



Constraints on methane emissions from geostationary observations

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# Constraints on methane emissions in North America from future geostationary remote sensing measurements

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## Abstract

The success of future geostationary (GEO) satellite observation missions depends on our ability to design instruments that address their key scientific objectives. In this study, an Observation System Simulation Experiment (OSSE) is performed to quantify the constraints on methane (CH<sub>4</sub>) emissions in North America obtained from Short Wave Infrared (SWIR), Thermal Infrared (TIR) and multi-spectral measurements in geostationary orbit compared to existing SWIR low earth (LEO) measurements. A stochastic algorithm is used to compute the information content of a variational inversion at high spatial resolution (0.5° × 0.7°) using the GEOS-Chem chemical transport model and its adjoint. Both the SWIR LEO and TIR GEO configurations generally provide poor constraints on CH<sub>4</sub> emissions (error reduction < 30%), with the exception of a few hotspots (e.g., Los Angeles, Toronto urban areas and Appalachian Mountains) where the error reduction is greater than 50%. On weekly time scales and for a GEO orbit, the degree of freedom for signal (DOFs) of the inversion from multi-spectral observations (500) is a factor of two higher than that obtained from a SWIR instrument (255) due to the increase in measurement sensitivity to boundary layer concentrations in the multi-spectral case. On a monthly time scale and for a GEO orbit, a SWIR instrument would reduce error in emission estimates by more than 70% for hotspots of CH<sub>4</sub> sources (emissions > 4 × 10<sup>5</sup> kg day<sup>-1</sup> grid<sup>-1</sup>) at model grid scale, while a TIR instrument would provide a relative error reduction of 25–60% over those areas. While performing similarly for monthly inversions, a multi-spectral instrument would allow for more than 70% error reduction for these emissions for 7 or 3 day inversions. Sensitivity of the inversions to error in boundary conditions are found to be negligible. Moreover, estimates of the model resolution matrix over significant emitting regions (CH<sub>4</sub> emissions > 2 × 10<sup>5</sup> kg day<sup>-1</sup> grid<sup>-1</sup>) show that for all instrument configurations in GEO orbit the inversion is able to independently constrain CH<sub>4</sub> sources at spatial scales smaller than 200 km. These results highlight the importance of using observations sensitive

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to boundary layer concentrations (i.e., SWIR) to achieve significant improvements in constraining CH<sub>4</sub> sources compared to current LEO capabilities.

## 1 Introduction

Methane (CH<sub>4</sub>) plays a key role in both atmospheric chemistry composition and climate. With a radiative forcing relative to preindustrial times that is one third that of carbon dioxide, CH<sub>4</sub> is the second most important greenhouse gas (Myhre, 2013). Further, as a precursor to tropospheric ozone, CH<sub>4</sub> also impacts surface-level air quality (Fiore et al., 2002; West et al., 2006; West and Fiore, 2005), crops (e.g., Shindell et al., 2012) and contributes to ozone radiative forcing (e.g., Fiore et al., 2008). Considerable uncertainty remains in our understanding of CH<sub>4</sub> sources (e.g., Dlugokencky et al., 2011; Kirschke et al., 2013), which include emissions from coal, wetlands, livestock, landfills, biomass burning, geologic seepage, and leaks from the production and distribution of natural gas.

Although there is a growing interest in using CH<sub>4</sub> emission regulations as an efficient lever to simultaneously address current air quality and global warming challenges (e.g., West et al., 2012), the lack of confidence in the available CH<sub>4</sub> emission estimates remains a problematic limitation to design of efficient environmental policies. Indeed, recent studies showed discrepancies of up to a factor of two between bottom-up inventories and top-down inversions using atmospheric CH<sub>4</sub> concentration observations (Katzenstein et al., 2003; Kort et al., 2008; Xiao et al., 2008; Karion et al., 2013; Miller et al., 2013; Wecht et al., 2012; Caulton et al., 2014; Turner et al., 2015; Wecht et al., 2014a). Extrapolation of local emission characteristics to larger areas and/or the use of proxy data (e.g., energy consumption, emission ratios applied to co-emitted species) are the main sources of error in bottom-up methods (Zhen et al., 2015). On the other hand, top-down approaches using space-based measurements of CH<sub>4</sub> from Low Earth Orbit (LEO) platforms allow a global spatial coverage within one to six days but at the same local time. However, as CH<sub>4</sub> emissions can exhibit significant diurnal cycles, e.g.,

