Supporting Information for

Mars Methane Sources in Northwestern Gale Crater Inferred from Back-Trajectory Modeling

Table S1. TLS methane signals investigated in this study. Adapted from Table S2 in Webster et al. (2018).

Name	Solar longitude (degrees)	Local time at Gale crater	*In situ* methane abundance (ppbv)
Spike 1	336.12	13:55	5.78
Spike 2	55.59	03:22	5.48
Spike 3	59.20	03:22	6.88
Spike 4	72.66	02:53	6.91
Spike 5	81.84	13:26	9.34
Background level before Spike 6	265.78	01:26	0.332
Spike 6	265.91	06:29	5.55

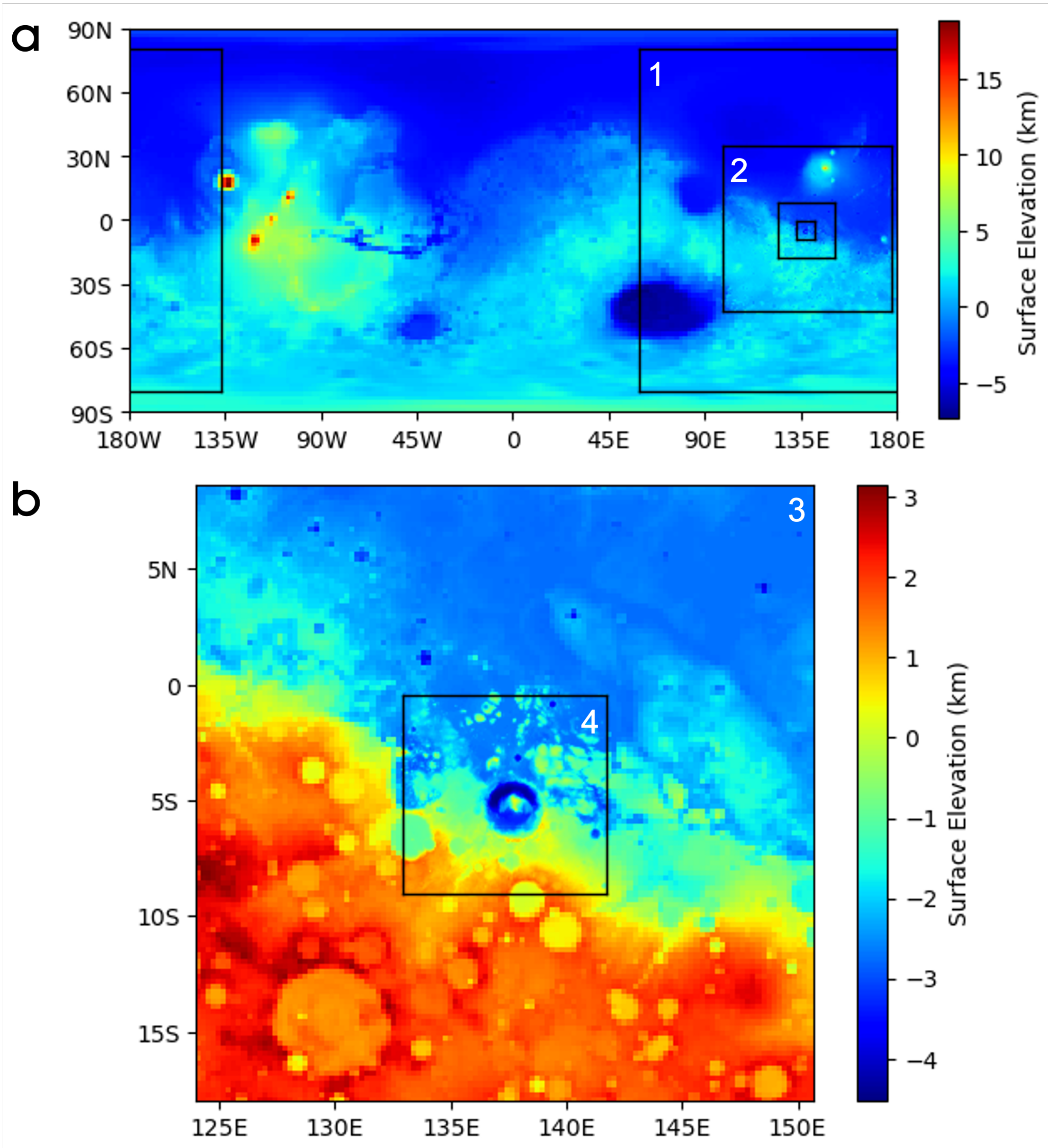

Fig. S1. MarsWRF domains on the four nesting levels. (**a**) Level 1, with a horizontal resolution of 2°×2° or 118 km×118 km, and level 2, 0.67°×0.67° or 39 km×39 km. (**b**) Level 3, 0.222°×0.222° or 13.1 km×13.1 km, and level 4, 0.074°×0.074° or 4.4 km×4.4 km. Colors show surface elevation in the corresponding horizontal resolutions of the four resolution levels.

