

Figure S1. Monthly TROPOMI sampling density at $0.25^\circ \times 0.3125^\circ$ resolution (~ 25 km) between May 2018 and April 2019, after filtering for data quality and clouds. The total number of observations, and percent over-land grid cell data coverage, is indicated in each panel.

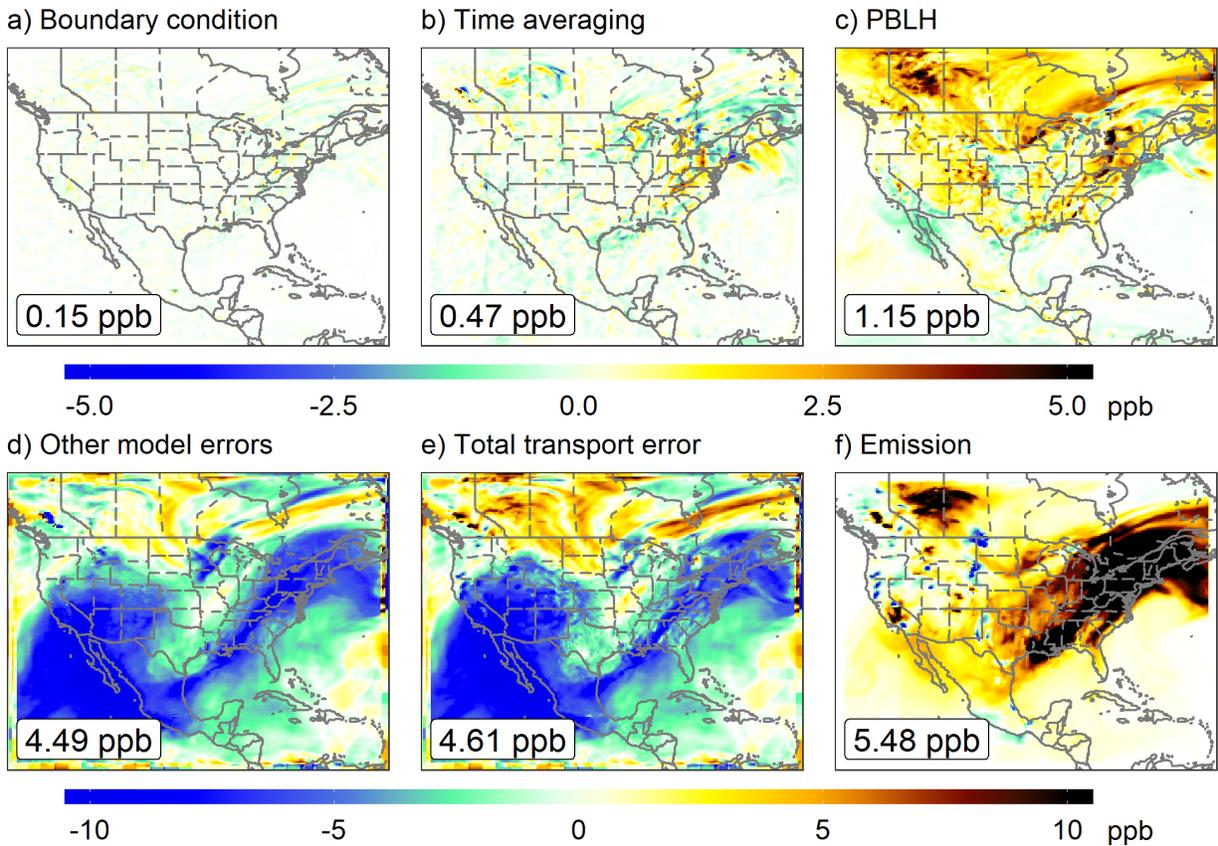


Figure S2. Methane column concentration differences for 2018-08-01 resulting from the individual model transport errors employed in the OSSE (see Sect. 2 for details). Shown are differences incurred from: a) using 6 versus 3 buffer grid cells at the domain boundary; b) averaging over 13:00-14:00 LT versus sampling the model instantaneously at the satellite overpass time; c) employing non-local versus full PBL mixing schemes; d) alternate convection and tropopause treatments; and e) all model transport errors. Shown for comparison are f) the column differences that arise from employing the true versus spatially biased prior emissions. The root-mean-square errors (RMSE) relative to the true fluxes are labeled in each panel.

