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The vertical transport of methane from different potential emission types on Mars

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Abstract

The contrasting evolutionary behaviour of the vertical profile of methane from three potential release scenarios is analysed using a global circulation model with assimilated temperature profiles. Understanding the evolving methane distribution is essential for interpretation of future retrievals of the methane vertical profile taken by instruments on the ExoMars Trace Gas Orbiter spacecraft. We show that at methane release rates constrained by previous observations and modelling studies, discriminating whether the methane source is a sustained or instantaneous surface emission requires at least ten sols of tracking the emission. A methane source must also be observed within five to ten sols of the initial emission to distinguish whether the emission occurs directly at the surface or within the atmosphere via destabilisation of metastable clathrates. Assimilation of thermal data is shown to be critical for the most accurate back-tracking of an observed methane plume to its origin.

Key Points

- Determining if a methane surface source is a sustained or instantaneous release requires ten sols of tracking the emission
- An instantaneous surface or atmospheric methane release is indistinguishable after around five sols of emission
- Data assimilation of thermal data is critical for the correct back-tracking of a methane plume to its source

1. Introduction

The existence of methane on Mars has recently been confirmed with an *in situ* detection by the NASA Curiosity rover [Webster *et al.*, 2015]. Recent modelling studies of methane surface release have shown that methane can form layers in the atmosphere [Viscardy *et al.*, 2016] and that surface releases of methane could potentially reconcile past observations from multiple different instruments [Holmes *et al.*, 2015]. The fact that a surface release of methane can evolve into a distinct atmospheric layer suggests that the local observations by the NASA Curiosity rover and past column observations by other instruments [Mumma *et al.*, 2009; Fonti and Marzo, 2010; Geminale *et al.*, 2011; Villanueva *et al.*, 2013] can potentially be reconcilable.

Multiple origins of methane on Mars have been hypothesised in the recent past, ranging from biogenesis [Summers *et al.*, 2002] to serpentinisation [Oze and Sharma, 2005; Atreya *et al.*, 2007] and destabilisation of methane clathrate hydrates [Chassefière, 2009]. By modelling transport of methane through the subsurface, Stevens *et al.* [2017] suggest that deep subsurface releases are not capable of providing the non-uniform sources of methane observed and that (if subsurface in origin) it must be as a result of fracturing or convective plumes from shallow sources. They also highlight that it is crucial to take into account transport processes when attempting to identify the source of any methane observed by future missions.

Future trace gas observations by the Nadir and Occultation for Mars Discovery (NOMAD) and Atmospheric Chemistry Suite (ACS) instruments on the ExoMars Trace Gas Orbiter (TGO) spacecraft will be the first to provide vertical profiles of methane, amongst multiple other species. For interpretation and understanding of the retrieved methane vertical profiles, modelling studies are required to scrutinise between the different proposed mechanisms of methane release into the

atmosphere, with global circulations models (GCMs) providing an invaluable tool to investigate the evolution of trace gas plumes and provide knowledge on where the original source could be located, and also potentially clues to its origin.

Previous modelling studies investigating the evolution of a methane source have been limited by the use of modelled wind fields [*Lefèvre and Forget, 2009; Viscardi et al., 2016*]. For the methane emission scenarios in this study, temperature retrievals from the Thermal Emission Spectrometer (TES) are assimilated using a modified form of the Analysis Correction (AC) scheme [*Lewis et al., 2007*]. Since there are no direct global wind observations, and the assimilation scheme ensures the wind fields are consistent with the thermal data input to the model, the assimilation process results in the best constraint available on the transport of tracers in the martian atmosphere and the best possible dynamical state of the atmosphere, of great importance when attempting to backtrack the methane sources to their source location. It was also shown by *Lewis et al. [2007]* that actual transient wave behaviour seen in observations is captured by the model when assimilating TES temperature profiles, rather than simply modifying the thermal state of the model to produce transient modes of different strengths and locations.