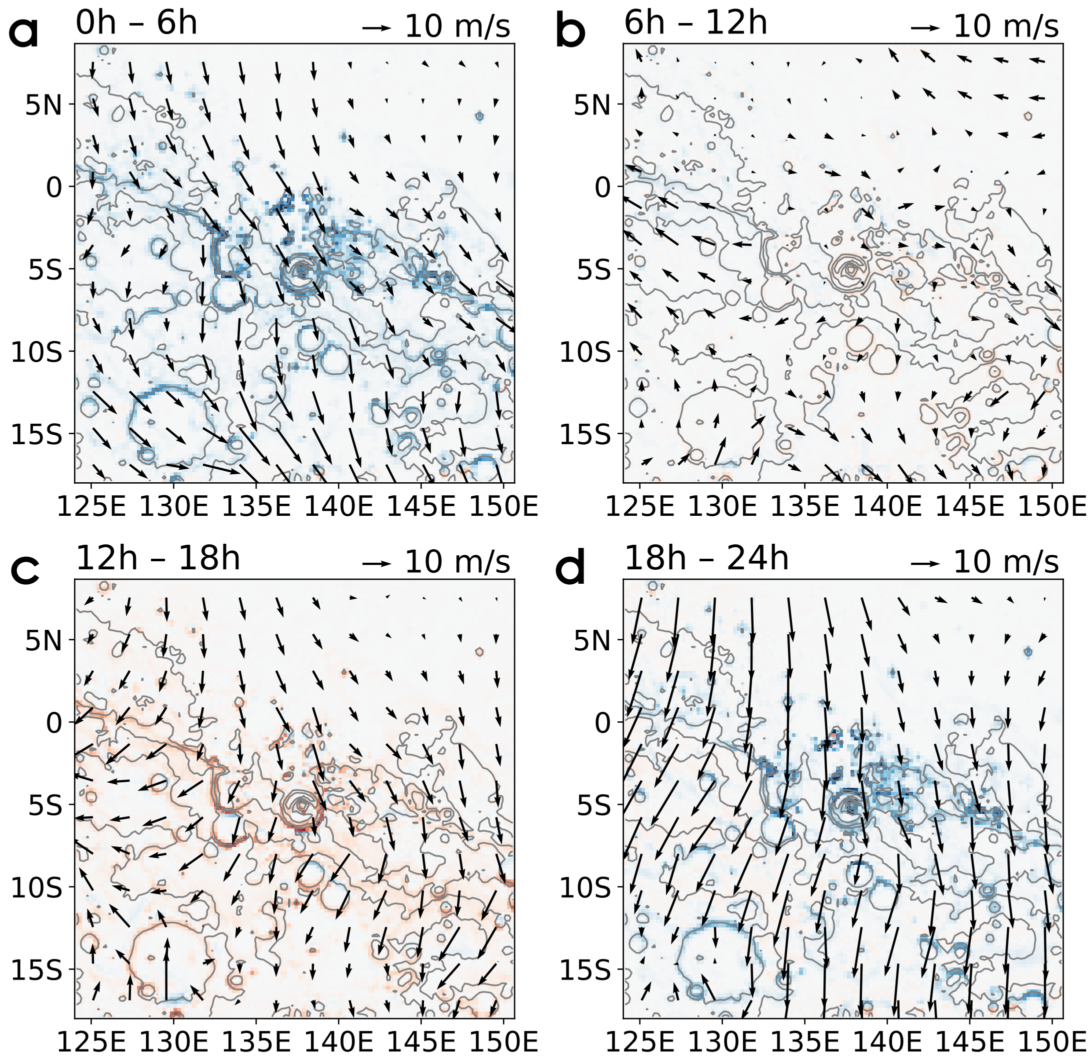

Fig. S2. MarsWRF-simulated winds averaged over the lowest 5 km of the atmosphere at 336.12° solar longitude (Spike 1). The plotted data is an average over six hours as indicated by the time period on the upper left of each subplot, and also an average over an ensemble of thirty-five sols. Weighted average by air density is performed in the vertical direction. Each arrow shows the horizontal wind averaged over a 131 km×131 km square. Red colors show rising air. Blue colors show sinking air. Contours show surface elevation. This figure can be directly compared to Fig. S17 in Giuranna et al. (2019). Between 00:00 and 06:00 local time and between 12:00 and 24:00 local time, the MarsWRF winds and the GEM-Mars winds are similar in directions – both of them are primarily northerlies, although the MarsWRF winds are stronger.

Between 06:00 and 12:00 local time, the GEM-Mars winds are weak easterlies, which was found responsible for transporting methane plumes from the regions to the east and the southeast of Gale crater to Gale (Giuranna et al., 2019). The MarsWRF winds in this time period are different, which are weak in general and do not show a dominant wind direction.

