



# How well can inverse analyses of high-resolution satellite data resolve heterogeneous methane fluxes? Observing system simulation experiments with the GEOS-Chem adjoint model (v35)

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**Abstract.** We perform observing system simulation experiments (OSSEs) with the GEOS-Chem adjoint model to test how well methane emissions over North America can be resolved using measurements from the TROPOspheric Monitoring Instrument (TROPOMI) and similar high-resolution satellite sensors. We focus analysis on the impacts of (i) spatial errors in the prior emissions and (ii) model transport errors. Along with a standard scale factor (SF) optimization we conduct a set of inversions using alternative formalisms that aim to overcome limitations in the SF-based approach that arise for missing sources. We show that 4D-Var analysis of the TROPOMI data can improve monthly emission estimates at 25 km even with a spatially biased prior or model transport errors (42 %–93 % domain-wide bias reduction;  $R$  increases from 0.51 up to 0.73). However, when both errors are present, no single inversion framework can successfully improve both the overall bias and spatial distribution of fluxes relative to the prior on the 25 km model grid. In that case, the ensemble-mean optimized fluxes have a domain-wide bias of  $77 \text{ Gg d}^{-1}$  (comparable to that in the prior), with spurious source adjustments compensating for the transport errors. Increasing observational coverage through longer-timeframe inversions does not significantly change this picture. An inversion formalism that optimizes emission enhancements rather than scale factors exhibits the best performance for identifying missing sources, while an approach combining a uniform background emission with the prior inventory yields the best performance in terms of overall spatial fidelity – even in the presence of model transport errors. However, the standard

SF optimization outperforms both of these for the magnitude of the domain-wide flux. For the common scenario in which prior errors are non-random, approximate posterior error reduction calculations (derived via gradient-based randomization) for the inversions reflect the sensitivity to observations but have no spatial correlation with the actual emission improvements. This demonstrates that such information content analysis can be used for general observing system characterization but does not describe the spatial accuracy of the posterior emissions or of the actual emission improvements. Findings here highlight the need for careful evaluation of potential missing sources in prior emission datasets and for robust accounting of model transport errors in inverse analyses of the methane budget.

## 1 Introduction

Increases in atmospheric methane since the pre-industrial era have enhanced global radiative forcing by  $0.97 \text{ W m}^{-2}$ , making it the second-most important anthropogenic greenhouse gas after carbon dioxide (IPCC, 2013). However, the limited spatial coverage of observations has made it difficult to constrain emission distributions (Dlugokencky et al., 2011; Dlugokencky, 2020; Kirschke et al., 2013; Saunio et al., 2020). New space-based measurements from TROPOMI (TROPOspheric Monitoring Instrument) provide near-global high-precision methane column observations at 7 km resolution,

potentially filling this gap. In this study, we present observing system simulation experiments (OSSEs) using a range of inversion strategies to explore the capabilities and limitations of high-resolution satellite-based column measurements for spatially resolving methane sources across North America.

Bottom-up methane emissions contain significant uncertainties. Recent global estimates for the 2008–2017 period range from 594–880 Tg yr<sup>-1</sup> (Saunois et al., 2020), with major disparities in spatial distribution. For example, while the total US anthropogenic methane flux in the gridded Environmental Protection Agency inventory (GEPA: 29 Tg yr<sup>-1</sup> for 2012; Maasakkers et al., 2016) is within 15 % of the corresponding estimate from the Emissions Database for Global Atmospheric Research v5 (EDGAR: 25 Tg yr<sup>-1</sup> for 2015; Crippa et al., 2019; EDGAR v5, 2019), these two datasets have a spatial correlation of just  $R = 0.08$  at  $0.1^\circ \times 0.1^\circ$  resolution. Such spatial errors limit our ability to diagnose the reasons for model–measurement disparities and thus hinder regional mitigation policies.

Atmospheric inversions are critical for testing and improving bottom-up methane flux estimates but carry their own uncertainties. Global top-down methane emission estimates for 2008–2017 range from 550–594 Tg yr<sup>-1</sup> and diverge substantially in their attribution of fluxes to source sectors, with differences of up to a factor of 2 (e.g., 21–50 Tg yr<sup>-1</sup> for non-wetland natural emissions; Dlugokencky et al., 2011; Saunois et al., 2020). Such uncertainties also manifest on regional scales. For example, recent top-down estimates for the US based on satellite (Greenhouse Gases Observing SATellite, GOSAT; Scanning Imaging Absorption Spectrometer for Atmospheric Chartography, SCIAMACHY) and in situ (tall tower, aircraft) measurements have varied between 30–45 Tg yr<sup>-1</sup> for different years, with differing source allocation (Maasakkers et al., 2021; Miller et al., 2013; Turner et al., 2015; Wecht et al., 2014a). Such disparities also manifest in other regions (Dlugokencky et al., 2011; Kirschke et al., 2013).

Prior spatial emission errors present one major barrier to top-down flux estimation. Inverse analyses commonly employ Bayesian scale factor (SF) optimization to improve flux estimates based on model–measurement concentration mismatches (Chen et al., 2018, 2021; Deng et al., 2014; Hooghiemstra et al., 2012; Jacob et al., 2016; Li et al., 2019; Maasakkers et al., 2021; Turner et al., 2015; Wecht et al., 2014a; Yu et al., 2021a; Zhang et al., 2018). However, this approach fails where emissions are either missing entirely in the prior inventory or are too low to sufficiently adjust without incurring a prohibitive cost function penalty (Chen et al., 2018). In such cases the optimization will then tend to attribute the required emissions to locations with higher prior emissions that require only modest adjustment and involve a smaller penalty (Jacob et al., 2016). Previous studies (e.g., Maasakkers et al., 2016; Turner et al., 2015) have further shown that prior spatial uncertainties in the sectoral allocation of emissions limit the accuracy of top-down source

attributions. Employing normalized spatial surrogates (e.g., livestock distribution) as prior constraints can eliminate the dependence on bottom-up inventories (Michalak et al., 2004; Miller et al., 2013), but the same limitations will apply given uncertainty in the spatial surrogates or variability in their relationship to fluxes.

Limited observational coverage has historically presented another major limitation to top-down methane analyses and exacerbates the prior dependencies outlined above. Ground-based networks provide a high-precision, temporally dense, and long-term record of atmospheric methane concentrations at globally distributed sites (NOAA ESRL GMD, 2021; Prinn et al., 2021; WMO WDCGG, 2021). However, these observations are spatially sparse compared to the heterogeneity of emissions. Airborne measurements (e.g., Davis et al., 2018; Gonzalez et al., 2021; Karion et al., 2015; Knox et al., 2019; Kort et al., 2008; Miller et al., 2013; Sheng et al., 2018b; Smith et al., 2017; Wofsy et al., 2018; Yu et al., 2020, 2021a) expand this spatial footprint but only for discrete snapshots. Satellite measurements to date have generally also provided only limited coverage. For example, short-wave infrared (SWIR) methane measurements from GOSAT are separated in space by 260 km (Kuze et al., 2016), while Greenhouse Gas Satellite (GHGSat) observations are high-resolution ( $0.05^\circ \times 0.05^\circ$ ) but measure only a limited set of targets. Thermal infrared (TIR) measurements provide broad sampling but with limited sensitivity to methane emissions (Jacob et al., 2016).

High-resolution measurements from TROPOMI offer a major advance over earlier satellite observations for mapping methane emissions. Prior OSSEs have demonstrated this potential. For example, Wecht et al. (2014b) and Sheng et al. (2018a) found that TROPOMI observations can provide comparable methane emission constraints as dedicated aircraft measurements spanning the same time intervals and regions. Other analyses have concluded that 1 week of TROPOMI methane observations is sufficient to resolve time-invariant fluxes at 30 km (Turner et al., 2018) and to achieve 100 % error reduction over emission hotspots (Bousserez et al., 2016), while a single satellite overpass is able to monitor the 20 highest-emitting locations in the GEPA inventory (Jacob et al., 2016). However, the above work has focused primarily on resolving emission magnitudes without explicitly considering the impacts of spatial errors.

Here, we apply the GEOS-Chem adjoint model in an OSSE framework to characterize the capabilities and limitations of TROPOMI and similar space-based sensors for resolving spatiotemporal patterns in methane emissions across local-to-regional scales. We perform an ensemble of synthetic inversions over North America and specifically assess the ability of the observing system to spatially quantify heterogeneous emissions given limited confidence in their prior distributions. We further evaluate multiple alternative inversion frameworks in terms of their strengths and weaknesses

in this context relative to the standard and widely used SF approach.

## 2 Methods

Figure 1 summarizes the OSSE framework employed here. We apply 1 month of synthetic TROPOMI observations over North America, with realistic instrument noise and sampling coverage, to evaluate the ability of different inversion frameworks to recover the true distribution of methane sources. We test the impact of spatial biases by supplying each inversion with incorrect (but plausible) prior flux fields, both in the presence and absence of model transport errors. As is broadly the case for source inversions, the framework here is formally inconsistent with the best linear unbiased-estimate (BLUE) 4D-Var problem. In general, we do not have an unbiased estimate of the prior, so the solution will not be the BLUE. As a practical matter we thus seek to evaluate alternative inversion formalisms in terms of their ability to recover the true emissions in spite of this limitation.

### 2.1 TROPOMI observations

TROPOMI is onboard Copernicus Sentinel-5 Precursor, a low-Earth polar-orbiting satellite launched in October 2017 with 13:30 local solar overpass time (LT). TROPOMI has a 2600 km swath width and provides near-global daily measurements at  $7 \times 7 \text{ km}^2$  nadir resolution in the shortwave infrared (SWIR) and  $7 \times 3.5 \text{ km}^2$  nadir resolution in the near infrared (NIR) (Hu et al., 2018). Methane columns are retrieved from NIR and SWIR spectral radiances with an estimated uncertainty of 1 % due to instrument noise (0.6 %) and forward model error (0.8 %) (Hu et al., 2016, 2018; Lorente et al., 2021; TROPOMI, 2021).