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consists of a 4D-Var inversion of CH<sub>4</sub> emissions using the GEOS-Chem chemical-transport model (CTM) over a 0.5° × 0.7° horizontal grid resolution covering North America. In practice, quantifying the information content of such a high-dimensional problem requires either Monte-Carlo simulations or, for linear models, a numerical approximation of the inverse Hessian matrix of the 4D-Var cost function (Tarantola, 2005). The computational cost of Monte-Carlo estimates can be prohibitive, since many perturbed inversions (typically about 50) are needed, each of them usually requiring numerous forward and adjoint model integrations. Therefore, computation of the information content in previous trace-gas Bayesian inversion studies has often relied on explicit calculations of the inverse Hessian matrix, by either considering a regional domain (e.g., Wecht et al., 2014a) or performing a prior dimension reduction of the control vector (e.g., Wecht et al., 2014b; Turner and Jacob, 2015). However, thus far dimension reduction methods for high-dimensional emission inversions have relied on suboptimal criteria.

In this study we use a gradient-based randomization algorithm to approximate the inverse Hessian of the cost function (Bousseres et al., 2015), which allows us to calculate the posterior errors as well as the model resolution matrix (or averaging kernel) of our CH<sub>4</sub> emission inversion at full grid-scale resolution. Such information is used to evaluate the impact of different instrumental designs (spatio-temporal sampling, vertical sensitivity of the measurements) on CH<sub>4</sub> emission constraints. In particular, the potential of CH<sub>4</sub> retrievals from existing Short Wave Infrared (SWIR) and Thermal Infrared (TIR) measurements as well as from a hypothetical multi-spectral instrument on geostationary orbit are examined. Section 2 describes the Observing System Simulation Experiment (OSSE) framework considered in this study, which comprises the 4D-Var method, the forward model, as well as the observations and prior information used. Section 3 presents the results of our experiments, where the information content of the inversion is analyzed in detail. A conclusion to this work is presented in the last section of the paper.



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This equivalence can be used to compute information content diagnostics prior to performing the inversion. In this study, following Bousseret et al. (2015), the diagonal elements of  $\mathbf{P}^a$  (error variances) are computed using a randomization estimate of  $\mathbf{H}^T \mathbf{R}^{-1} \mathbf{H}$ . Here an ensemble of 500 random gradients of the cost function are used, based on the convergence of the uniform norm ( $\|\cdot\|_\infty$ ) of the inverse Hessian approximation. Bousseret et al. (2015) showed that good approximation of both the error variances and the error correlations can be obtained using this approach. For the present study we further validated our method by comparing direct finite-difference estimates of selected diagonal elements of  $\mathbf{P}^a$  to their stochastic approximations and found a relative error smaller than 10 %.

The model resolution matrix (or averaging kernel  $\mathbf{A}$ ) is defined as the sensitivity of the analysis  $\mathbf{x}_a$  (optimized  $\text{CH}_4$  emissions) to the truth  $\mathbf{x}_t$  (true emissions):

$$\mathbf{A} \equiv \frac{\partial \mathbf{x}_a}{\partial \mathbf{x}_t}. \quad (4)$$

The model resolution matrix in Eq. (4) can be rewritten in matrix form:

$$\mathbf{A} = \mathbf{I} - \mathbf{P}^a \mathbf{B}^{-1}. \quad (5)$$

Since  $\mathbf{B}$  is diagonal in our experiments, Eq. (5) allows us to calculate any element of  $\mathbf{A}$  using:

$$\mathbf{A}_{i,j} = \delta_{ij} - \frac{\mathbf{P}_{i,j}^a}{\mathbf{B}_{j,j}}. \quad (6)$$

Finally, the degree of freedom for signal (DOFs) of the inversion is defined as the trace of  $\mathbf{A}$ , that is:  $\text{DOFs} = \sum_i \mathbf{A}_{i,i}$ .

## 2.2 Forward model and prior emissions

The forward model in Eq. (1) includes the GEOS-Chem chemistry-transport model, which relates the  $\text{CH}_4$  emissions to the 3-D concentration field of atmospheric  $\text{CH}_4$ ,

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and the satellite observation operator that transforms the CH<sub>4</sub> concentration profiles into their corresponding retrieved profile or columns. The GEOS-Chem simulation used in our experiment is described in (Wecht et al., 2014a; Turner et al., 2015). It consists of a nested simulation over North America at 0.5° × 0.7° horizontal resolution and 72 vertical levels, driven by offline meteorological data provided by GEOS-5 reanalysis from the NASA Global Modeling and Assimilation Office (GMAO). Boundary conditions for the nested domain are used every three hours from a global 4° × 5° GEOS-Chem simulation.