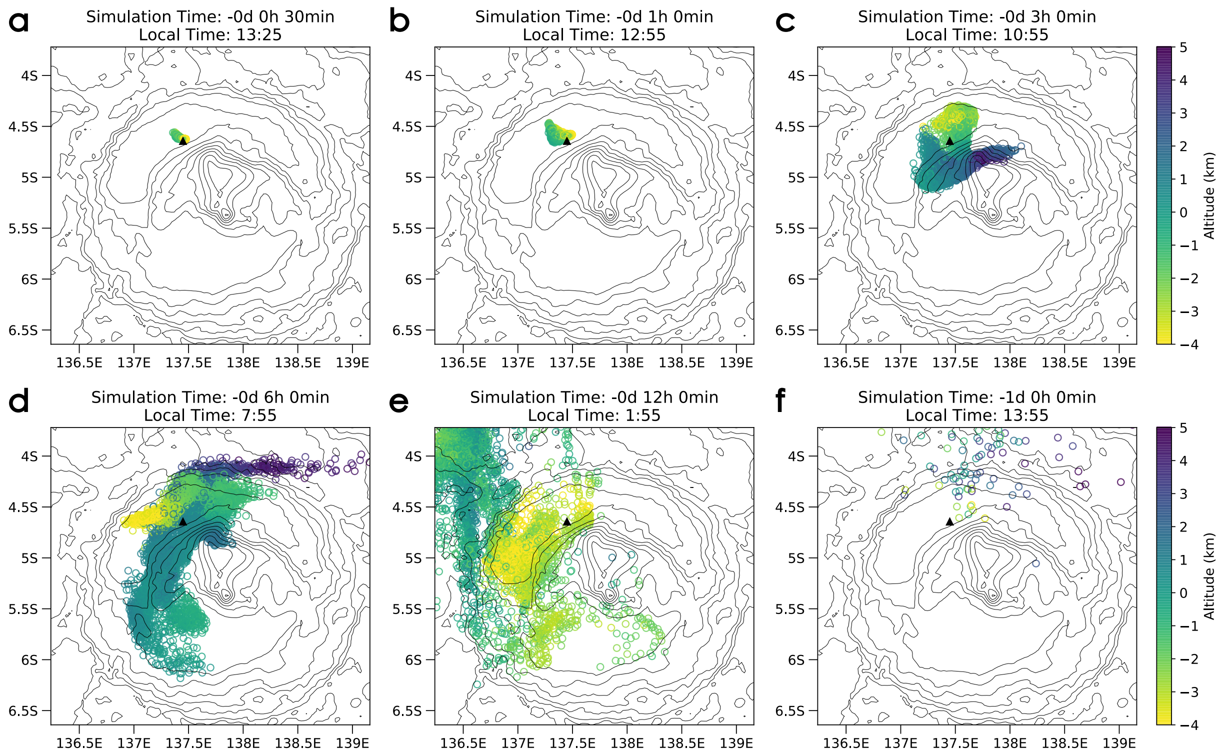

Fig. S3. Dispersion of backward-traveling particles within Gale crater (a) thirty minutes, (b) one hour, (c) three hours, (d) six hours, (e) twelve hours, and (f) one sol after particles are released. The STILT simulation is for Spike 1. Each circle shows the position of a single particle. Ten thousand particles are released in the simulation. Colors show the altitude of the particles relative to the Mars datum elevation. Contours show surface elevation. The black triangles mark the position of *Curiosity*. Almost all particles are ventilated out of Gale crater after one sol, indicating an exchange timescale of shorter than one sol, consistent with the findings in (Pla-García et al., 2019).