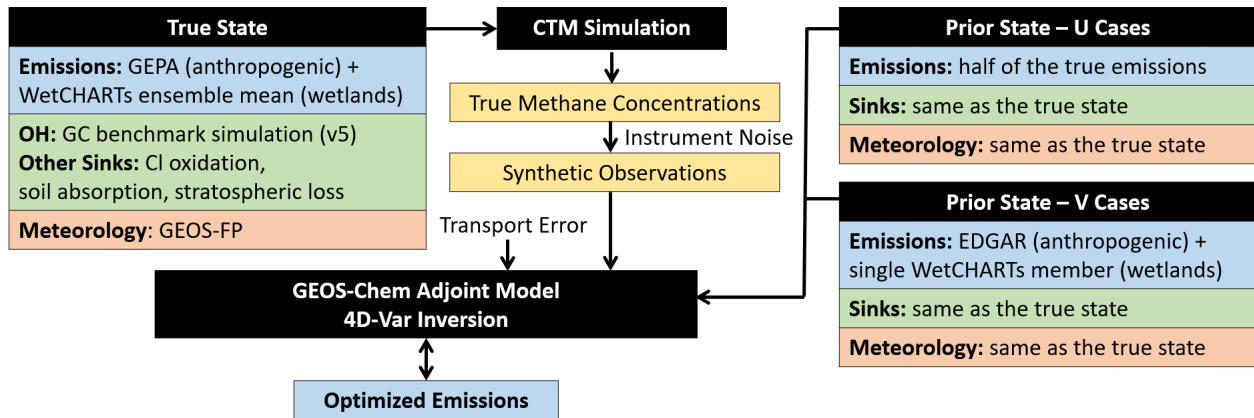
We use synthetic observations for August 2018 in our analyses and apply standard data filtering procedures based on the actual TROPOMI retrieval quality parameters for clouds, spectral fit, albedo, aerosols, and viewing geometry (Table S1 in the Supplement). Figure 2 shows the resulting data coverage for 1 d to 1 month intervals on the  $0.25^\circ \times 0.3125^\circ$  analysis grid. Over 247 000 observations are available for August 2018 over North America, covering 66 % of terrestrial grid cells, with the highest sampling density over the western US and northern Ontario. This level of data coverage is typical for TROPOMI: monthly overland coverage at  $0.25^\circ \times 0.3125^\circ$  ranges from 42 %–79 % between May 2018 and April 2019 when using the data selection criteria in Table S1 (Fig. S1 in the Supplement). For comparison, Fig. 2f shows August 2018 data coverage for the GOSAT satellite sensor, with overland sampling density just 0.4 % that of TROPOMI. Despite its significant sampling gaps, TROPOMI thus offers unprecedented new information for advancing scientific understanding of global methane sources and their spatiotemporal variability.

### 2.2 Chemical transport model and true state

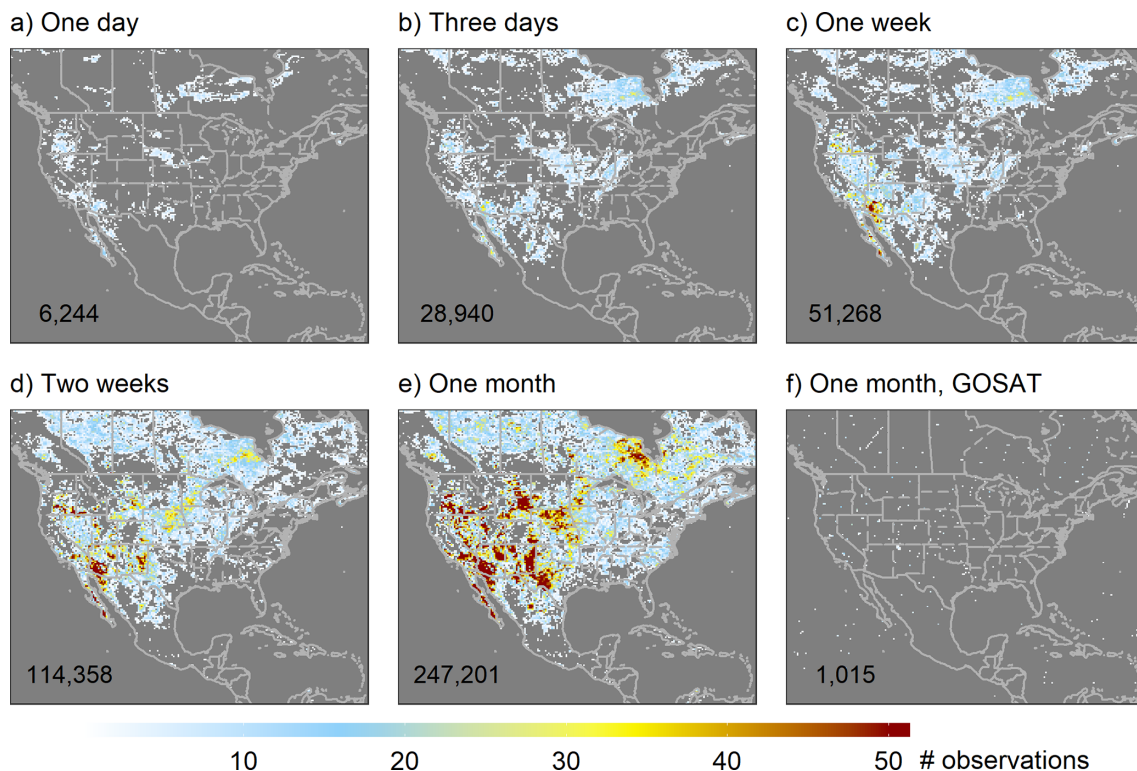
Our OSSE analyses employ the GEOS-Chem (v11-2) chemical transport model (CTM) and its adjoint (v35) to optimize methane emissions on a  $0.25^\circ \times 0.3125^\circ$  grid over North America ( $9.75\text{--}60^\circ \text{ N}$ ,  $60\text{--}130^\circ \text{ W}$ ). Simulations use GEOS-FP meteorological fields and 5 and 10 min time steps for transport and emissions, respectively. Three-hourly dynamic boundary conditions are from simultaneous global simulations at  $2^\circ \times 2.5^\circ$ . Initial conditions are based on a global 25-year spinup for 2016 at  $2^\circ \times 2.5^\circ$ , followed by a 2-week regional spinup over the nested domain at  $0.25^\circ \times 0.3125^\circ$ . As described next, inversions are performed for scenarios considering instrument error only and for scenarios considering both instrument error and model transport error. This permits comparison of these key observing system errors in terms of their impacts on solution accuracy.

Base-case analyses include instrument error only, with the same transport scheme used to drive the adjoint model and to generate the true-state methane fields. Specifically, this relies on the transport implementation in v35 of the GEOS-Chem adjoint model, which includes fully instantaneous planetary boundary layer (PBL) mixing (Wu et al., 2007), a relaxed Arakawa–Schubert scheme (Moorthi and Suarez, 1992) for convection, and a multi-dimensional flux-form semi-Lagrangian (FFSL) treatment of advection (Lin and Rood, 1996). The simulation also employs a six-cell ( $0.25^\circ \times 0.3125^\circ$ ) buffer region at the boundary between the global and nested simulation domains. We add 0.6 % random error to the resulting methane column concentrations to represent instrument noise, and we apply the TROPOMI observation operator (averaging kernel and prior methane profiles) to the model output sampled instantaneously at the time and location of each satellite retrieval. In this way, the applied cloud coverage and other data filters are consistent with the actual TROPOMI measurements.

Analyses that also incorporate model transport error rely on true-state tropospheric methane concentrations generated using v11-2 of the GEOS-Chem forward model. Transport here employs a non-local PBL mixing scheme (Lin and McElroy, 2010) and updated implementations of convection and advection (Zhang et al., 2021). The nested domain boundary uses a three-cell (rather than the six above) buffer region. Instrument error and the TROPOMI observation operator are then applied as before but to time-averaged (13:00–14:00 LT) rather than instantaneous tropospheric methane fields. The resulting TROPOMI methane columns have a mean root-mean-square error (RMSE) of 12 ppb relative to the base case, with similar error contributions from transport and from the use of alternative emissions (Fig. S2). For comparison, Locatelli et al. (2013) reported a mean inter-model standard deviation of  $> 15$  ppb for surface concentrations between 10 CTMs (with identical emissions but differing transport) across a global ensemble of observing stations.



**Figure 1.** Schematic illustrating the OSSE framework used here.



**Figure 2.** TROPOMI sampling density at  $0.25^\circ \times 0.3125^\circ$  resolution ( $\sim 25$  km) for 1 d to 1 month observing intervals after filtering for data quality and clouds. GOSAT coverage (with no data filter) is shown for comparison. The total number of observations is labeled in each panel.