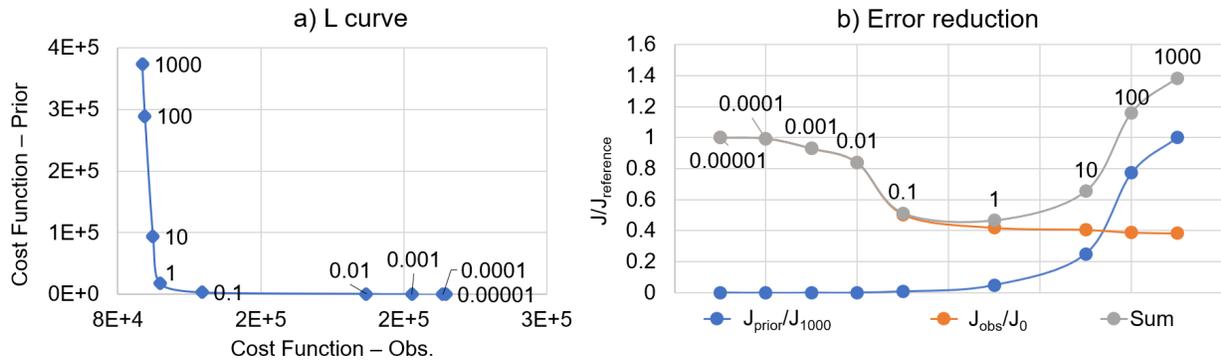


Figure S3. Cost function analysis and determination of the regularization parameter γ based on one-week inversions with spatially uniform prior errors. Panel a) shows the L curve comparing the penalty and observational deviation terms in the cost function as a function of γ . Panel b) shows the penalty term divided by the total cost function computed at $\gamma = 1000$ (where the solution is mostly determined by the observations; blue line), the observational term divided by the total cost function computed at $\gamma = 0$ (where the solution is solely determined by the prior; orange line), and the sum of the blue and orange lines (in grey).