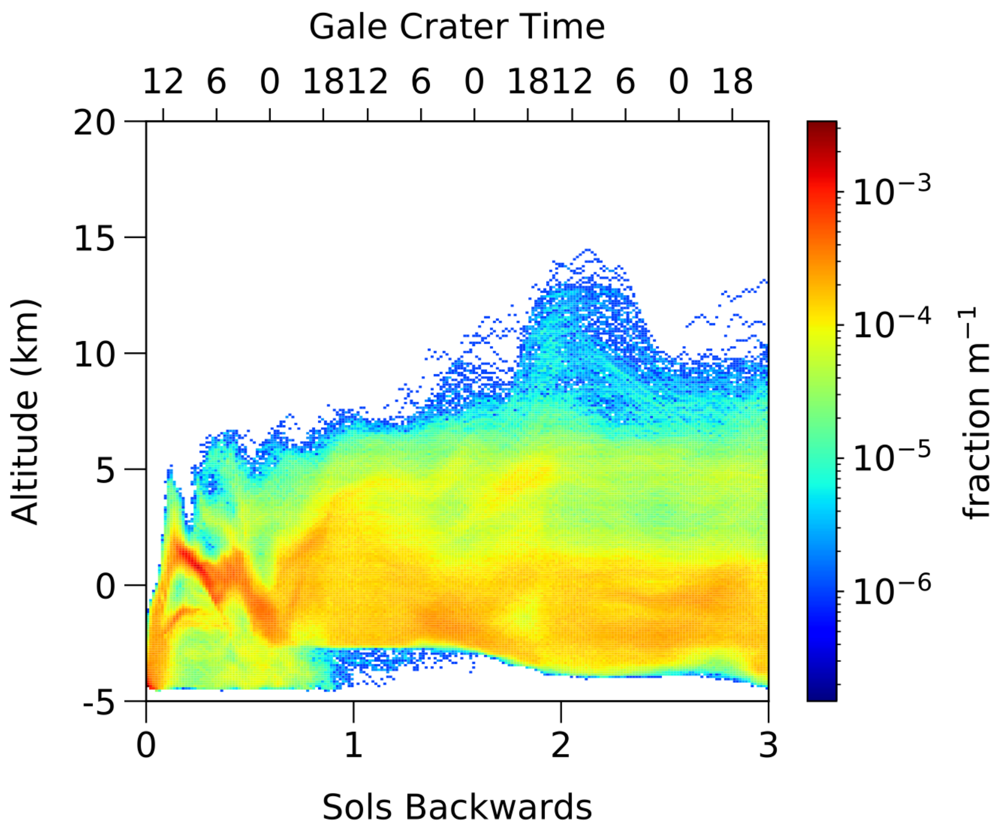

Fig. S4. Vertical dispersion of particles in a STILT simulation. An example simulation for Spike 1 is shown. Ten thousand particles are released in the simulation from the lower left corner of the figure. Colors show the fraction of the ten thousand particles with one-meter resolution in the vertical direction, indicating the number density of particles at different altitudes. Note that the particles are also dispersed in the horizontal direction, and almost all of the particles have left Gale crater one sol after the release (refer to Fig. S3). Immediately after the particle release, the majority of the particles climb up the northwestern slope of Mount Sharp, as is shown by the rapid ascent in this figure. This is consistent with the downslope wind along Mount Sharp in the early morning. In the daytime convective PBL, the convection parameterized in STILT randomly redistributes particles in the vertical direction. This results in a nearly homogeneous distribution of particles within the daytime PBL. Three sols after the release, some particles are still in contact with the surface. Extending the simulation out to thirty sols ultimately produces a homogeneous distribution across the lower atmosphere.