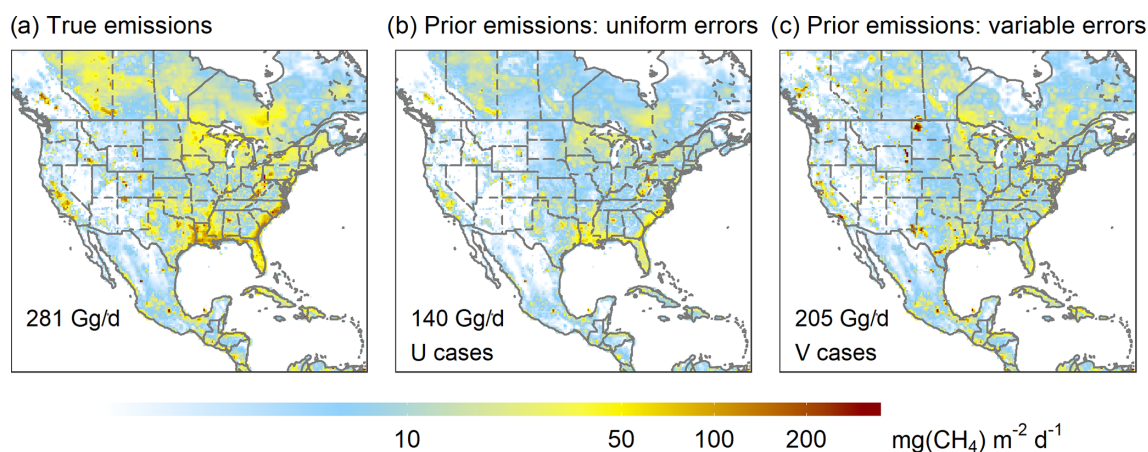
### 2.3 Methane sources and sinks

We use differing emission inventories to generate the true-state atmosphere and as prior for the inversions (Fig. 3). True-state anthropogenic fluxes are from the year-2012 gridded EPA inventory (Maasakkers et al., 2016) over the US and from year-2012 EDGAR v4.3.2 (2017) elsewhere, totalling  $124 \text{ Gg d}^{-1}$  over the North America simulation domain. Wetland emissions use the WetCHARTs ensemble mean ( $146 \text{ Gg d}^{-1}$  for the year 2017; Bloom et al., 2017),

while biomass burning emissions are from the Quick Fire Emissions Dataset (QFED,  $6 \text{ Gg d}^{-1}$  for the year 2017; Darmenov and da Silva, 2015). Geological seep and termite emissions follow Maasakkers et al. (2019) and Fung et al. (1991), respectively (together  $5 \text{ Gg d}^{-1}$ ).

Prior emissions include scenarios with spatially uniform and spatially variable errors (designated as U and V cases, respectively; see Table 1). In the U cases, we scale the true-state emissions uniformly by  $0.5\times$ . This is informative when the prior emissions have strong spatial fidelity with the truth and





**Figure 3.** True-state and prior emissions used in the OSSE analyses. The total emission for the North American domain is labeled in each panel.

is a common OSSE approach (e.g., Bousserez et al., 2016; Sheng et al., 2018a; Turner et al., 2018). However, when the spatial allocation of emissions is uncertain, as is frequently the case for methane, such analyses are likely to yield overly optimistic results. We therefore also include prior scenarios based on an alternate set of inventories: EDGAR v4.3.2 for anthropogenic emissions (yielding a domain-wide anthropogenic source of  $112 \text{ Gg d}^{-1}$ ), a single WetCHARTs ensemble member for wetlands (with  $\text{CH}_4 : \text{C } q_{10} = 1$ , GLOBCOVER wetland extent, and a resulting flux of  $80 \text{ Gg d}^{-1}$  for the year 2017), and QFED biomass burning emissions for a different year than in the true state ( $8 \text{ Gg d}^{-1}$  for the year 2018). The minor sources from geological seeps and termites are treated as before ( $5 \text{ Gg d}^{-1}$ ).

Figure 3 compares the true-state emissions with the above spatially perturbed prior. Across the domain, they differ by  $76 \text{ Gg d}^{-1}$  (27 % of the true state), with large spatial disparities yielding an overall  $R$  of 0.51. Major discrepancies are evident over oil and gas production regions (e.g., Bakken formation, Alberta oil sands), wetlands (e.g., Hudson Bay lowlands, US south coast), and agricultural areas (e.g., California Central Valley, Upper Midwest).

Methane sinks in the model include oxidation by the hydroxyl radical (OH, 90 % of the total simulated removal over the nested domain), computed using archived monthly fields from a full-chemistry simulation (v5-07-08). Stratospheric oxidation contributes a further 6 % and is computed based on NASA Global Modeling Initiative monthly loss frequencies (Murray et al., 2013). Other minor sinks include soil absorption (3 %; following Fung et al., 1991) and oxidation by chlorine (2 %; following Sherwen et al., 2016).

## 2.4 Optimization framework

We apply the GEOS-Chem adjoint model (Henze et al., 2007) to optimize the total methane flux ( $\mathbf{x}$ ) in each  $0.25^\circ \times 0.3125^\circ$  grid cell via iterative reduction in the fol-

lowing cost function:

$$J(\mathbf{x}) = (\mathbf{x} - \mathbf{x}_a)^T \mathbf{S}_a^{-1} (\mathbf{x} - \mathbf{x}_a) + \gamma (\mathbf{y} - F(\mathbf{x}))^T \mathbf{S}_{\text{obs}}^{-1} (\mathbf{y} - F(\mathbf{x})), \quad (1)$$

where  $F(\mathbf{x})$  are the model-predicted methane columns,  $\mathbf{y}$  are the synthetic TROPOMI observations,  $\mathbf{x}_a$  are the prior emissions, and  $\mathbf{S}_a$  and  $\mathbf{S}_{\text{obs}}$  are, respectively, the prior and observational error covariance matrices. We employ a regularization parameter ( $\gamma$ ) to avoid overfitting, defined based on 1-week sensitivity inversions with uniformly biased prior emissions (Fig. S3). We then scale  $\gamma$  for other time windows according to the number of observations ( $\gamma = 8.1, 1.6, 1.0, 0.5$ , and  $0.2$ , for 1 d, 3 d, 1 week, 2 weeks, and 1 month, respectively). We do not optimize boundary conditions.

By default, minimization of  $J(\mathbf{x})$  in the GEOS-Chem adjoint proceeds through derivation of grid-level SFs  $\mathbf{s}$  that are then applied as  $\mathbf{x} = \mathbf{s} \circ \mathbf{x}_a$ . Equation (1) can thus be equivalently expressed in terms of  $\mathbf{s}$  rather than  $\mathbf{x}$ , in which case  $\mathbf{S}_a$  describes relative rather than absolute errors. We first explore inversion performance in this framework, including the impacts of spatial emission errors and model transport errors. We subsequently evaluate four alternative inversions as candidates to address shortcomings of this SF approach; these are summarized in Table 1 and detailed in Sect. 4.

Prior error covariances for the GEPA anthropogenic emissions include magnitude and displacement components and are computed following Maasackers et al. (2016). Those for wetlands are estimated as the standard deviation of the WetCHARTs model ensemble (Bloom et al., 2017). Other emissions assume an error standard deviation of 50 %, consistent with previous work (Maasackers et al., 2019; Sheng et al., 2018b; Turner et al., 2015; Wecht et al., 2014a; Zhang et al., 2018). The above components, weighted by the corresponding flux amount, are added in quadrature to obtain the diagonal elements of  $\mathbf{S}_a$ . The resulting prior errors have a median value of 67 % for the uniform-bias inversions (which

**Table 1.** Inversion frameworks.

Emission bias		Inversion	Framework <sup>1</sup>	Rationale
Spatially uniform emission errors	U-SF	Base-case SF <sup>2</sup>	$\mathbf{x} = \mathbf{s} \circ \mathbf{x}_a$	Explore reliability of optimized fluxes when spatial distribution is well-known in prior
	V-SF	Base-case SF	$\mathbf{x} = \mathbf{s} \circ \mathbf{x}_a$	Explore influence of spatial emission errors on base-case SF inversion
Spatially varying emission errors	V-flat	Flat prior	$\mathbf{x} = x_{a\_ave} \mathbf{s}$	Identify constraints solely from TROPOMI without bottom-up knowledge
	V-AddBG	Background increment	$\mathbf{x} = \mathbf{s} \circ (0.5\mathbf{x}_a + 0.5x_{a\_ave})$	Identify missing sources
	V-OBSGuess	Observational guess	$\mathbf{x} = \mathbf{s} \circ (\mathbf{x}_a + \mathbf{x}_{ObsGuess})$	Resolve and optimize emission hotspots
	V-EH	Enhancement	$\mathbf{x} = x_{inc} \mathbf{s} + \mathbf{x}_a$	Identify missing sources
	Sensitivity inversions including transport error	* – * – T		Same as U and V cases above

<sup>1</sup> $\mathbf{x}$ : optimized emissions;  $\mathbf{s}$ : scale factors;  $\mathbf{x}_a$ : prior emissions;  $x_{a\_ave}$ : uniformly distributed prior emissions;  $\mathbf{x}_{ObsGuess}$ : observationally informed initial guess;  $x_{inc}$ : emission increment. <sup>2</sup>SF: scaling factor.

employ GEPA as anthropogenic prior) and 142 % for the others (which employ EDGAR v4.3.2). Finally, we employ an exponentially decaying 200 km correlation length scale to populate the off-diagonal elements of  $\mathbf{S}_a$ , in line with previous studies (200–500 km, Monteil et al., 2013; Wecht et al., 2014a; Yu et al., 2021a). Sensitivity tests varying this length scale yield broadly similar results: derived scale factors for 1-week V-SF inversions using 100, 200, and 400 km correlation lengths are spatially correlated to  $R = 0.82$ – $0.94$  and have  $< 1$  % domain-wide emission differences.

Observational error covariances are prescribed as the relative residual standard deviation of the column mismatch between the true-state synthetic observations and the prior simulations over a  $2^\circ \times 2^\circ$  moving window (Heald et al., 2004). We impose on the derived values a lower limit of 60 ppb<sup>2</sup>, corresponding to the 0.25 quantile of the overall error distribution. The resulting observing system errors average 9 ppb (range: 8–29 ppb) and mainly reflect instrument noise. The 9 ppb estimate is in line with and slightly smaller than observational error estimates for previous methane inversions using data from TROPOMI (e.g., 11 ppb; Zhang et al., 2020) and GOSAT (e.g., 13 ppb; Maasackers et al., 2019); it is therefore an appropriate representation for our OSSE analyses. Note that any systematic measurement errors (Lorente et al., 2021) are inherently not accounted for in our framework and would need separate correction.