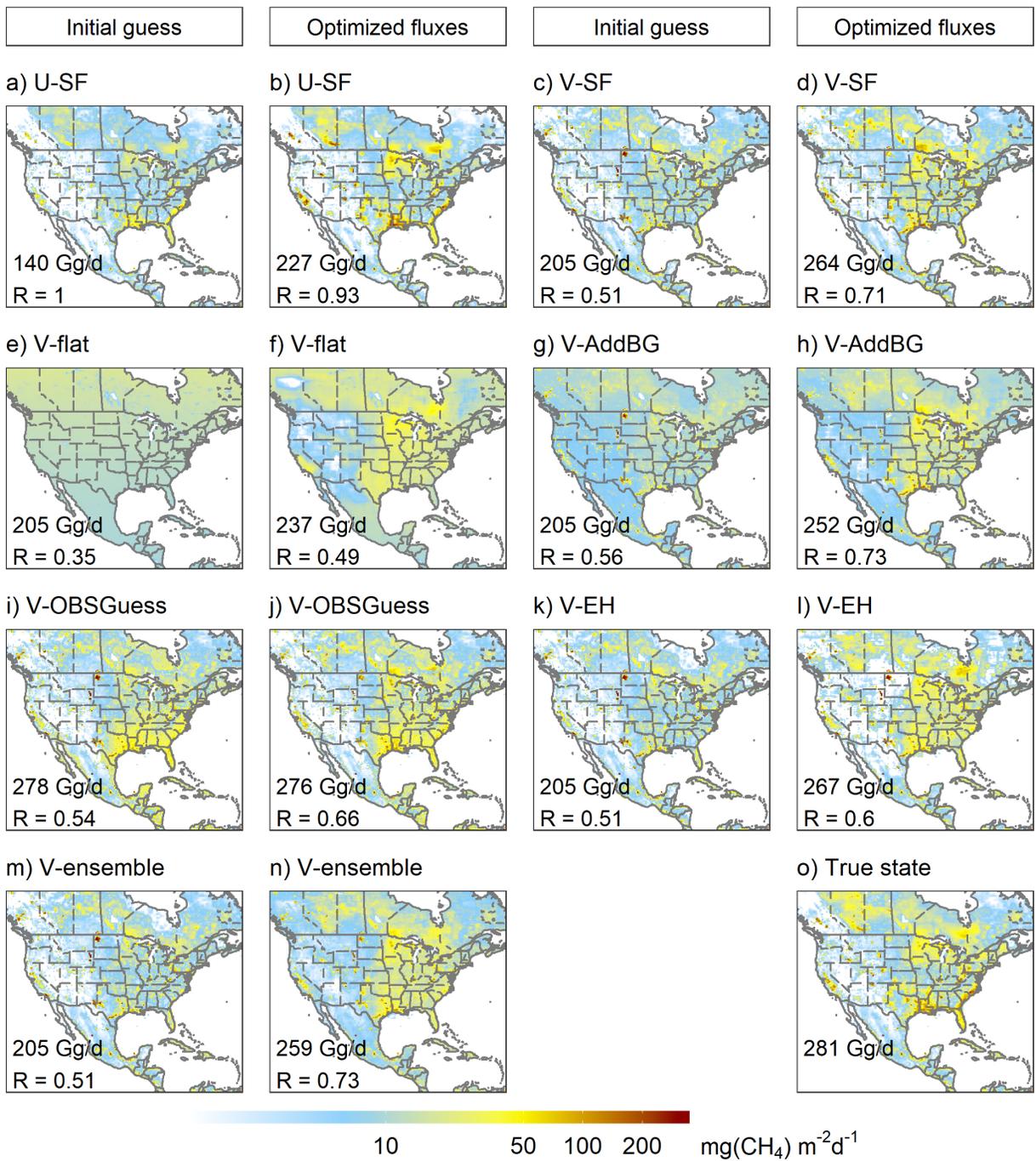


Figure S4. Initial guess and optimized emissions for each inversion framework. Labels inset indicate the domain-wide total emissions and spatial correlation to the true fluxes.

Emission increments

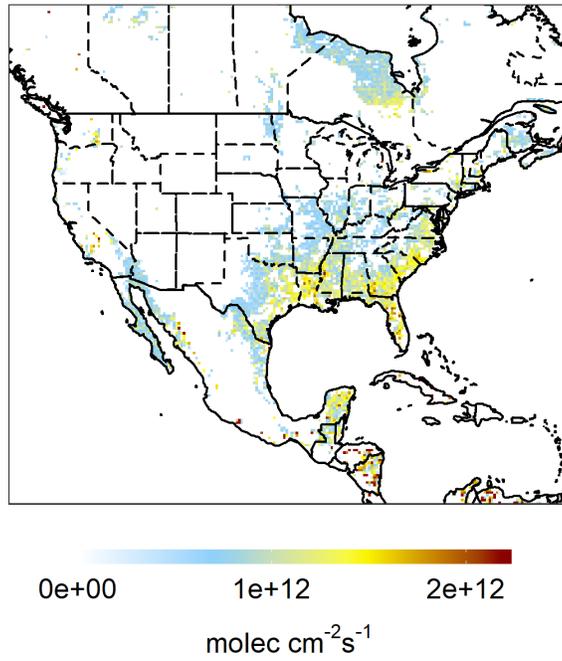


Figure S5. Emission increments added to the prior fluxes in the V-OBSGuess inversions. See Sect. 4 for details.