The prior methane emissions we use are from the EDGARv4.2 anthropogenic methane inventory (European Commission, 2011), the wetland model from Kaplan (2002) as implemented by Pickett-Heaps et al. (2011), the GFED3 biomass burning inventory (van der Werf et al., 2010), a termite inventory and soil absorption from Fung et al. (1991), and a biofuel inventory from Yevich and Logan (2003). Figure 1 shows the total average daily prior methane emissions for the entire North America nested domain. Strong hotspots of CH<sub>4</sub> sources clearly appear over the Canadian wetlands, the Appalachian Mountains (an extensive coal mining area) and densely urbanized areas (e.g., southern California and the East Coast). Following previous assessments of the range of the prior error (Wecht et al., 2014a; Turner et al., 2015), we assume a relative prior error of 40 % for our reference case.

### 2.3 Observations

We consider several instrument configurations for our study, which are associated with different vertical sensitivities. Constraints on CH<sub>4</sub> emissions are evaluated for the following CH<sub>4</sub> retrievals: the Greenhouse gases Observing SATellite (GOSAT) Proxy XCH<sub>4</sub> v3.2 data described by Parker et al. (2011) (available from <http://www.leos.le.ac.uk/GHG/data/>), which consists of CH<sub>4</sub> column mixing ratio  $X_{\text{CH}_4}$  obtained from Short Wave Infrared (SWIR) measurements near 1.6 μm; the Tropospheric Emission Spectrometer (TES) V005 Lite product (Worden et al., 2012) (<http://tes.jpl.nasa.gov/data/>), which consists of CH<sub>4</sub> vertical profile retrievals from Thermal Infra Red (TIR) measure-

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ments at 7.58–8.55  $\mu\text{m}$ ; and a hypothetical multi-spectral  $\text{CH}_4$  profile retrievals, which allows for significantly increased sensitivity of the retrieval to boundary layer concentrations. The multi-spectral retrieval are obtained from the 1.6 and 8  $\mu\text{m}$  bands as well as constraints developed for the TES algorithm. The Signal-to-noise ratio (SNR) is then adjusted such that the mapped profile gives a total column that is consistent with the observation error for a GOSAT retrieval. Since the DOFs for the TES retrievals is less than 2, we use an equivalent TES  $X_{\text{CH}_4}$  column instead of the retrieved  $\text{CH}_4$  profiles. Conversely, the multi-spectral retrievals have a DOFs between 2 and 3, and therefore the entire  $\text{CH}_4$  profiles were considered for this instrument.

Figure 2 shows the column averaging kernel for the GOSAT and TES  $X_{\text{CH}_4}$  retrievals as well as the averaging kernels at three different levels for the multi-spectral retrieval. The GOSAT retrieval sensitivity is nearly uniform throughout the troposphere, with averaging kernel values close to 1. The TES retrieval is mostly sensitive to  $\text{CH}_4$  concentrations in the upper troposphere, with a peak of the column averaging kernel around 300 hPa. The multi-spectral profile retrieval shows a distinct signal in the boundary layer, with weaker sensitivities above. Observational errors for methane columns ( $X_{\text{CH}_4}$ ) are uniformly set to 8 ppb for both GOSAT and TES. This value is consistent with GOSAT column errors reported in Parker et al. (2011). For the multi-spectral retrieval, a Singular Value Decomposition (SVD) of the covariance matrix of observation errors is performed for each profile in order to decorrelate errors between retrieval levels. As in Wecht et al. (2014b), model transport errors are assumed to be 16 ppb and are added in quadrature with measurement errors. As shown by Locatelli et al. (2013), taking into account transport errors is critical in order to mitigate uncertainties in the inversion, since neglecting them can lead to discrepancies in the posterior estimates of more than 150 % of the prior flux at model grid scale. Finally, contamination by clouds is taken into account for each grid cell by multiplying the corresponding GEOS-5 cloud fraction by the total number of observations within the grid cell and subtracting it from the total number of retrievals.







trations and aircraft-based observations from the HIAPER Pole-to-Pole Observations (HIPPO) experiment (Turner et al., 2015). These results show that a 2 % positive perturbation of the boundary conditions results in absolute posterior emission scaling factor responses smaller than 0.05 for all observational configurations. Moreover, the noisy characteristic of the scaling factor perturbations is evident for all inversions. Therefore, errors in boundary conditions of a few percent or less are found to have a negligible impact on our CH<sub>4</sub> source inversions.