We derive posterior errors and degrees of freedom for signal (DOFS) for each of the inversions using a gradient-based

randomization approach (Bousserez et al., 2015). The posterior error matrix  $\mathbf{S}_{opt}$  is given by

$$\begin{aligned} \mathbf{S}_{opt} &= \left( \mathbf{S}_a^{-1} + \gamma \mathbf{H}^T \mathbf{S}_{obs}^{-1} \mathbf{H} \right)^{-1} \\ &= \left( \mathbf{S}_a^{-1} + \gamma \overline{\nabla J(\mathbf{x}_a) \nabla J(\mathbf{x}_a)^T} \right)^{-1}, \end{aligned} \quad (2)$$

where  $\mathbf{H}$  is the forward model operator and  $\nabla J(\mathbf{x}_a)$  is the cost function gradient at  $\mathbf{x} = \mathbf{x}_a$ . The  $\overline{\nabla J(\mathbf{x}_a) \nabla J(\mathbf{x}_a)^T}$  term is computed from an ensemble of cost function gradients, each relying on synthetic data that have been perturbed with random noise based on the error characteristics of the original dataset; it thus reflects the combined effects of data coverage and the system error characteristics. The DOFS are then computed as the trace of  $\mathbf{I} - \mathbf{S}_{opt} \mathbf{S}_a^{-1}$ . Our computed DOFS converge at approximately 100 ensemble members.

We compute these information content metrics because they are commonly used for evaluating satellite instrument capabilities (Bousserez et al., 2016; Sheng et al., 2018a). However, posterior error reduction estimates can only match the emission improvements if the prior emissions are unbiased, which is not usually the case. For this reason, we compare the posterior error reduction  $\rho_{est}(i) = 1 - \mathbf{S}_{opt}(i, i) / \mathbf{S}_a(i, i)$  estimated as above against the actual grid-level emission improvement:

$$\rho_{true}(i) = 1 - (\hat{\mathbf{x}}(i) - \mathbf{x}_{true}(i))^2 / (\mathbf{x}_a(i) - \mathbf{x}_{true}(i))^2, \quad (3)$$

where  $\hat{x}(i)$  and  $x_{\text{true}}(i)$  are, respectively, the optimized and true emissions for grid cell  $i$ . For computational reasons we employ only the diagonal elements of  $S_a$  in the calculation of  $\rho_{\text{est}}$ ; an evaluation using a random subset of grid cells suggests that this approximation alters the results by  $< 25\%$ .

### 3 Role of prior errors in biasing inversions

#### 3.1 Inversions with spatially uniform prior biases

Our results show that in the absence of model transport errors, a 1-month standard SF inversion of TROPOMI observations can effectively minimize a uniform prior emission bias while maintaining spatial fidelity with the truth (U-SF inversion; Table 1). Figure 4a–b shows that for this scenario the prior bias of  $140 \text{ Gg d}^{-1}$  is reduced by 61% (to  $54 \text{ Gg d}^{-1}$ ), while spatial correlation with the true fluxes decreases only slightly from  $R = 1$  to  $R = 0.93$ . The U-SF inversion successfully improves the prior estimates for both small ( $< 50 \text{ mg m}^{-2} \text{ d}^{-1}$ ) and large ( $\geq 50 \text{ mg m}^{-2} \text{ d}^{-1}$ ) sources, in all cases maintaining  $R > 0.8$  with the true fluxes and decreasing model RMSE at the 25 km grid resolution (Fig. 5). Domain-wide flux estimates for these source categories are improved by 46% and 95%, respectively (Fig. 6). Partitioning of emissions between anthropogenic and natural sources (by scaling the derived total fluxes to the prior grid-level source fractions) is also accurate, with  $R > 0.9$  and decreased RMSE for every case except livestock (which had low error to begin with) and improved domain-wide flux accuracy (Figs. 5 and 6).

Despite this strong overall performance, we do see an influence from the prior emission distributions on the inversion results. The spatial correlation between the derived SFs and the prior emissions is  $R = 0.58$ , whereas the true solution (SF = 2 for all grid cells) would have no such correlation. This reflects a tendency for SF inversions to overcorrect large sources while undercorrecting small sources (along with the fact that the satellite data themselves are less sensitive to small sources). If the actual prior emission errors were random and normally distributed (i.e., no mean bias), the impacts of this tendency would manifest equally everywhere and would not lead to spatially coherent SF-prior correlations. Here, we employ a uniform prior emission bias, which (like most real scenarios) breaks the assumption of unbiased Gaussian emission errors. SF inversions are widely used despite this limitation, and we see here that the approach broadly succeeds under a uniform-bias scenario even with the tendency for large-source overcorrection.

The U-SF inversion has  $\text{DOFS} = 382$ , with derived posterior error reductions that reflect the TROPOMI spatial sampling density for this month (Fig. 7). However, this computed error reduction  $\rho_{\text{est}}$  (derived via gradient-based randomization) has no meaningful spatial correlation with the actual emission improvement  $\rho_{\text{true}}$  ( $R = 0.07$ ). This reflects

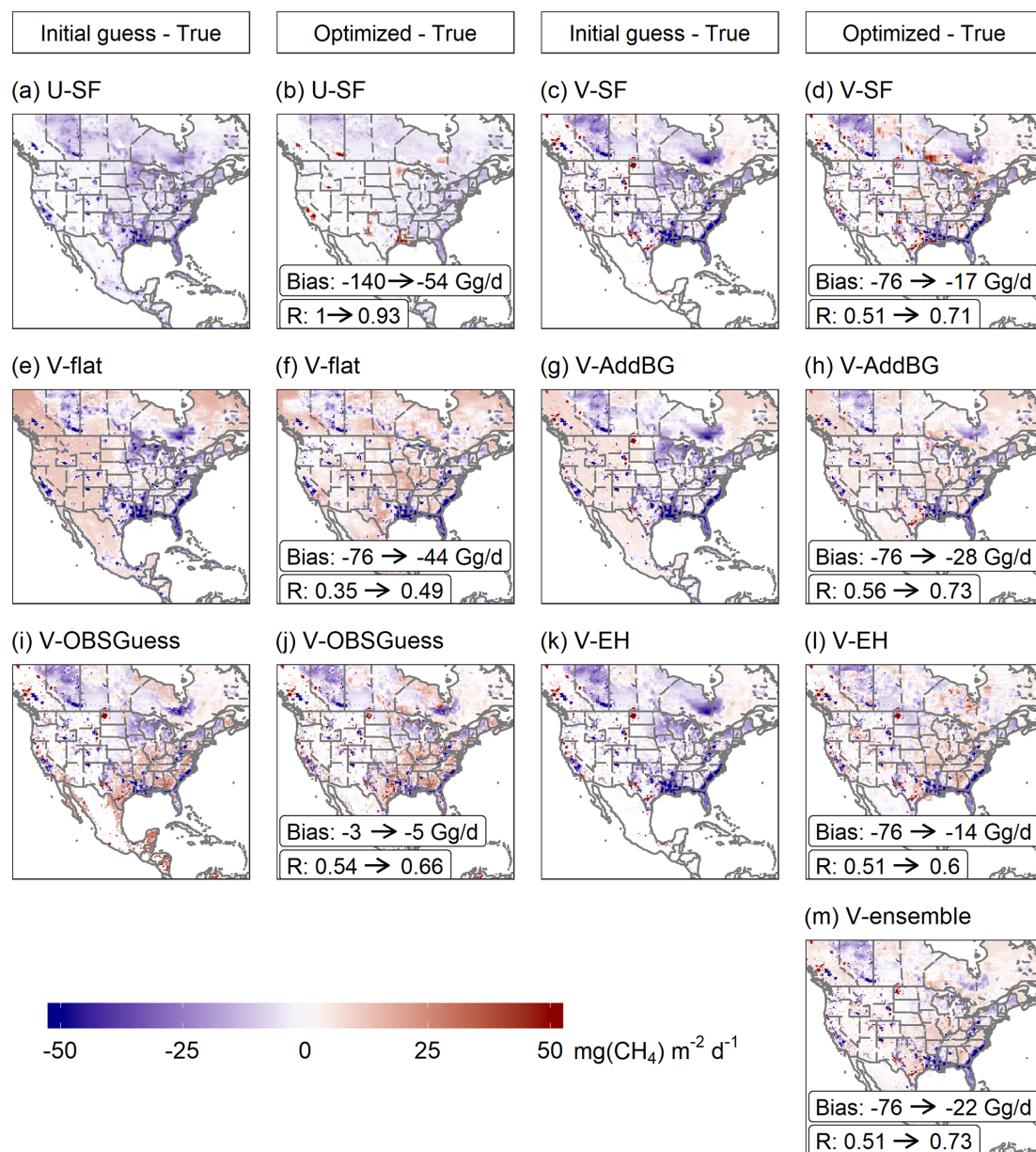
the fact that the posterior error reductions and DOFS contain no information on where the prior emissions are actually in error and can therefore be improved. For a scenario where the prior emissions had random and normally distributed disparities relative to the truth, areas with the largest computed posterior error reduction would also tend to have the greatest emission improvement – since those locations would have the strongest observational constraints. DOFS and error reduction analyses are thus useful for general observing system characterization but do not describe the spatial accuracy of posterior emissions or the actual emission improvements for realistic scenarios where the real prior errors are non-random.

The imposition of model transport errors does not strongly degrade the above performance. In this case (U-SF-T inversion; Table 1), the domain-wide optimized emission magnitude for North America is no less accurate than before (in fact slightly more so; Fig. 8b). The spatial distribution of the derived emissions, while less precise than in the case with perfect model transport, maintains high spatial fidelity with the truth ( $R > 0.8$ ; Fig. 8b). As we will see later, the same is not true when spatially varying prior errors are present.

