km and $X_{\text{CH}_4}^k < 10^{-4} \text{ ppbv}$ at 10 km for the localized scenario) and that it would require an enhanced loss mechanism, even if this region is the only source emitting methane on Mars ($\tau_{\text{CH}_4}^k \sim 4 \text{ Earth years}$ vs. a nonlimiting $\tau_{\text{CH}_4}^k = 700 \text{ Earth years}$ for the localized scenario). Other simulated scenarios that could be responsible for the background levels of methane detected by MSL, including methane sources far away from Gale Crater, even inside the crater but outside the northwest interior of Gale Crater, are probably incompatible with nondetection by TGO ($X_{\text{CH}_4}^k \gg 0.05 \text{ ppbv}$). Furthermore, simulations in Viúdez-Moreiras (2021a) show that a localized emission source in the northwest interior of Gale Crater is also compatible with the development of methane spikes, which are not observed from a nonlocalized emission source or from nonlocal regions in a general case, at least considering a continuous emission flux. A localized emission source in the northwest interior of Gale Crater is compatible with emissions through fracture media, for example, in the highly fractured Murray outcrops or in some of the sulfate-bearing strata on Aeolis Mons, as stated in Viúdez-Moreiras, Arvidson, et al. (2020).

However, the results also point to scenarios that are improbable or problematic. The three-dimensional simulations performed in this study indicate that methane detected by MSL would be compatible with our current understanding of the atmospheric chemistry on Mars if, and only if, the localized emission source responsible for these levels of methane is almost the unique emitting methane on Mars. This possibility seems improbable for a natural source, and would imply that the current detection of methane in the Mars atmosphere might be wrong. Thus, a natural origin of methane detected by MSL in the Mars atmosphere seems to necessarily require the invocation of an unknown loss mechanism able to destroy methane faster (even orders of magnitude) than that expected based on the standard chemistry applied to Mars (and that successfully reproduces the methane chemistry in the Earth's atmosphere), in line with what previous works have suggested (e.g., Mumma et al., 2009; Moores et al., 2019). As further emission locations around the planet are considered, the faster the destruction rate is required to be. This raises potential additional problems in atmospheric chemistry in terms of including such as yet unknown loss mechanism in the chemical scheme without affecting the remaining species that are successfully predicted to date. The fundamental problem between the potential locations emitting methane on Mars versus the hypothetical rapid destruction mechanism of methane has no answer to date.

Data Availability Statement

This is a modeling paper in which all data used to parameterize the model are described in the manuscript. The topographic maps used to input the model are obtained from the NASA's Planetary Data System (PDS) (Neumann et al., 2003). The data used for generating the figures displayed in this article are available on Mendeley Data (Viúdez-Moreiras, 2021b).

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