### 3.3 Spatial resolution of the inversion

An objective measure of the spatial resolution of the inversion, i.e., the ability of the observational system to constrain grid-scale emissions independently from each other, is provided by the rows of the model resolution matrix (see Eq. 5). Figure 7 shows the model resolution matrix rows corresponding to five different locations, chosen to span a range of characteristics, in terms of emissions magnitude and error reduction. Table 1 summarizes the coordinates and CH<sub>4</sub> emissions corresponding to each location. The gain in spatial resolution as the sensitivity of the retrieval to boundary layer CH<sub>4</sub> concentrations increases is evident, especially for eastern Canadian wetlands and the Los Angeles area. For all instrument configurations, observations allow for constraints on CH<sub>4</sub> emissions at spatial resolutions between 80 km × 80 km and 160 km × 160 km. Note that over regions such as the wetlands in eastern Canada, the Appalachian Mountains and the Los Angeles area, using a multi-spectral instrument from geostationary orbit would allow complete constraints of CH<sub>4</sub> sources at grid-scale resolution (0.5° × 0.7°).

## 4 Conclusions

In this paper we evaluated top-down constraints on methane emissions in North America provided by potential geostationary observation missions (GEO-CAPE) and existing low-earth orbit remote sensing instruments (GOSAT). For the first time, a rigorous es-

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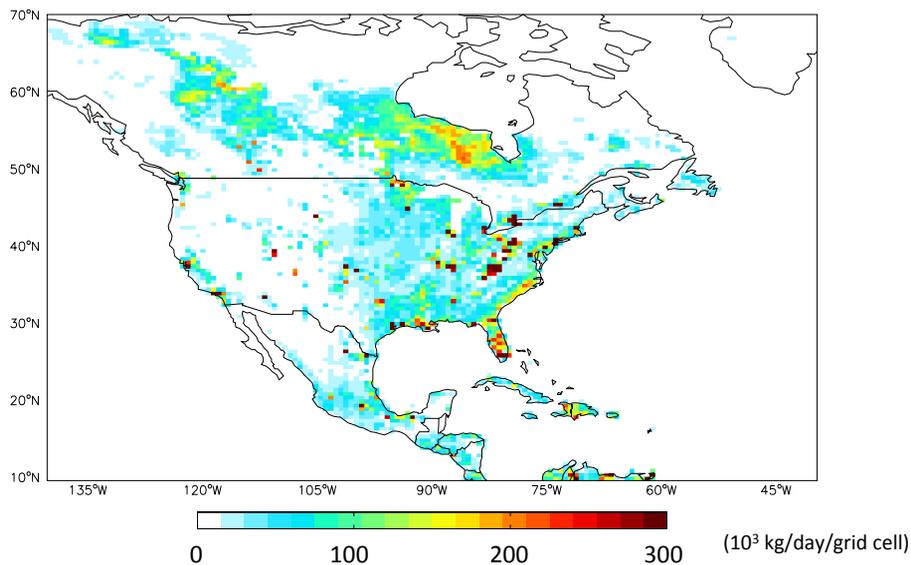
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**Figure 1.** Total average daily prior methane emissions for the nested North America domain ( $0.5^\circ \times 0.7^\circ$ ).