#### 3.2 Inversions with spatially varying prior biases

When spatially varying biases are present in the prior emissions but transport errors are absent, the standard SF approach (V-SF inversion; Table 1) can still successfully minimize the domain-wide flux bias (78% reduction, from 76 to  $17 \text{ Gg d}^{-1}$ ) and has moderate success in recovering the true spatial distribution of emissions ( $R$  increases from 0.51 to 0.71). However, unlike the strong performance seen across all sources in the uniform-bias case, Figs. 5 and 6 show that the V-SF inversion reduces the domain-wide emission biases for both small and large sources but fails to improve the spatial allocation of large sources (RMSE and  $R$  improve by just 0.2% and 0.02, respectively). Comparing Fig. 4c–d and 5, we see that the V-SF inversion successfully corrects erroneous hotspots that are overestimated in the prior (e.g., Bakken and Permian shales) but lacks the ability to identify missing sources (e.g., wetland emissions in Alberta, the Hudson Bay lowlands, and the US south coast). Thus, given the presence of spatially varying prior biases, the SF tendency to overcorrect large sources and undercorrect small sources discussed above now manifests in systematic ways that bias the derived fluxes for particular regions and sectors.

The V-SF inversion has  $\text{DOFS} = 1281$ , higher than the U-SF case due to the larger prior error estimates for EDGAR versus GEPA. However, as with the U-SF inversion, the estimated posterior error reductions  $\rho_{\text{est}}$  and the actual emission improvements  $\rho_{\text{true}}$  have no significant spatial correlation at the 95% confidence level – for the reasons discussed earlier. As an example, the computed posterior error reductions shown in Fig. 7b show large uncertainty decreases over the Hudson Bay lowlands (with high observational density), but

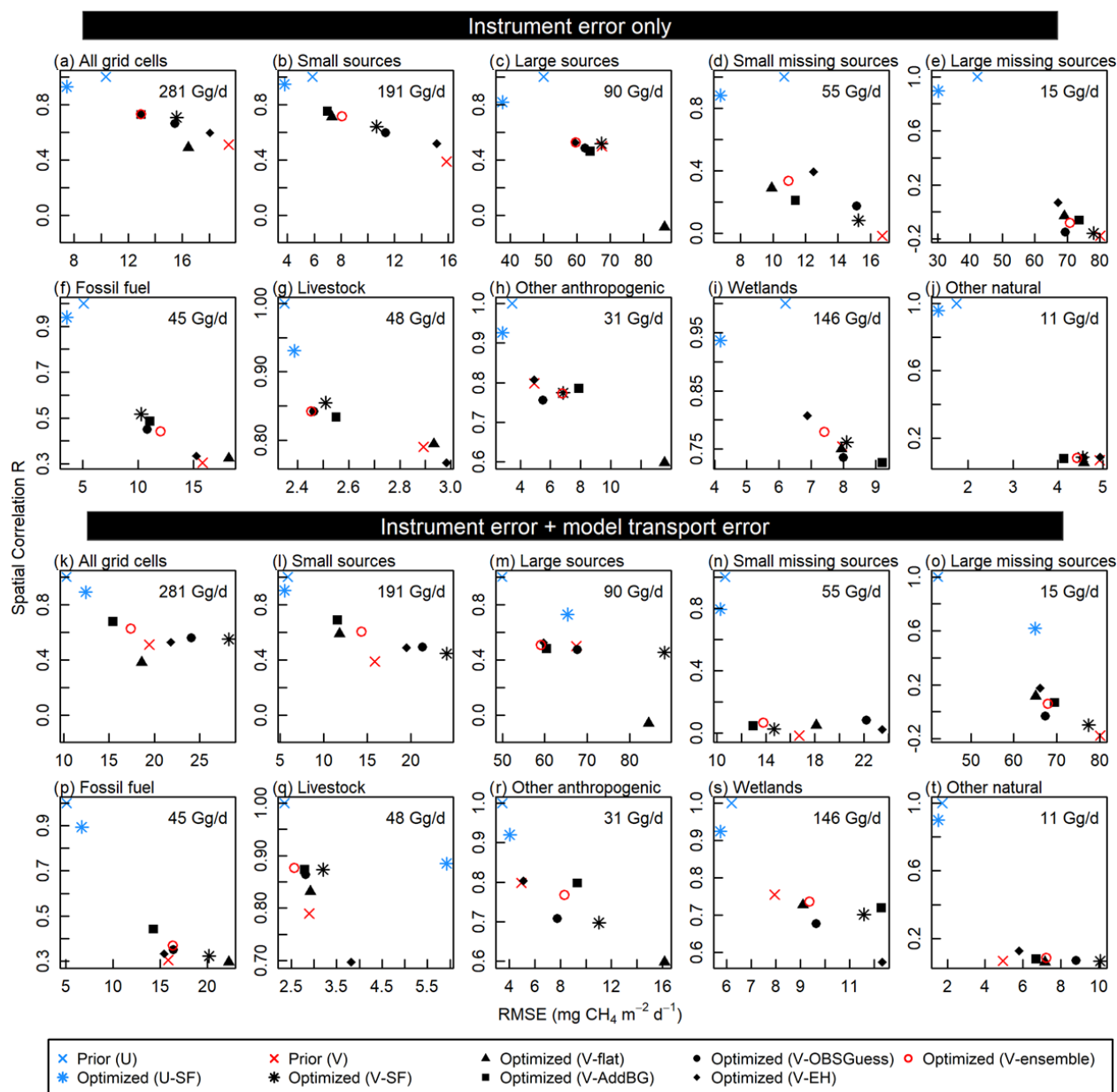


**Figure 4.** Differences between the initial-guess emissions and the true state and between the optimized emissions and the true state, for each inversion framework. Labels inset indicate the change in domain-wide bias and spatial correlation with respect to the true fluxes achieved through each optimization. More details are provided in Table 1 (inversion specifications) and Fig. S4 (initial-guess and optimized emissions). Inversions shown here do not include model transport errors; Fig. S7 shows results when such errors are included.

the derived fluxes over this region do not in fact improve towards the truth.

Combining model transport errors with the spatially varying prior emission errors substantially worsens the SF inversion performance. We then obtain an overcorrection of the domain-wide flux and a resulting positive bias (23 %) that is almost as large as the prior negative bias, with the optimization also failing to meaningfully improve the spatial accuracy of the prior emissions (Fig. 8b). As a result, the inversion has little ability to resolve sectoral sources: with the exception of

livestock, none of the posterior sectoral fluxes improve over their prior estimates in terms of RMSE or correlation, and only wetland sources are improved in terms of domain-wide bias (Figs. 5 and 6). As discussed in Sect. 5, extending the duration of the analysis period does not significantly improve the situation (Fig. 8b). This finding aligns with a previous ensemble analysis of surface observations (Locatelli et al., 2013), in which the optimized fluxes varied by 23 %–48 % regionally and up to 150 % at the grid level ( $2.5^\circ \times 3.75^\circ$ ) depending on model transport scheme. These disparities clearly



**Figure 5.** Inversion performance by source size and sector in terms of root-mean-square error (RMSE) and spatial correlation coefficient ( $R$ ) relative to the true fluxes. Results from base-case inversions (instrument error only) are shown for (a) all grid cells, (b) small sources ( $< 50 \text{ mg m}^{-2} \text{ d}^{-1}$ ), (c) large sources ( $\geq 50 \text{ mg m}^{-2} \text{ d}^{-1}$ ), (d) small missing sources (prior  $< 10 \text{ mg m}^{-2} \text{ d}^{-1}$ ; truth  $\in [10, 50] \text{ mg m}^{-2} \text{ d}^{-1}$ ), (e) large missing sources (prior  $< 10 \text{ mg m}^{-2} \text{ d}^{-1}$ ; truth  $\geq 50 \text{ mg m}^{-2} \text{ d}^{-1}$ ), (f) fossil fuel, (g) livestock, (h) other anthropogenic, (i) wetland, and (j) other natural emissions. Panels (k)–(t) show results including model transport error. The total true fluxes are indicated in each panel.

point to model transport error as one of the primary reasons behind the large spread in top-down methane source attributions (Locatelli et al., 2013).

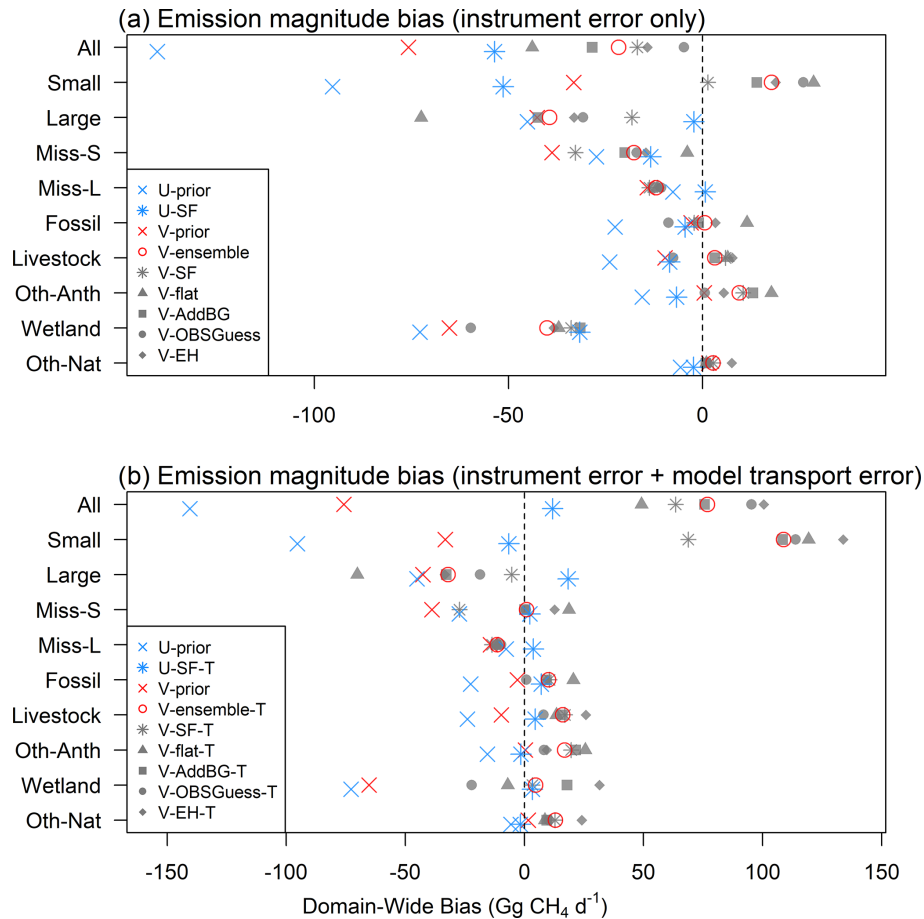
In summary, while TROPOMI is ground-breaking in providing high-precision, high-resolution methane observations daily and on a global basis, the combination of (i) spatial errors in prior emission estimates with (ii) model transport errors continues to limit inversion performance. Careful evaluation of potential missing sources in the prior inventories,

along with rigorous evaluation of model transport errors, is critical for robust inverse analyses.