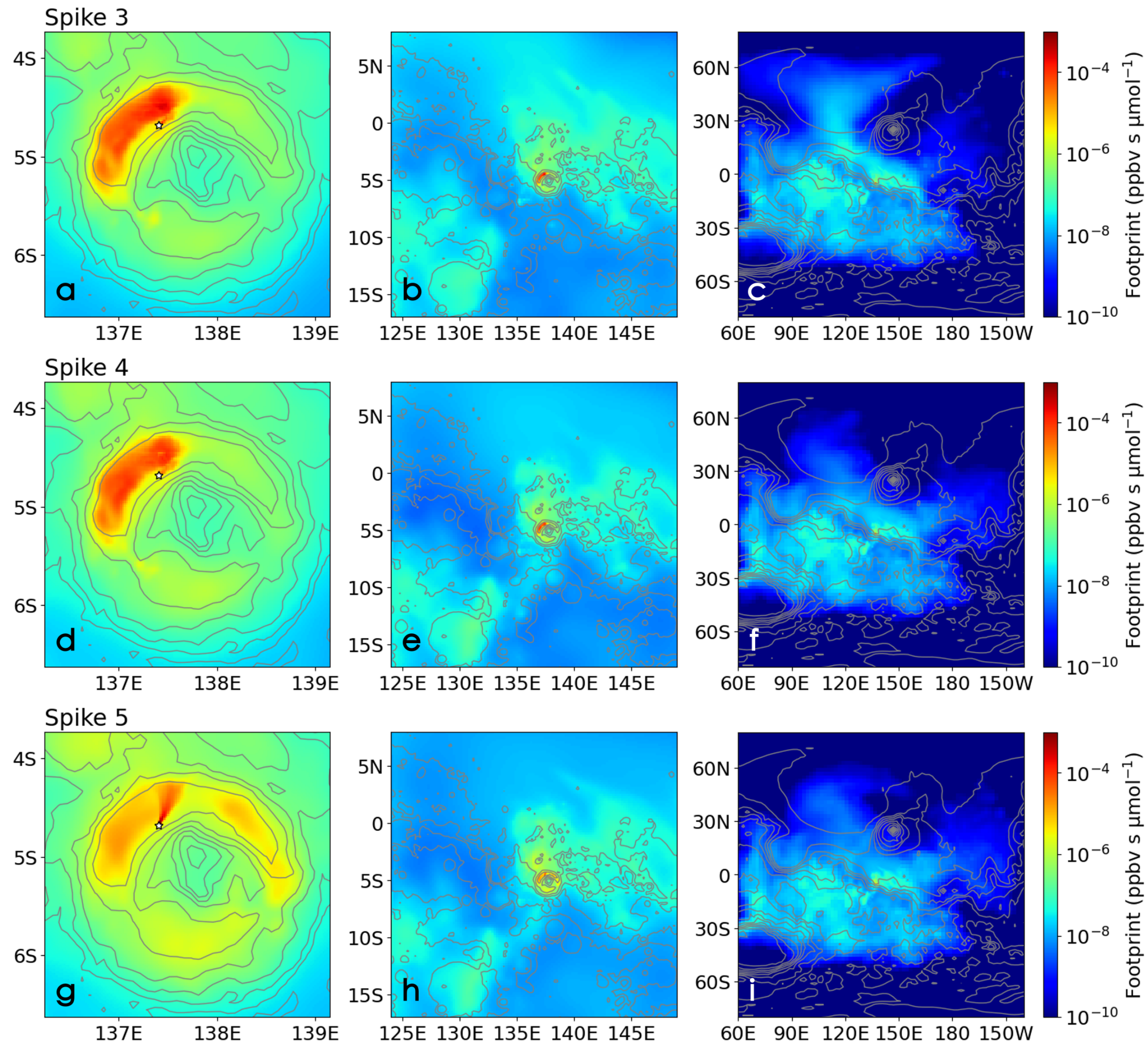

Fig. S5. Same as Fig. 3, but for (a–c) Spike 3, (d–f) Spike 4, (g–i) Spike 5.

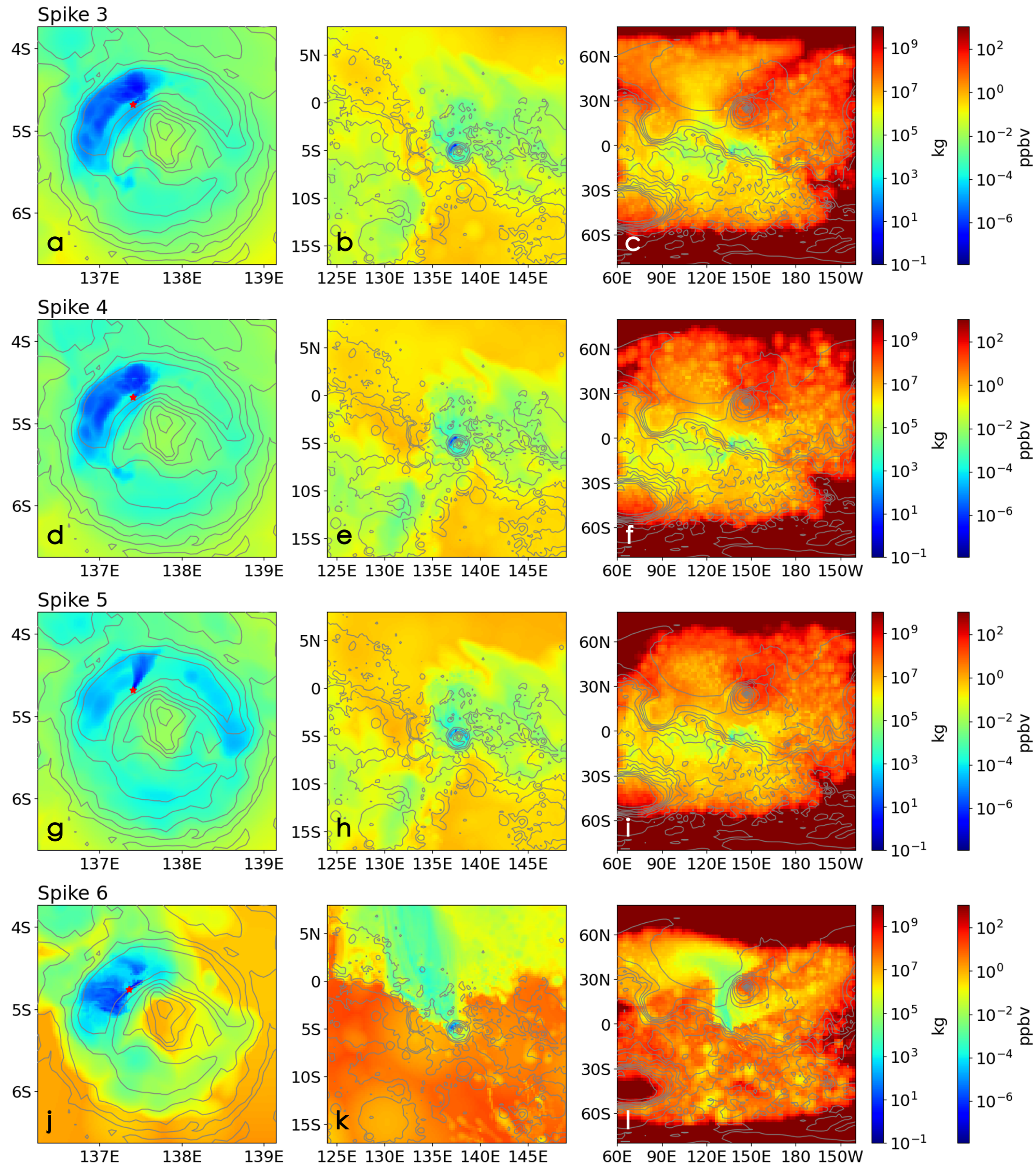

Fig. S6. Same as Fig. 4, but for (a–c) Spike 3, (d–f) Spike 4, (g–i) Spike 5, and (j–l) Spike 6.