This study investigates the vertical evolution of methane from multiple different source emission scenarios, using a state-of-the-art Mars GCM coupled to the AC assimilation scheme. Section 2 details the GCM and observational data used in this investigation, with the evolving methane vertical profile from three different source emission scenarios analysed in Section 3. Finally, the difference in the evolution of the vertical profile of methane with/without the assimilation of thermal data is detailed in Section 4.

2. Methods

This section details the GCM and observational data used to construct the simulations, followed by a description of the different emission scenarios investigated in this study.

2.1. Model and observational data

The Mars GCM used for this investigation is the UK version of the LMD GCM [Forget *et al.*, 1999], hereafter MGCM, which has been developed in a collaboration between the Laboratoire de Météorologie Dynamique, the Open University, the University of Oxford and the Instituto de Astrofísica de Andalucía. This model uses physical parameterisations shared with the LMD GCM, which are coupled to a spectral dynamical core and semi-Lagrangian advection scheme [Newman *et al.*, 2002] to transport tracers. Tracers such as methane are transported by the semi-Lagrangian advection scheme with mass conservation [Priestley, 1993]. The advection scheme uses wind fields updated by the dynamical core to determine the methane concentration at each model grid point every 15 minutes.

The MGCM is similar to the model used in Holmes *et al.* [2015], but now includes additional sub-models to improve on modelling of the planetary boundary layer and water and dust cycles. A thermal plume model is used to better represent turbulent structures in the planetary boundary layer [Colaïtis *et al.*, 2013], of critical importance for the evolution of tracers (e.g. methane) released from the surface. Regarding the martian water cycle, the most recent cloud microphysics package is now included [Navarro *et al.*, 2014] which also accounts for the effects of radiatively active water ice clouds. A ‘semi-interactive’ two-moment scheme is used to freely transport dust in the model [Madeleine *et al.*, 2011], although the dust column optical depth at each grid point is scaled to match the observed dust distribution maps created by Montabone *et al.* [2015] using

an interpolation of numerous sets of observations from orbiters and landers based on a kriging method. The model is truncated at wavenumber 31 resulting in a 5° longitude-latitude grid with 35 vertical levels extending to an altitude of ~ 105 km (full details of the vertical levels used in this study are supplied in Supporting Information S1).

2.2. Simulations

Three different methane emission scenarios were used to investigate the evolutionary behaviour of the methane vertical profile from an initial source. The area and timing of the methane release was chosen to coincide with the observed methane plume in 2003 by *Mumma et al.* [2009], using the closest GCM grid point located at 50°E , 2.5°N . Two different release methods are chosen, with either sustained emission over a 30 sol period starting at $L_S = 148^\circ$ in MY 26 or an instantaneous emission in which methane is released over a single 15 minute (one physics timestep) time window. The release rate for the sustained emission scenario is constrained by the optimal scenario in *Lefèvre and Forget* [2009] for the development of the plume observed by *Mumma et al.* [2009], in which 150,000 tonnes of methane is released over a 120 sol period. The resulting release rate, equivalent to continuous emission of methane at a rate of $3.3 \times 10^{-10} \text{ kg m}^{-2} \text{ s}^{-1}$ at the surface over the 30 sol period, is of similar magnitude to the best fit release rate estimated by *Holmes et al.* [2015] when attempting to reconcile the evolution of the observed plume [*Mumma et al.*, 2009] with observations later in time from a separate instrument [*Fonti and Marzo*, 2010].

Two separate simulations are performed for the instantaneous emission scenario. The release rate of methane in both simulations is constrained by the estimated observed plume mass of 19,000 tonnes [*Mumma et al.*, 2009]. The first instantaneous emission scenario emits methane from the surface at 50°E , 2.5°N on the first model timestep and then allows for the methane