#### 4 Alternative approaches to mitigate impacts of spatially varying prior errors

The impacts of prior spatial errors discussed above present a general challenge to inverse analyses. The fact that SF-based





**Figure 6.** Inversion performance in terms of domain-wide emission bias for (top to bottom): all grid cells, small sources ( $< 50 \text{ mg m}^{-2} \text{ d}^{-1}$ ), large sources ( $\geq 50 \text{ mg m}^{-2} \text{ d}^{-1}$ ), small missing sources (prior  $< 10 \text{ mg m}^{-2} \text{ d}^{-1}$ ; truth  $\in [10, 50) \text{ mg m}^{-2} \text{ d}^{-1}$ ), large missing sources (prior  $< 10 \text{ mg m}^{-2} \text{ d}^{-1}$ ; truth  $\geq 50 \text{ mg m}^{-2} \text{ d}^{-1}$ ), and fossil fuel, livestock, other anthropogenic, wetland, and other natural emissions. Panel (a) shows results for synthetic observations subject to instrument error only, while panel (b) shows results that also include model transport errors.

emission adjustments of a given magnitude incur less penalty over high-emission grid cells can be problematic for sources with poorly known spatial distributions, such as wetlands. For methane, with its long atmospheric lifetime, such issues are compounded by model transport biases, since the inversion cost function is then heavily weighted by downwind observations with their accumulating errors.

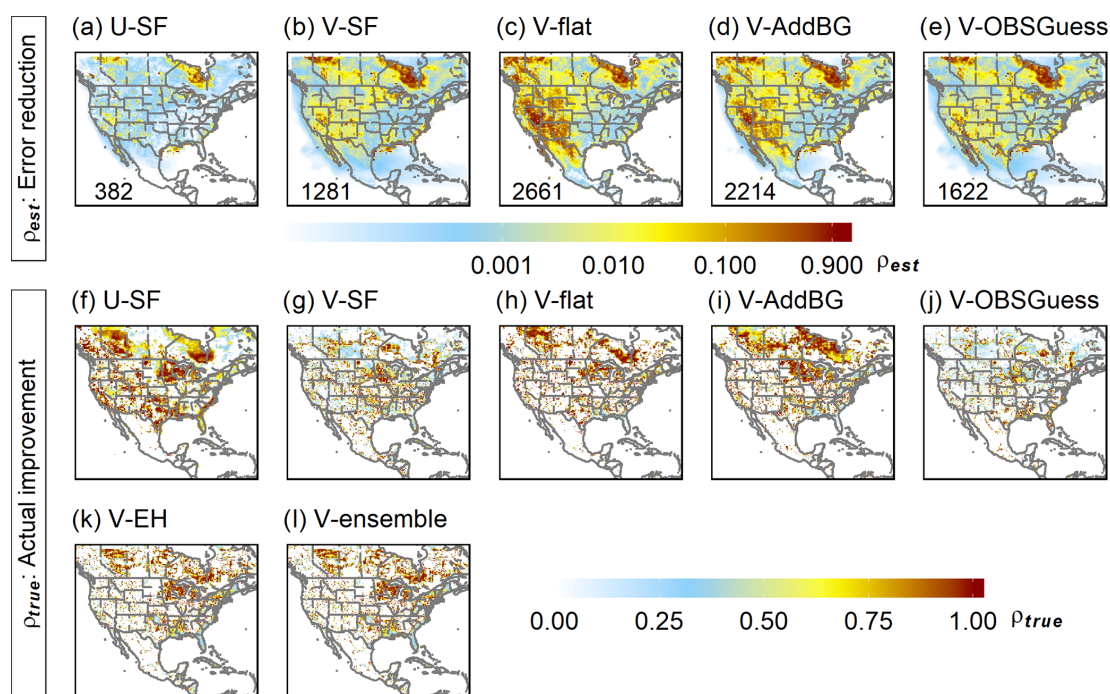
We examine here four alternative inversion strategies in terms of their capacity for addressing these limitations. As summarized in Table 1 and described below, these include three SF inversions in which the initial guesses are modified from the standard prior (V-flat, V-AddBG, and V-OBSGuess) and a fourth inversion that optimizes absolute emission enhancements (V-EH) rather than SFs. As before, we evaluate inversion performance in each case based on 1 month of synthetic TROPOMI observations both in the absence and presence of model transport errors.

#### 4.1 Flat-prior inversion (V-flat): good spatial performance for small sources but poor constraints on large sources

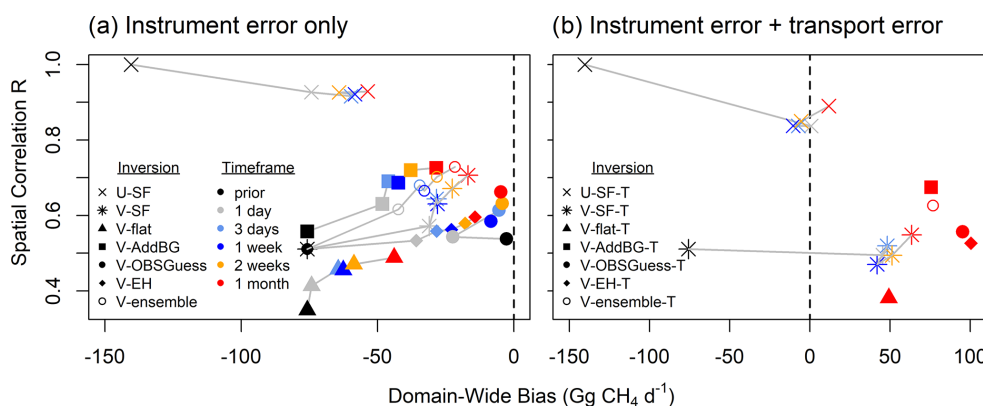
Our first alternative inversion (V-flat; Table 1) employs initial-guess emissions that are distributed uniformly among model land grid cells, with the aim of resolving spatial flux patterns entirely from the TROPOMI observations while also addressing the inability of the SF inversion to recover missing sources. The initial domain-wide flux magnitude is consistent with that in the standard prior, and the inversion penalty term is computed with respect to the revised initial guess.

Figure 4e–f shows that in the absence of transport error the V-flat inversion reduces the total prior emission bias by 42 % (from  $-76$  to  $-44 \text{ Gg d}^{-1}$ , the weakest performance among all inversions) and recovers a significant portion of the true spatial variance ( $R = 0.49$ ; however, this is still lower than for the original prior emissions). The optimization captures





**Figure 7.** Posterior error reduction derived via gradient-based randomization (top row; degrees of freedom for signal are labeled inset;  $\rho_{\text{est}}(i) = 1 - \mathbf{S}_{\text{opt}}(i, i) / \mathbf{S}_{\text{a}}(i, i)$ ) and actual grid-level emission improvement (bottom rows;  $\rho_{\text{true}}(i) = 1 - (\hat{\mathbf{x}}(i) - \mathbf{x}_{\text{true}}(i))^2 / (\mathbf{x}_{\text{a}}(i) - \mathbf{x}_{\text{true}}(i))^2$ ) for each inversion framework. Table 1 provides the inversion details for each case.



**Figure 8.** OSSE performance for timescales ranging from 1 d to 1 month (colors) as a function of inversion framework (symbols). Table 1 provides the inversion details for each case. Panel (a) shows results for synthetic observations subject to instrument error only, while panel (b) shows results that also include model transport errors (panel b shows only the 1 month results for the alternative formalisms).

some broad patterns in the true fluxes, with higher emissions in the eastern US and over Canada (Fig. S4), but does not resolve key finer-scale features. In particular, the optimized fluxes yield no improvement for large sources, either in terms of spatial distribution or emission magnitude (Figs. 5 and 6). The inversion performs well at recovering the spatial distribution of small sources (RMSE decreases by 54%;  $R$  increases from 0.39 to 0.71), and it captures their combined source magnitude to within 15%.

When transport error is included (V-flat-T, Table 1), the magnitude of the domain-wide emission bias increases from  $-44$  to  $+49 \text{ Gg d}^{-1}$ . The V-flat-T inversion is still able to improve the RMSE (by 25%) for small sources but dramatically overestimates their domain-wide magnitude (by 63%) to compensate for a 78% underestimate of large sources (Fig. 6). Given the V-flat inability to constrain large sources and poor overall spatial correlation with the truth, we move on to examine other inversion approaches below.

#### 4.2 Background-increment inversion (V-AddBG): best spatial performance

Our second alternative framework (V-AddBG; Table 1) occupies a middle ground between the V-SF inversion and the V-flat inversion. Here we construct an initial guess field as the mean of the standard prior and the flat initial guess used above, with the aim of incorporating prior knowledge while also enabling the SF inversion to recover missing sources.