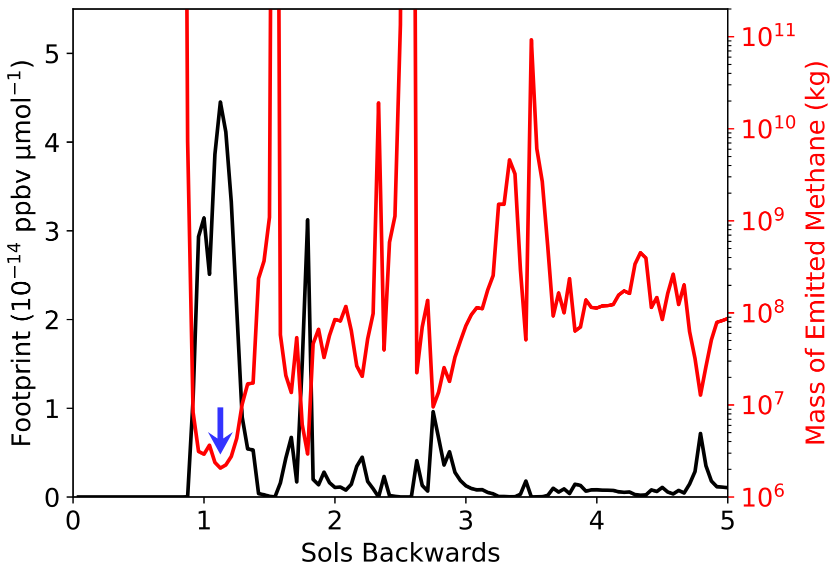

Fig. S7. An example time series of the footprint magnitude for Spike 1 at the center of the "E8" region in Giuranna et al. (2019) (4°S, 144°E). The emission site is about 390 km away from the *Curiosity* rover. The black curve shows the STILT footprint versus time backwards with respect to the time of detection. The zero footprint within the first twenty hours means that any emission that takes place at this emission site less than twenty hours before the detection will not reach the detection site at *Curiosity*. The red curve shows the mass of emitted methane that can give rise to the methane signal, assuming methane is instantaneously emitted. The blue arrow indicates the moment of maximum influence of the emission site on the methane signal, which manifests itself as the smallest emitted methane abundance. To produce Spike 1, this emission site has to emit at least two thousand tons of methane, which is equivalent to 0.23 ppbv global mean methane concentration. If the emission occurred at a random time within five sols before the detection, this figure shows that in general 10^4 to 10^5 tons of methane needs to be emitted. The emitted mass for the "E8" region found in Giuranna et al. (2019) is 1170 to 2740 tons, which is about the same as the lower limit estimated in this study.