distribution to only be altered by transport processes. Comparison of the methane vertical distribution between the sustained and instantaneous surface emission scenario could potentially be used to highlight any expected differences based on the different method of release. A third simulation also uses the instantaneous emission scenario but the methane is released into the atmosphere at an altitude of 10 km instead of from the surface. In the case of destabilisation of methane clathrate hydrates being the source of methane, the release could potentially be atmospheric rather than from the surface [Chassefière, 2009]. This process suggests that methane is transported into the atmosphere from the subsurface in a metastable clathrate form and decomposes into gaseous methane through the condensation-sublimation process of water vapour. At the time and latitude of the modelled methane release, water vapour is primarily confined to altitudes below 10 km [Steele *et al.*, 2014]. Comparison of the methane vertical profile in the instantaneous surface and atmosphere emission scenarios could potentially be used to highlight if it is possible to determine whether the atmospheric methane present originated from an initial surface/atmospheric source.

Temperature profiles from the Thermal Emission Spectrometer (TES) instrument [Conrath *et al.*, 2000] on the Mars Global Surveyor spacecraft are assimilated in all three different release scenarios to provide the most realistic thermodynamical state of the atmosphere, with almost one million temperature profiles assimilated over the 30 sol period of each simulation. The TES temperature profiles cover from the surface to an altitude of ~ 40 km, encompass all latitudes and are spaced by $\sim 30^\circ$ in longitude. One final simulation identical to the instantaneous surface emission scenario except without any data assimilation is included in Section 4 to identify how

the addition of thermal profiles by the assimilation process alters the distribution of the evolving methane plume.

3. Contrasting evolution of methane vertical profile

The horizontal movement of a methane source emission was detailed in *Holmes et al.* [2015] by analysing the vertically-integrated zonal and meridional flux of methane, whilst here we investigate and compare the evolution of the methane concentration from three different emission scenarios as a function of height in the atmosphere. This approach is taken as it represents the way in which these measurements will be made in future by the ExoMars TGO mission.

The evolution of the methane volume mixing ratio vertical profile at a latitude of 2.5°N (i.e. the latitude of the initial source emission) for the three different emission scenarios is shown in Figure 1. After one sol, the sustained and instantaneous surface emission scenario, shown in Figure 1a and 1b respectively, are similarly transported, with only a difference in the amount transported as a result of the increased mass of methane added to the atmosphere in the instantaneous surface emission. Methane emitted from the surface source is predominantly transported by weak near surface easterlies to around 30°E, while the local circulation patterns result in a portion of methane greater than 10 ppb (parts per billion, by volume) reaching altitudes beyond 10 km.

The longitudinal transport of methane in the instantaneous atmosphere emission scenario (Figure 1c) is likely to be more extensive as the zonal winds at an altitude of 10 km (the altitude of the atmospheric source emission) and higher are marginally stronger than near surface winds. The complex nature of local circulation patterns as a result of turbulent structures in the planetary

boundary layer results in methane at a level of ~ 1 ppb reaching the surface at longitudes close to the initial longitude of the source (50°E).

As expected from a sustained or instantaneous surface source emission, after one sol peak amounts of methane greater than 10 ppb are seen closest to the surface with methane decreasing at increasing altitude, whereas the instantaneous atmosphere emission scenario displays a peak in methane abundance centred at around 11 km altitude. After only two sols, the methane vertical profile directly over the initial source location in the instantaneous surface emission scenario (Figure 1e) has changed drastically, with a methane abundance greater than 10 ppb centred at ~ 10 km altitude and less than 5 ppt (parts per trillion, by volume) near the surface as a result of consistent westward transport of methane near the surface at this latitude. The sustained surface emission scenario after two sols, shown in Figure 1d, still has a large methane abundance near the surface (as a result of continued emission in this scenario) which is at least two orders of magnitudes larger than in the instantaneous surface emission scenario. Comparison of the methane vertical profile 20° westward of the initial source longitude however would indicate little difference between the sustained and instantaneous surface emission scenario except in the peak methane abundance resulting from the stronger initial release in the instantaneous surface emission scenario.