Figure 4g–h shows that in the absence of model transport error, the V-AddBG inversion successfully reduces the regional mean bias from the prior value of  $-76$  to  $-28$  Gg d<sup>-1</sup>. While this overall bias correction is not as strong as the standard V-SF approach (63 % vs. 78 %), the V-AddBG inversion yields higher spatial correlation with the truth across the domain ( $R = 0.73$  vs. 0.71). Sectoral performance combines the strengths of the V-SF and V-flat inversions. Like V-flat, V-AddBG has strong spatial performance for small sources (reducing the RMSE by 56 % and increasing  $R$  from 0.39 to 0.75; Fig. 5) while mitigating the V-flat tendency to overestimate small sources and underestimate large sources. Like V-SF, V-AddBG can effectively correct false hotspots (e.g., Bakken and Permian shales) while being better able to resolve missing sources (e.g., Hudson Bay lowlands). Some missing sources (e.g., US Gulf Coast) are still not resolved due to limited observational coverage.

Transport errors (V-AddBG-T inversion; Table 1) increase the domain-wide bias in the derived fluxes from  $-28$  to  $+76$  Gg d<sup>-1</sup>. However, this framework still provides the best spatial accuracy among all approaches examined, with  $R = 0.67$  relative to the true fluxes even with transport error. It has better spatial performance than V-SF-T for every source category except wetlands, where the two are comparable (Fig. 5). For several emission categories where the standard V-SF-T solution is spatially inferior to the prior (small sources, large sources, fossil fuel, livestock), V-AddBG-T delivers meaningful improvements – improving the grid-level RMSE by up to 27 % and the spatial correlation to the truth by up to 76 % (Fig. 5). Overall, we find that V-AddBG provides the best spatial fidelity across all inverse approaches, but the domain-wide bias improvements are in general not as strong as the standard V-SF approach.

#### 4.3 Observational-guess inversion (V-OBSGuess): exploiting long-term observations to identify missing sources

Our third alternative framework (V-OBSGuess; Table 1) exploits the TROPOMI observations themselves to derive initial-guess emissions that enable SF-based recovery of spatially heterogeneous and missing sources. Specifically, we use 1 year of synthetic TROPOMI data (generated as above for the true-state atmosphere and averaged to the model resolution; Sun et al., 2018) to identify locations  $i$  with high methane column  $\Omega_i$  but low prior emissions  $x_i$  based on the

following index (see Fig. S5):

$$\beta_i = (x_{\max} - x_i) / x_{\max} \times \Omega_i / \Omega_{\max}. \quad (4)$$

For locations exceeding the 0.8 quantile for this index we linearly scale  $\beta_i$  to a corresponding prior emission increment. The scaling relationship is derived from scaled-emission sensitivity simulations with the resulting increments capped at 50 mg m<sup>-2</sup> d<sup>-1</sup>. The role of atmospheric transport means that this conversion is merely an approximation, but it is suitable for our purposes as input for a source inversion – and in particular, addresses the failure of standard SF inversions for scenarios with a near-zero prior where a source actually exists. The initial guess flux field derived in this way reduces the regional emission bias in the standard prior by 97 % (from  $-76$  to  $-3$  Gg d<sup>-1</sup>) and slightly improves its spatial fidelity to the truth (from  $R = 0.51$  to 0.54), thus providing an improved starting point for the 4D-Var optimization. This inversion thus optimizes emissions in two steps, using the synthetic observations to first spatially identify missing sources then to optimize grid-level emission magnitudes.

Figure 4i–j shows that in the absence of transport error, the V-OBSGuess inversion maintains the low domain-wide bias present in its initial guess; however, this does reflect some compensation between large-source underestimates and small-source overestimates (Fig. 6). The spatial accuracy of emissions is improved from  $R = 0.54$  to  $R = 0.66$ . Specific improvements are apparent for locations with erroneous hotspots in the prior (e.g., Bakken and Permian shales) as well as those with missing sources (e.g., Hudson Bay lowlands). Emissions are likewise improved over southern US coastal regions, where observational coverage is low for this month, thanks to the revised initial guess that employs a full year of data. V-OBSGuess yields moderate spatial performance for sector-specific emissions (Fig. 5), outperforming V-SF slightly for missing sources but otherwise not exhibiting a particular benefit for any specific emission category. Overall, when omitting transport error, the V-OBSGuess approach provides the lowest domain-wide bias of any inversion, with a comparable spatial performance to the standard V-SF approach.

However, this approach is highly sensitive to model transport error (V-OBSGuess-T; Table 1). In its presence, the domain-wide emission bias increases from  $-5$  to 95 Gg d<sup>-1</sup>, which is worse than the original prior. The spatial allocation of the derived emissions is not substantially improved over the revised initial guess ( $R = 0.56$  versus 0.54) or the original prior ( $R = 0.51$ ). Therefore, while V-OBSGuess can achieve a low regional-mean bias and strong spatial fidelity given accurate model transport, its flexibility in spatially correcting emissions also makes it more sensitive to meteorological errors than other approaches.

#### 4.4 Emission enhancement inversion (V-EH): recovering large missing sources

Finally, we optimize emission enhancements rather than SFs (V-EH; Table 1). In this case, the prior error covariances in Eq. (1) are computed with respect to absolute flux increments rather than with respect to the prior emissions; the inversion thus has increased flexibility to add emissions everywhere regardless of the local prior. We employ an increment of  $x_{\text{inc}} = 10 \text{ kg grid}^{-1} \text{ min}^{-1}$  and optimize emissions as  $\mathbf{x} = x_{\text{inc}}\mathbf{s} + \mathbf{x}_a$ . For context, approximately half of the domain-wide prior emissions are contributed by grid cells with emissions greater than  $x_{\text{inc}}$ . We further set a lower bound for the scaled increment (of  $-\mathbf{x}_a$ ) to avoid negative fluxes and prescribe zero fluxes for ocean grid cells outside of offshore oil/gas production fields.

Figure 4k–l shows that in the absence of model transport error, the V-EH inversion reduces the regional emission bias from  $-76$  to  $-14 \text{ Gg d}^{-1}$  (by 81 %) while improving the spatial correlation from  $R = 0.51$  to  $0.60$ . It succeeds at removing some of the false hotspots present in the prior (i.e., Bakken and Permian shales), but incorrectly smears those corrections spatially due to atmospheric transport. The V-EH inversion exhibits the best overall performance for missing sources (prior emissions  $< 10 \text{ mg m}^{-2} \text{ d}^{-1}$ ; Figs. 5 and 6): the RMSE and domain-wide bias are reduced by 25 % and 62 %, respectively, for small missing sources (with  $R$  increasing from 0 to 0.39 with respect to the truth) and by 16 % and 22 %, respectively, for large missing sources (but with no clear correlation improvement in this case).

Transport errors significantly degrade the optimized fluxes in this framework (V-EH-T; Table 1), increasing the domain-wide bias from  $-14$  to  $101 \text{ Gg d}^{-1}$  and reducing the spatial fidelity to  $R = 0.53$ . The latter is only a slight improvement over the prior ( $R = 0.51$ ) and lower than is obtained with the standard V-SF-T approach ( $R = 0.55$ ). V-EH-T still yields the best overall improvement for large missing sources, but in other cases the performance is mixed (Figs. 5 and 6).

#### 4.5 Summary and ensemble inversion performance

The inversion approaches explored above offer distinct advantages and disadvantages, which we summarize below.

1. In the absence of model errors (Fig. 5), V-flat and V-AddBG exhibit strong spatial performance for small sources ( $< 50 \text{ mg m}^{-2} \text{ d}^{-1}$ , representing 97 % of grids cells and 70 % of total emissions) but overestimate their emissions while underestimating those for large sources. For large sources ( $\geq 50 \text{ mg m}^{-2} \text{ d}^{-1}$ , 3 % of grid cells, 30 % of total emissions), all inversions except V-flat yield modest improvements (e.g., 0.2 %–12 % reduction in grid-level RMSE relative to the true fluxes). V-EH performs best at recovering missing sources (e.g., for wetlands), while the V-AddBG results generally have the highest spatial correlation with the truth. At the

sectoral level, the V-SF, V-AddBG, and V-OBSGuess inversions are all able to improve over the prior estimates for fossil fuel emissions (characterized by false hotspots in the prior) and livestock emissions (which have a spatially accurate prior). The alternative approaches allow more spatial flexibility in source allocation than the standard SF inversion, but the trade-off is a greater propensity to introduce spurious sources.

2. When subject to model errors, the V-SF-T, V-AddBG-T, and V-OBSGuess-T inversions all overcorrect large sources, while the V-flat-T and V-EH-T inversions create some spurious sources to compensate for transport biases. The V-AddBG-T inversion is the only framework able to reduce the grid-level emission RMSE despite transport errors, and it achieves the highest spatial  $R$  across all inversions (Fig. 5). However, it is unable to reduce the domain-wide emission bias given the transport errors imposed here. Conversely, the V-SF-T and V-flat-T inversions are able to improve the overall emission bias in the presence of transport errors, but they fail to improve their spatial accuracy. Short-lived species that are less influenced by transport error would likely yield better performance.
3. In all cases, the posterior error reductions  $\rho_{\text{est}}$  derived via Eq. (2) have no significant correlation with the actual emission improvements  $\rho_{\text{true}}$  (Fig. 7).

Together, the above inversions provide a range of possible emission solutions to fit the TROPOMI data. For optimizations using real data, analyzing the mean and spread across inversion frameworks provides a way to combine those various constraints and to diagnose robust aspects of the solution (e.g., Tarantola, 2006; Yu et al., 2021a). Here, in the absence of transport error, the ensemble mean solution reduces the domain-wide bias from  $-76$  to  $-22 \text{ Gg d}^{-1}$  (by 72 %; Fig. 4) with higher spatial accuracy ( $R$  increases from 0.51 to 0.73, RMSE is reduced by 33 %) than any individual inversion. In the presence of transport errors, the ensemble mean offers performance intermediate among the individual cases for both domain-wide emission bias (77 vs.  $-76 \text{ Gg d}^{-1}$  in the original prior) and overall spatial fidelity ( $R$  increases from 0.51 to 0.63, RMSE is reduced by 11 %).