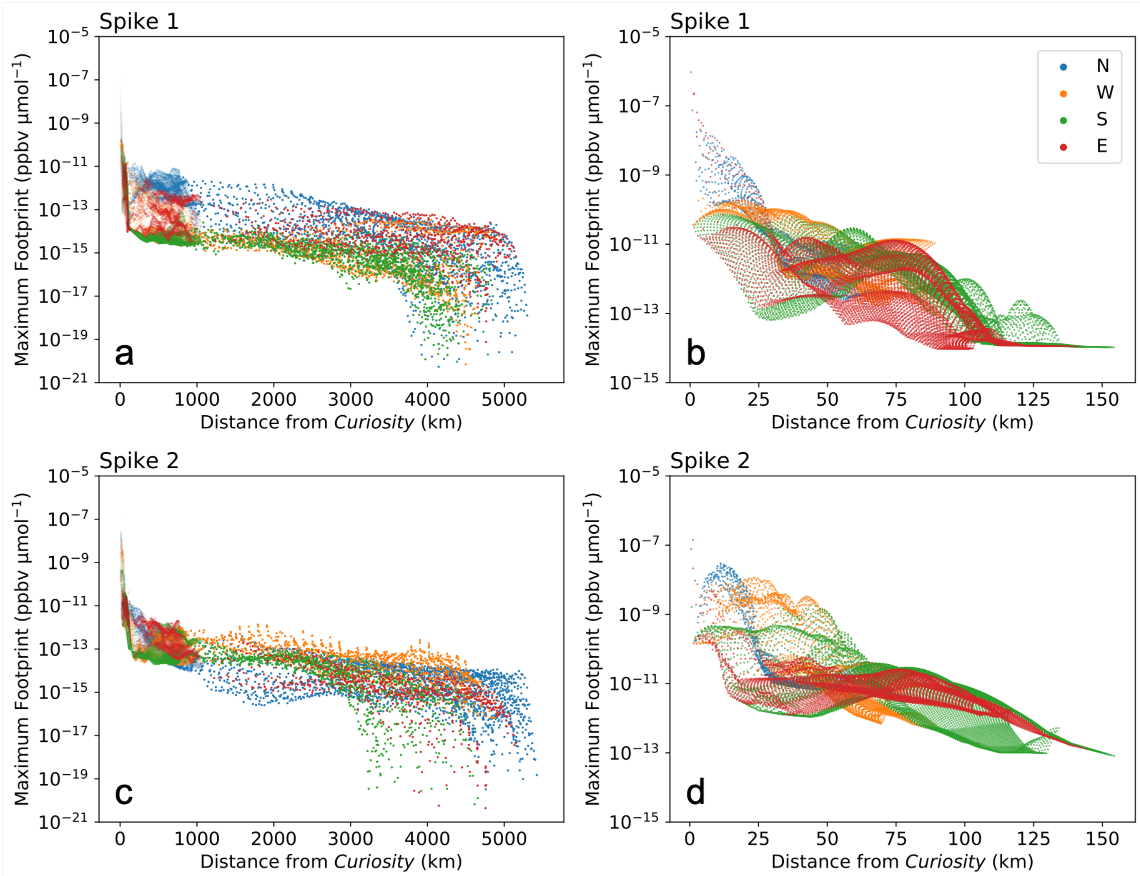

Fig. S8. Decay of STILT footprint with distance. Shown is the maximum STILT footprint (**a**) and (**b**) for Spike 1 and (**c**) and (**d**) for Spike 2 at every putative emission site around the detector versus the distance between the emission site and the detector. (**a**) and (**c**) show all the emission sites in Fig. 3(**c**). (**b**) and (**d**) only show emission sites within and in the vicinity of Gale crater. Blue dots indicate emission sites in the northern quadrant, yellow dots, in the western quadrant, green dots, in the southern quadrant, and red dots, in the eastern quadrant. The drop of footprint magnitude at long distances (> 3000 km) in (**a**) and (**c**) is due to an insufficient number of particles that reach these distant places in the STILT simulations. It is found that the maximum footprint decays rapidly with distance while within Gale crater. Outside Gale crater, the decay is slower. This figure demonstrates of the necessity of modeling atmospheric dispersion when one wants to build a linkage between emission fluxes and methane signals, because the linkage strongly depends on the distance between the emission site and the detector.