Over the initial source location ($50^\circ\text{E}, 2.5^\circ\text{N}$) after two sols, the methane vertical profile in the instantaneous atmosphere emission scenario (Figure 1f) is similar to the instantaneous surface emission scenario (Figure 1e) except for a slight difference in the centre of the atmospheric layer (15 km in the instantaneous atmosphere emission scenario as opposed to 10 km). It is only by knowing the vertical profile of methane westward of the initial source longitude that we could

distinguish between the surface or atmospheric origin of methane, suggesting frequent spatial mapping of methane vertical profiles in longitudinal space are necessary.

The longitudinal extent of the initial methane source is evidently greater in the instantaneous atmosphere emission scenario when compared to the two other emission scenarios after two sols, and further evident after five sols by which time methane at levels of less than 0.5 ppb has reached longitudes greater than 180°E in Figure 1i and less than 1 ppt present for the majority of western longitudes in the sustained and instantaneous surface emission scenarios (Figure 1g and 1h respectively). After five sols, differences in the two surface emission scenarios are also becoming apparent, with less than 5 ppb methane present below 10 km near the initial source longitude in the instantaneous surface emission scenario (Figure 1h) whereas the sustained surface emission scenario in Figure 1g still has methane levels greater than 10 ppb below 10 km. This suggests that a surface emission of methane will need to be observed for at least 5 sols in order to distinguish if the initial source is sustained or instantaneous (otherwise the sustained surface emission scenario could be potentially incorrectly interpreted as a weaker/stronger instantaneous emission).

Similar peak values of 5–10 ppb of methane at 10–15 km altitude in all three emission scenarios occur in the vicinity of 0° longitude, with all three different emission scenarios displaying similar vertical profiles at this location. The atmospheric layer formed by the emission scenarios in this study at 2.5°N latitude are at a marginally lower altitude than those simulated by *Viscardy et al.* [2016], potentially as a result of the different latitude of initial emission of methane resulting in weaker ascending motion at the boundary of the ascending branch of the northern and southern Hadley cell.

The different evolutionary behaviour of a potential sustained and instantaneous surface emission scenario of methane (beginning to be distinguishable after around five sols) is more obvious after 10 sols (Figure 1j and 1k respectively). While an instantaneous surface emission scenario with a plausible release rate is relatively well mixed at levels of up to 5 ppb after 10 sols, the sustained surface release scenario now has its greatest methane abundance of more than 100 ppb near the source location. Methane levels eastward of the initial source longitude at altitudes higher than 20 km are, however, still rather similar. Ten sols after the initial instantaneous release of methane, the instantaneous surface emission scenario and instantaneous atmosphere emission scenario (displayed in Figure 1k and 1l respectively) are practically identical for the majority of the domain displayed, with differences primarily less than 1 ppb. This suggests that to determine the initial altitude of the methane source requires observing a release of methane within the first 10 sols, otherwise determining the altitudinal origin of methane will be impossible.

The analysis so far has only investigated a meridional cross-section of the methane vertical profile that contains the initial source latitude, but are differences apparent between the three different emission scenarios away from the initial source latitude? Figure 2 displays the evolution of the methane volume mixing ratio vertical profile at a latitude of 27.5°S for the three different emission scenarios. While little difference is evident after one sol between the three different emission scenarios, the instantaneous atmosphere emission scenario displayed in Figure 2c shows a peak of around 10 ppt methane at 20 km altitude. This results from stronger meridional transport of methane at higher altitudes coupled with the initial higher altitude of methane in the instantaneous atmosphere emission scenario, with a local maximum of methane greater than 5 ppb at 10 km slightly west of the initial source longitude in Figure 2f after two sols.

After five sols, methane levels of up to 5 ppb are present from 60–180°E in the instantaneous atmosphere emission scenario (Figure 2i) and methane levels are up to two orders of magnitude lower in the same region for the sustained and instantaneous surface emission scenarios (displayed in Figure 2g and 2h respectively). Westward of the initial source longitude, the methane vertical profile is similar in all three different emission scenarios, with a peak in methane generally centred at around 10–12 km. The lack of methane below 5 km in all emission scenarios indicates it would be difficult to know whether the sustained and instantaneous surface emission scenarios originated from a surface emission unless you have observed the methane plume much closer in latitude to the actual source location.