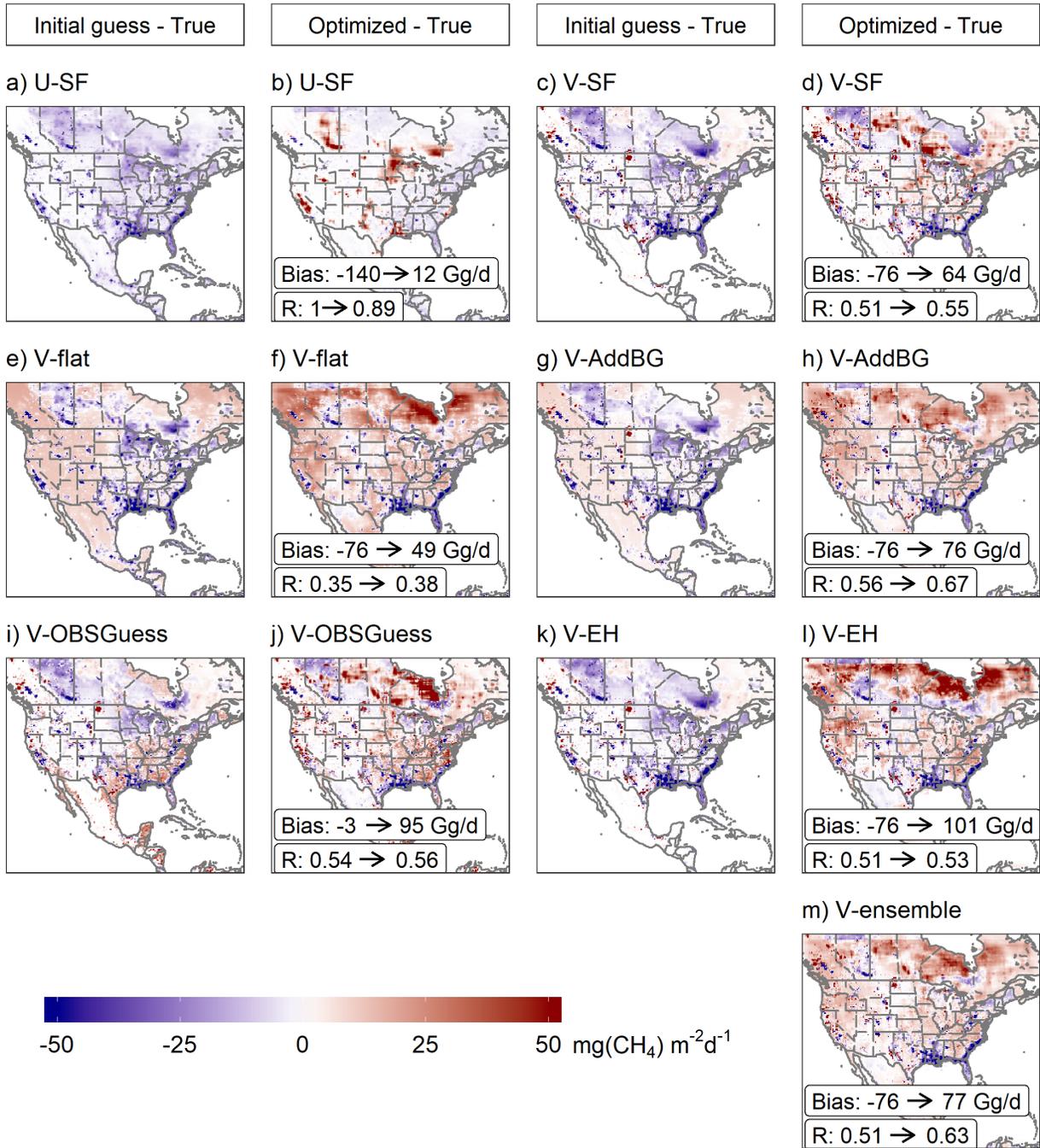


Figure S6. Same as Fig. 4, but showing results with model transport error.

Table S1. Filters applied for TROPOMI data quality assurance

| Parameter | Range |
|--|--------------------------------|
| Solar Zenith Angle (SZA) | $\leq 70^\circ$ |
| Viewing Zenith Angle (VZA) | $\leq 60^\circ$ |
| Surface albedo | ≥ 0.02 |
| Aerosol Optical Thickness (AOT) | < 0.3 |
| XCH ₄ precision (noise-related error) | < 10 ppb |
| Signal-to-noise ratio | ≥ 50 |
| χ^2 | < 100 |
| Fraction of non-corrupted/unphysical spectral pixel ¹ | $\geq 70\%$ |
| Cloudiness level | Confidently clear ¹ |

¹Pixel quality is determined per orbit, for details see the TROPOMI Product User Manual (2019).

Reference

TROPOMI product user manual: <http://www.tropomi.eu/data-products/methane/>, last access: 15 April 2019.