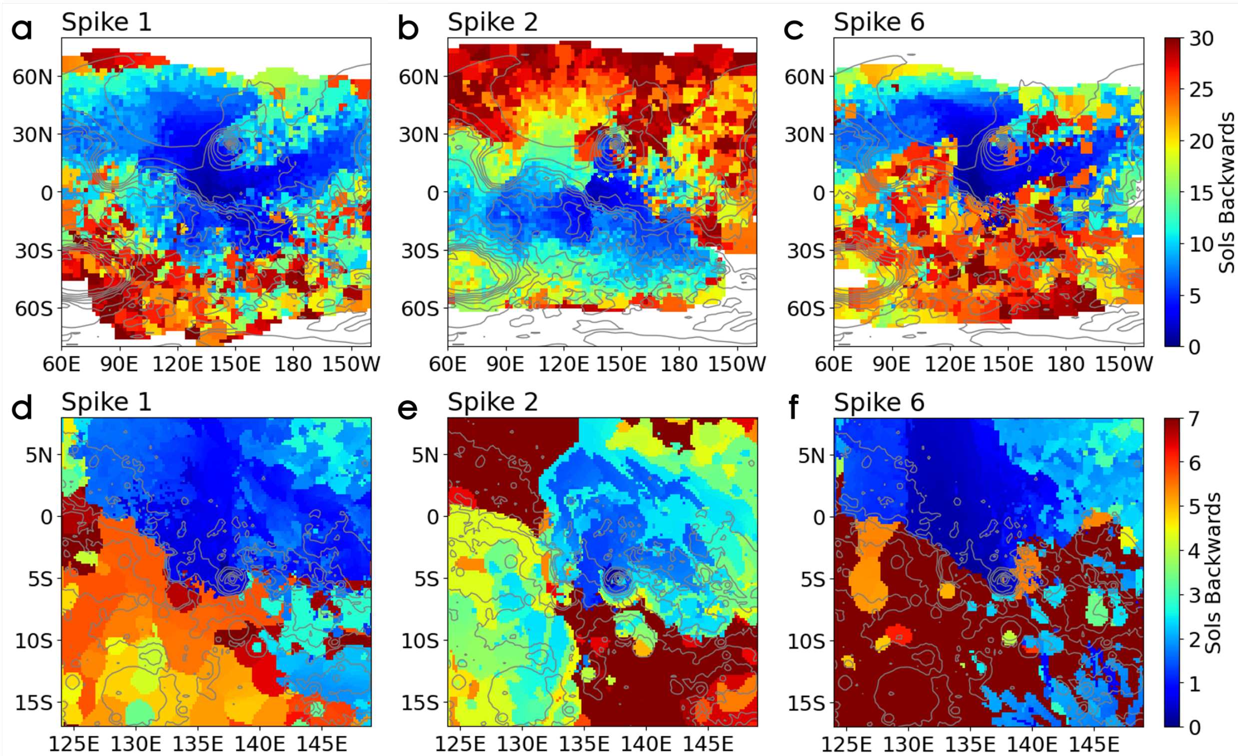

Fig. S9. Time of maximum STILT footprint for (a) and (d) Spike 1, (b) and (e) Spike 2, and (c) and (f) Spike 6 at all emission sites. One can refer to the blue arrow in Fig. S7 for the meaning of "maximum STILT footprint". This figure shows the transport timescales. (**a**–**c**) show the entire domain of the simulation. (**d**–**f**) zoom into the vicinity of Gale crater. It takes less than one sol to transport methane signals emitted from anywhere within Gale crater to the *Curiosity* rover. For Spikes 1 and 6, it takes less than one week to transport methane signals emitted from Elysium Planitia, Utopia Planitia, and Amazonis Planitia to the detector, whereas it can take up to a few weeks to more than one month to transport methane signals emitted from the southern highlands to the detector. For Spike 2, it takes about one week to transport methane signals emitted from Hesperia Planum and Terra Cimmeria to the *Curiosity* location, whereas it can take up to a few weeks or longer to transport methane signals emitted from the northern lowlands to the detector. The figures for Spikes 3, 4, and 5 are similar to those for Spike 2.

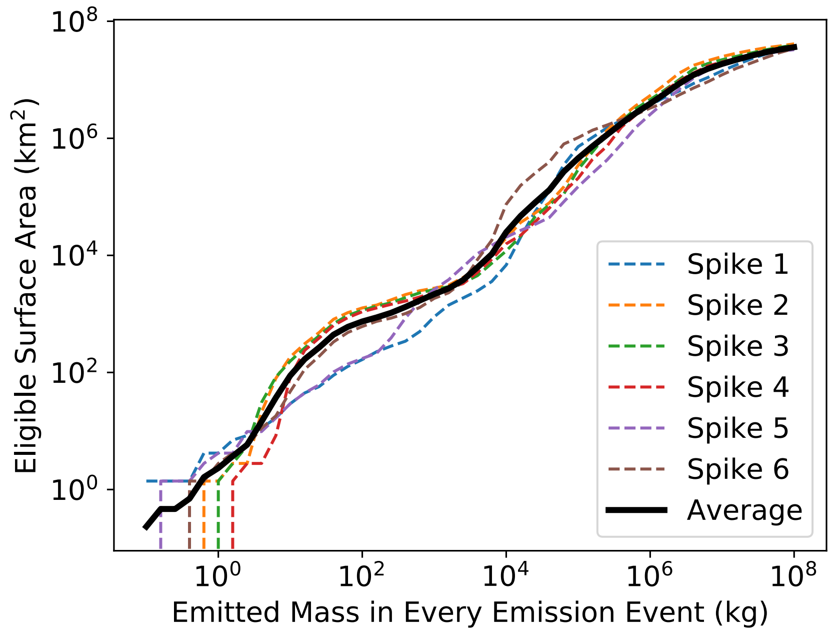

Fig. S10. The maximum area of the emission regions where certain amounts of emitted methane can produce the observed methane spikes. This figure shows the information in Fig. 4 and Fig. S6 from another perspective. Recalling that TGO reported an upper limit of 0.02 ppbv for the long-term steady-state methane abundance in the atmosphere assuming methane is a long-lived species, if the lifetime of methane is 330 years as suggested by standard photochemical models, no more than ~520 kg of methane is replenished in the atmosphere every year on average. TLS detected six methane spikes during its 4.6 Earth years of operation, meaning that no fewer than six emission events happened during this period of time (and possibly many more). Should the emission events not significantly perturb the long-term steady state methane abundance, no more than 400 kg of methane can be emitted by each emission event. This figure shows that only an area of 1300 km^2 around the detector (about 7% the total area of Gale crater) is able to produce an observed methane spike by emitting 400 kg of methane. This quantitatively demonstrates how lucky we were to send *Curiosity* to the exact place that is very close to an active surface emission site. Alternatively, there must be fast methane removal mechanisms at work, or either the TLS spikes or the TGO upper limits need to be reevaluated.