The methane vertical profile at 27.5°S latitude for the sustained and instantaneous surface emission scenarios are almost identical after five sols, but begin to diverge from one another after ten sols (see Figure 2j and 2k), with consistent levels of up to 5 ppb methane present for the majority of the atmosphere below 20 km in the sustained surface emission scenario. The timescale for divergence of a sustained and instantaneous surface emission scenario is similar to when looking at the meridional cross-section of methane vertical profile at the source latitude in Figure 1. The continued influx of methane in the sustained surface emission scenario results in methane at an abundance greater than 3 ppb from 60–120°E in Figure 2j, whilst after ten sols the instantaneous surface and atmosphere emission scenario are largely well-mixed with peak methane abundance of ~1 ppb (Figure 2k and 2l respectively). Similar to the meridional cross-section of methane vertical profile at the source latitude, ten sols after the initial emission of methane the instantaneous surface and atmosphere emission scenarios are almost indistinguish-

able, with differences generally less than 1 ppb. Marginally more methane is present at higher altitudes above 35 km in the instantaneous atmosphere emission scenario (Figure 2l).

For all three different emission scenarios, there is a higher abundance of methane in the vertical profiles at 180–30°W at a latitude of 27.5°S than at the initial source latitude (compare Figure 2j–l with Figure 1j–l). Methane is dispersed more extensively in longitude at more southerly latitudes as methane is transported by stronger zonal winds in the southern polar jet at this time of the year.

4. Impact of data assimilation on methane transport and backtracking

This section looks at the impact on the evolution of a methane plume as a result of the assimilation of TES temperature profiles. Previous studies have indicated how assimilation of thermal data is able to capture the actual transient waves [Lewis *et al.*, 2007] and modify the transient baroclinic wave behaviour in a consistent way [Lewis *et al.*, 2016], altering the weather and in particular correcting the amplitude and phase of waves in a GCM. Deviations in the methane distribution as it evolves in time with/without assimilation of temperature profiles are equivalent to deviations in the thermal structure of the atmosphere (since methane is a passive tracer). As the backtracking of methane to its source depends primarily on the circulation patterns and hence thermal structure of the atmosphere, any deviations indicated as the methane plume evolves forward in time with/without assimilation will equivalently translate to inaccuracies in locating spatially the initial source when using a modelling approach to retrace the evolved methane distribution back in time.

The above point is illustrated in Figure 3, which displays the evolution of methane for the instantaneous surface emission scenario without the assimilation of TES temperature profiles.

The assimilation of thermal profiles adjusts the local circulation patterns, and after one sol the vertical shape of the plume is already different, with a westward tilt evident in the methane plume when no thermal profiles are assimilated (compare Figure 1b with Figure 3a). Differences in the northward/southward transport of methane away from the initial source latitude also contribute to the differences in local methane abundance displayed in Figure 3b. The increased westward transport of methane continues and after two sols, methane at an altitude of ~ 10 km has already formed a distinct atmospheric layer above the equator in Figure 3d, whilst this feature is only apparent later when the thermal profiles are assimilated (Figure 1h). The dipole in deviation of local methane abundance close to the initial source location below 10 km i.e. generally more/less methane eastward/westward when thermal profiles are not assimilated in Figure 3e indicates very different vertical profiles would be retrieved over the initial source location. As the time from the initial emission increases, the observed deviation between methane abundance in the instantaneous surface emission scenario with/without assimilation of thermal profiles extends in longitude. After ten sols (compare Figure 1k and Figure 3j), peak differences in local methane abundance are around 1 ppb, which corresponds to as much as a 50% difference in local methane abundance at any point in the domain (Figure 3k), and differences are apparent across almost all longitudes.