Restricting the analysis to grid cells where the derived emission adjustments have a consistent sign across all inversions yields a larger improvement over the prior. In the instrument-error-only cases, the spatial fidelity across these grid cells (totalling  $127 \text{ Gg d}^{-1}$ , 45 % of total emissions, and 31 % of the grid cells) improves from  $R = 0.21$  to  $R = 0.53$  with a 46 % reduction in the grid-level RMSE, and total emission magnitude bias improves by 79 % (from  $-52$  to  $-11 \text{ Gg d}^{-1}$ ). In the presence of both instrument and model transport errors, the spatial fidelity of these consistent grid cells (totalling  $147 \text{ Gg d}^{-1}$ , 52 % of total emissions, and 40 % of the grid cells) improves from  $R = 0.25$  to  $R = 0.50$  with

a 27 % grid-level RMSE reduction, but total emission magnitude bias only improves by 2 % (from  $-61$  to  $60 \text{ Gg d}^{-1}$ ).

## 5 Influence of inversion timeframe on solution accuracy

The above analyses are all based on 1 month of synthetic observations and performed on the  $0.25^\circ \times 0.3125^\circ$  model grid. Below we extend the analysis to alternate timeframes to further evaluate the impact of data coverage on inversion performance.

We find that in the absence of both spatially varying prior emission errors and model transport errors (U cases; Table 1), a single day of TROPOMI measurements can reduce a domain-wide emission bias by 47 % (from  $-140$  to  $-74 \text{ Gg d}^{-1}$ , Fig. 8a) while maintaining high spatial fidelity with the truth ( $R > 0.9$ ). Extending the inversion timeframe to 3 d reduces the bias by an additional 10 % (to  $-60 \text{ Gg d}^{-1}$ ), but longer timeframes beyond this offer no additional benefit (our true emissions in the OSSE do not vary on a sub-monthly timescale). When model transport error is present but prior emissions are still spatially accurate, we again find similar performance across 1 d to 1 month timeframes, with strong bias reduction and spatial accuracy in each case (Fig. 8b, U-SF-T, Table 1). Therefore, given spatially reliable bottom-up inventories, TROPOMI can constrain methane emissions at high time resolution and resolve day-to-day temporal variability even in the presence of realistic model transport errors.

Spatial errors in the prior change this picture. In that case, when model transport error is absent (V cases; Table 1), both the emission bias and spatial fidelity continue to improve as the inversion timeframe increases from 1 d to 1 month (Fig. 8a). Given accurate transport, additional TROPOMI observations thus allow all the inversions to progressively correct prior spatial inaccuracies. However, the combination of transport errors with spatial emission errors in the prior strongly compromises inversion performance in all cases. Except for V-AddBG, we see at best marginal spatial improvements with increasing observational coverage (Fig. 8b). Furthermore, the domain-wide bias progressively worsens as the inversion timeframe increases, and after 1 month the bias in the derived fluxes has a comparable magnitude to that in the prior. While longer inversion windows benefit from increased data coverage, this comes at the cost of accumulating transport error as atmospheric enhancements are related to emissions farther and farther upwind. In such cases, multiple short inversions, rather than a single long inversion, may be preferable.

Given the combined effects of imperfect model transport, prior spatial errors, and limited observational coverage, one might expect better inversion accuracy when evaluated at coarser resolution rather than on the native  $0.25^\circ \times 0.3125^\circ$  model grid. When aggregating the solutions to  $4^\circ \times 5^\circ$  we

indeed obtain a higher correlation with the true fluxes for all cases with spatially variable prior errors (Fig. S6). However, this result mainly arises because the prior itself is more spatially accurate at this scale: the degree of improvement actually achieved through the inversion is no better than at fine resolution. Overall, neither the use of alternative timeframes nor of spatial aggregation changes our finding that inversion experiments can improve both the spatial distribution and magnitude of fluxes if either spatially variable prior errors or transport errors are present – but not when both errors are present.

## 6 Conclusions and implications

In this paper we examine three factors that limit the accuracy of top-down methane source estimates: (i) observational coverage, (ii) spatial inaccuracies in prior emission estimates, and (iii) model transport accuracy. The TROPOMI satellite sensor provides unprecedented, high-density and high-precision measurements of atmospheric methane columns over land, representing a major step forward in addressing the first of these constraints. We employ here a series of OSSE experiments to evaluate a range of inversion approaches in terms of their ability to spatially resolve methane emissions from high-coverage satellite sensors such as TROPOMI given the remaining limiting factors above.

The widely used SF-based inversion approach can be problematic for sources with poorly known spatial distributions, since adjusting grid cells with missing or near-zero sources in the prior may incur a prohibitive cost function penalty. The required emissions are then unfortunately allocated to higher-emission locations. We examine four alternate inversion strategies that aim to alleviate this issue. Three use a revised initial guess to allow the SF-based recovery of missing sources (V-flat: flat prior; V-AddBG: adds a background emission to the standard prior; V-OBSGuess: uses satellite data to pre-allocate missing sources), while the fourth optimizes emission enhancements (V-EH) rather than scale factors. The V-EH inversion performs best at resolving missing sources, whereas V-AddBG has the best performance in terms of the overall spatial fidelity of the solution – even in the presence of model transport errors. However, the standard V-SF approach yields better domain-wide bias reduction when model transport errors are present. The spread and mean across the ensemble solutions help in identifying robust aspects and uncertainties in the optimized flux patterns. For example, grid cells in which the emission corrections have a consistent sign across the ensemble members exhibit improved grid-level RMSE reduction (here, 27 % in the presence of transport error) compared to other grid cells ( $-8$  %, degraded performance).

We find that 4D-Var source optimization based on TROPOMI observations can provide robust constraints on monthly methane emissions at 25 km resolution, even when

provided with spatially incorrect prior emissions or in the presence of significant model transport error. However, performance is substantially degraded when both of these errors are present. Then, only one of the inversion frameworks is able to improve upon the prior spatial distribution of emissions (V-AddBG-T;  $R$  increases from 0.51 to 0.67 and RMSE reduces by 21 %), but it is unable to reduce the domain-wide emission bias. The two inversion frameworks that successfully reduce the prior bias in this scenario (V-SF-T and V-flat-T; 16 %–36 % bias reduction) are unable to improve the spatial distribution (grid-level RMSE). In many cases spurious emission adjustments are derived to compensate for the transport errors. Increasing observational coverage through longer-timeframe inversions does not resolve the situation, providing only a modest spatial improvement but with progressively worsening domain-wide bias due to accumulating transport errors.

We show through the OSSE analysis that the computed error reduction (approximated via gradient-based randomization) has no meaningful correlation with the actual emission improvements that are obtained in the inversions. This arises because, in general, the true prior emission disparities are not randomly distributed with zero mean (as is formally required in the best linear unbiased-estimate, or BLUE, 4D-Var problem) but rather have coherent spatial patterns associated with specific source types and regions. The same issue would also apply had the posterior errors instead been derived exactly via an analytical Bayesian solution. While often applied for observing system characterization, this approximate information content analysis should not be used to assess inversion accuracy or the spatial reliability of derived fluxes.

Findings here show that improving the spatial accuracy of bottom-up methane emission estimates is one key need for advancing top-down source assessments – for example through wetland extent surveys, a better assessment of the environmental drivers of fluxes, and precise facility-level information for livestock, fossil fuel, and industrial facilities. However, even with best efforts in these areas, the temporally sporadic nature of certain fluxes, combined with uncertainties in sectoral partitioning and in emission drivers, will inevitably lead to some bottom-up spatial biases. Such challenges provide the main motivation for pursuing top-down approaches in the first place.

Fundamental advancement will therefore require both the minimization of model transport errors and explicit representation of such errors in inverse analyses. Ongoing efforts to improve CTM representation of inter-hemispheric transport, convection, and boundary layer mixing offer promise for reducing the influence of model transport errors in future inverse analyses (Lin and McElroy, 2010; Patra et al., 2011; Saito et al., 2013). Including model error terms in the cost function for optimization, for example via weak-constraint inverse modeling, can alleviate the perfect-model dependence of standard (strong-constraint) inverse approaches and would improve inversion results for long-lived tracers such

as methane (Stanevich et al., 2020, 2021). Furthermore, the current version of the GEOS-Chem adjoint model assumes that the observational error covariance matrix is diagonal. However, model transport errors have clear spatial correlation structure (Fig. S2), and future work accounting for off-diagonal observational errors may thus improve performance. As such developments progress, current inverse analyses of space-borne methane measurements require careful evaluation of possible missing sources and of model transport errors, along with a thoughtful assessment of the potential for multiple viable solutions.

*Code and data availability.* TROPOMI data are publicly available at <http://www.tropomi.eu/data-products/level-2-products> (TROPOMI, 2021). The GEOS-Chem adjoint code is available at [http://adjoint.colorado.edu:8080/gcadj\\_std.git](http://adjoint.colorado.edu:8080/gcadj_std.git) (last access: 19 December 2021). The modified code used here is archived at <https://doi.org/10.13020/g5xc-nj81> (Yu et al., 2021b).

*Supplement.* The supplement related to this article is available online at: <https://doi.org/10.5194/gmd-14-7775-2021-supplement>.

*Author contributions.* XY performed the 4D-Var inversions. DBM and DKH supervised this study. All authors contributed to the interpretation of results. XY and DBM wrote the paper. All authors reviewed and edited the paper.

*Competing interests.* The contact author has declared that neither they nor their co-authors have any competing interests.

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