Regarding the potential backtracking of a methane source to its initial location, one of the primary science goals of the ExoMars TGO mission, it is important to investigate the direct impact of data assimilation on the evolving wind fields. A primary source of error in the accuracy of backtracking will be in the zonal wind deviations (which in the lower atmosphere are generally larger in magnitude than meridional winds) between the simulation with/without assimilation

of thermal profiles. As a result of the regional variability of the surface of Mars [Ehlmann and Edwards, 2014], an accurate backtracking of methane is key to provide evidence of the most likely origin of the methane source. Figure 3c,f, i and l display the zonal wind speed deviation at 2.5°N of the simulation without assimilation after 1, 2, 5 and 10 sols respectively (averaged over the preceding sol) from the simulation that includes assimilation of TES temperature profiles. Changes in the zonal wind reflect the modifications to the thermal structure of the atmosphere through the assimilation process. The assimilation of thermal profiles results in a strengthening of the easterly zonal winds east of the initial source location above 10 km. The difference in zonal wind speed at the altitude of the atmospheric layer formed (between 10–20 km) is in the range of $5\text{--}10\text{ ms}^{-1}$ eastward of the initial source longitude (see Figure 3i and l), which if consistent over sols 5 to 10 of the evolution of the methane source equates to a difference of $35\text{--}70^{\circ}$ in longitude. This potential inaccuracy in locating the initial source longitude in the 10–15 km altitude range is (in this specific case study) mitigated by the negative zonal wind speed difference of a similar magnitude west of the initial source longitude, but methane which is present at marginally higher altitudes (i.e. just above 20 km altitude) will show a large difference in the initial source longitude since the deviation is generally positive over the majority of longitudinal space.

5. Conclusions

Analysis of the vertical evolution of methane for three different methane emission scenarios has been performed to determine if it is possible to distinguish between the evolution of different methane release scenarios. This is of high importance for the contextual interpretation of retrievals of the methane vertical profile, possible in the future from the NOMAD and ACS

instruments on the ExoMars TGO spacecraft, to determine the original emission location of atmospheric methane.

Using release rates of methane constrained by previous modelling studies and the spatial location of an observed methane plume [Mumma *et al.*, 2009], a comparison was made between a sustained and instantaneous surface release of methane. Distinguishing between a sustained and instantaneous surface emission requires at least ten sols of tracking the emission (otherwise a continuously emitting methane source could potentially be interpreted incorrectly as a weaker instantaneous emission).

To determine if a methane source is atmospheric rather than from the surface, which can help to identify the underlying release mechanism of methane into the atmosphere, the source of methane must be observed within ten sols of the initial release. After this timescale, the methane vertical profile for a surface or atmospheric release are practically indistinguishable from one another. An atmospheric source of methane is spread faster in latitudinal space, and so comparison of retrievals of the methane vertical profile spaced up to 30° apart in latitude can give clues to the initial altitude of the methane source, as long as the atmosphere is observed on the timescale covering less than ~5 sols after the initial methane source emission.

The longitudinal distribution of an evolving methane plume depends strongly on an accurate representation of the thermal state of the atmosphere during the evolution of the plume to capture the most realistic circulation patterns. Although the magnitude of deviations will be model-dependent, assimilation of thermal profiles from available data will be critical to provide the best estimate of the thermal state of the atmosphere which includes weather and large scale wave activity. Accurate back-tracking of a methane plume to its source location can only be

performed using assimilation of both methane vertical profiles and thermal profiles into a GCM, which will be possible using data from instruments on the ExoMars TGO spacecraft in the future.

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References

- Atreya, S. K., P. R. Mahaffy, and A.-S. Wong (2007), Methane and related trace species on Mars: Origin, loss, implications for life, and habitability, *Planet. Space Sci.*, *55*, 358–369.
- Chassefière, E. (2009), Metastable methane clathrate particles as a source of methane to the martian atmosphere, *Icarus*, *204*, 137–144, doi:10.1016/j.icarus.2009.06.016.
- Colaïtis, A., A. Spiga, F. Hourdin, C. Rio, F. Forget, and E. Millour (2013), A thermal plume model for the Martian convective boundary layer, *J. Geophys. Res. (Planets)*, *118*, 1468–1487, doi:10.1002/jgre.20104.
- Conrath, B. J., J. C. Pearl, M. D. Smith, W. C. Maguire, P. R. Christensen, S. Dason, and M. S. Kaelberer (2000), Mars Global Surveyor Thermal Emission Spectrometer (TES) observations: Atmospheric temperatures during aerobraking and science phasing, *J. Geophys. Res.*, *105*, 9509–9520, doi:10.1029/1999JE001095.

- Ehlmann, B. L., and C. S. Edwards (2014), Mineralogy of the Martian Surface, *Annu. Rev. Earth Pl. Sci.*, *42*, 291–315, doi:10.1146/annurev-earth-060313-055024.
- Fonti, S., and G. A. Marzo (2010), Mapping the methane on Mars, *Astron. Astrophys.*, *512*, A51.
- Forget, F., F. Hourdin, R. Fournier, C. Hourdin, O. Talagrand, M. Collins, S. R. Lewis, P. L. Read, and J.-P. Huot (1999), Improved general circulation models of the Martian atmosphere from the surface to above 80 km, *J. Geophys. Res.*, *104*, 24,155–24,176.
- Geminale, A., V. Formisano, and G. Sindoni (2011), Mapping methane in Martian atmosphere with PFS-MEX data, *Planet. Space Sci.*, *59*, 137–148.
- Holmes, J. A., S. R. Lewis, and M. R. Patel (2015), Analysing the consistency of martian methane observations by investigation of global methane transport, *Icarus*, *257*, 23–32, doi:10.1016/j.icarus.2015.04.027.
- Lefèvre, F., and F. Forget (2009), Observed variations of methane on Mars unexplained by known atmospheric chemistry and physics, *Nature*, *460*, 720–723.
- Lewis, S. R., P. L. Read, B. J. Conrath, J. C. Pearl, and M. D. Smith (2007), Assimilation of thermal emission spectrometer atmospheric data during the Mars Global Surveyor aerobraking period, *Icarus*, *192*, 327–347.
- Lewis, S. R., D. P. Mulholland, P. L. Read, L. Montabone, R. J. Wilson, and M. D. Smith (2016), The solsticial pause on Mars: 1. A planetary wave reanalysis, *Icarus*, *264*, 456–464, doi:10.1016/j.icarus.2015.08.039.
- Madeleine, J.-B., F. Forget, E. Millour, L. Montabone, and M. J. Wolff (2011), Revisiting the radiative impact of dust on Mars using the LMD Global Climate Model, *J. Geophys. Res. (Planets)*, *116*, E11010, doi:10.1029/2011JE003855.

- Montabone, L., F. Forget, E. Millour, R. J. Wilson, S. R. Lewis, B. Cantor, D. Kass, A. Kleinboehl, M. Lemmon, M. D. Smith, and M. J. Wolff (2015), Eight-year Climatology of Dust Optical Depth on Mars, *Icarus*, *251*, 65–95.
- Mumma, M. J., G. L. Villanueva, R. E. Novak, T. Hewagama, B. P. Bonev, M. A. DiSanti, A. M. Mandell, and M. D. Smith (2009), Strong Release of Methane on Mars in Northern Summer 2003, *Science*, *323*, 1041–1045.
- Navarro, T., J.-B. Madeleine, F. Forget, A. Spiga, E. Millour, F. Montmessin, and A. Määttänen (2014), Global climate modeling of the Martian water cycle with improved microphysics and radiatively active water ice clouds, *J. Geophys. Res. (Planets)*, *119*, 1479–1495, doi:10.1002/2013JE004550.
- Newman, C. E., S. R. Lewis, P. L. Read, and F. Forget (2002), Modeling the Martian dust cycle, 1. Representations of dust transport processes, *J. Geophys. Res.*, *107*, 5123.
- Oze, C., and M. Sharma (2005), Have olivine, will gas: Serpentinization and the abiogenic production of methane on Mars, *Geo. Res. Letts.*, *32*, L10203, doi:10.1029/2005GL022691.
- Priestley, A. (1993), A Quasi-Conservative Version of the Semi-Lagrangian Advection Scheme, *Mon. Wea. Rev.*, *121*, 621–629.
- Steele, L. J., S. R. Lewis, and M. R. Patel (2014), The radiative impact of water ice clouds from a reanalysis of Mars Climate Sounder data, *Geo. Res. Letts.*, *41*, 4471–4478.
- Stevens, A. H., M. R. Patel, and S. R. Lewis (2017), Modelled isotopic fractionation and transient diffusive release of methane from potential subsurface sources on Mars, *Icarus*, *281*, 240–247, doi:10.1016/j.icarus.2016.08.023.
- Summers, M. E., B. J. Lieb, E. Chapman, and Y. L. Yung (2002), Atmospheric biomarkers of

subsurface life on Mars, *Geo. Res. Letts.*, *29*, 2171, doi:10.1029/2002GL015377.

Villanueva, G. L., M. J. Mumma, R. E. Novak, Y. L. Radeva, H. U. Käufl, A. Smette, A. Toku-
naga, A. Khayat, T. Encrenaz, and P. Hartogh (2013), A sensitive search for organics (CH₄,
CH₃OH, H₂CO, C₂H₆, C₂H₂, C₂H₄), hydroperoxyl (HO₂), nitrogen compounds (N₂O, NH₃,
HCN) and chlorine species (HCl, CH₃Cl) on Mars using ground-based high-resolution infrared
spectroscopy, *Icarus*, *223*, 11–27.

Viscardy, S., F. Daerden, and L. Neary (2016), Formation of layers of methane in the atmosphere
of Mars after surface release, *Geo. Res. Letts.*, *43*, 1868–1875, doi:10.1002/2015GL067443.

Webster, C. R., P. R. Mahaffy, S. K. Atreya, G. J. Flesch, M. A. Mischna, P.-Y. Meslin, K. A.
Farley, P. G. Conrad, L. E. Christensen, A. A. Pavlov, J. Martin-Torres, M.-P. Zorzano, T. H.
McConnochie, T. Owen, J. L. Eigenbrode, D. P. Glavin, A. Steele, C. A. Malespin, P. Douglas
Archer Jr., B. Sutter, P. Coll, C. Freissinet, C. P. McKay, J. E. Moores, S. P. Schwenzer,
J. C. Bridges, R. Navarro-Gonzalez, R. Gellert, M. T. Lemmon, and M. S. Team (2015), Mars
methane detection and variability at Gale crater, *Science*, *347*, 415–417.

Figure 1. Meridional cross-section of methane vertical profile at 2.5°N in the sustained surface release scenario (left), instantaneous surface release scenario (middle) and instantaneous atmosphere release scenario (right) for the first (a–c), second (d–f), fifth (g–i) and tenth (j–l) sol of simulation. The dashed vertical black line indicates the longitude of the initial source emission (50°E, 2.5°N). White indicates methane at levels lower than ten parts per trillion by volume.

Figure 2. Meridional cross-section of methane vertical profile at 27.5°S in the sustained surface release scenario (left), instantaneous surface release scenario (middle) and instantaneous atmosphere release scenario (right) for the first (a–c), second (d–f), fifth (g–i) and tenth (j–l) sol of simulation. The dashed vertical black line indicates the longitude of the initial source emission (50°E, 2.5°N). White indicates methane at levels lower than ten parts per trillion by volume.

Figure 3. Meridional cross-section of methane vertical profile at 2.5°N in the instantaneous surface release scenario with no assimilation (left), the methane deviation from the instantaneous surface release scenario (middle) and the zonal wind deviation from the instantaneous surface release scenario for the first (a-c), second (d-f), fifth (g-i) and tenth (j-l) sol of simulation. The dashed vertical black line indicates the longitude of the initial source emission (50°E, 2.5°N). White in the left and middle figures indicates methane at levels lower than ten parts per trillion by volume. Zonal wind deviations are averaged over the sol preceding the sol displayed in each row.