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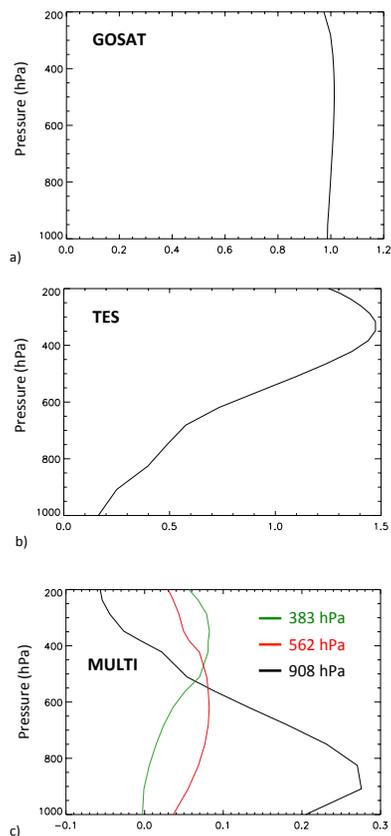
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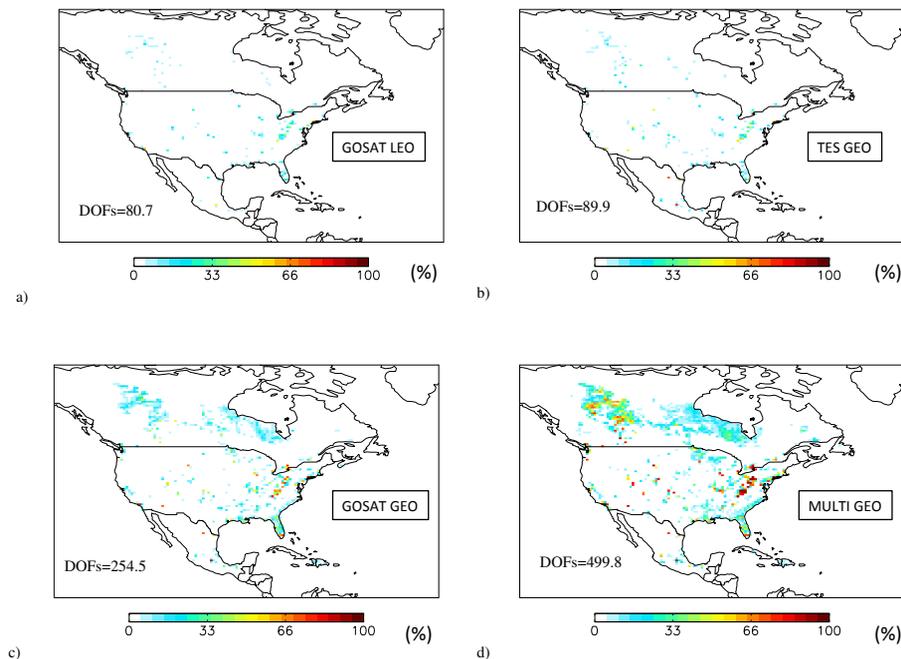
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**Figure 2.** Averaging kernels for the different instrument configurations: **(a)** GOSAT column averaging kernel; **(b)** TES column averaging kernel; **(c)** Multi-spectral averaging kernels at three pressure levels: 908, 562 and 383 hPa.

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**Figure 3.** Reduction (%) in methane emission standard errors for a 7 day inversion (1–8 July 2008) using: **(a)** GOSAT low-earth orbit observations (GOSAT\_LEO); **(b)** GEO-CAPE observations with a TES-like instrument (TES\_GEO); **(c)** GEO-CAPE observations with a GOSAT-like instrument (GOSAT\_GEO); **(d)** GEO-CAPE observations with a multi-spectral instrument (MULTI\_GEO). The degree of freedom for signal (DOFs) of each inversion is also indicated.

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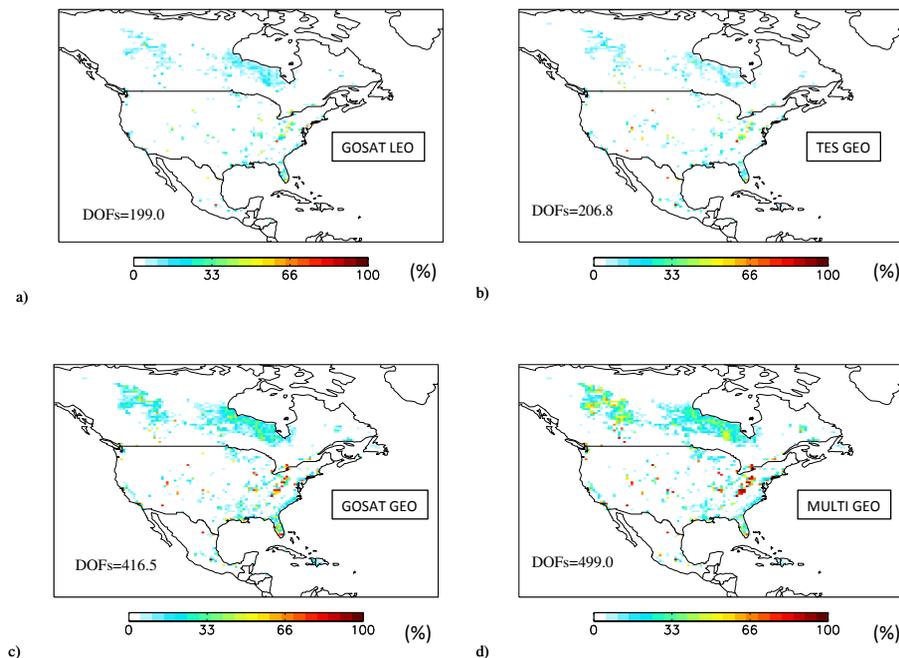
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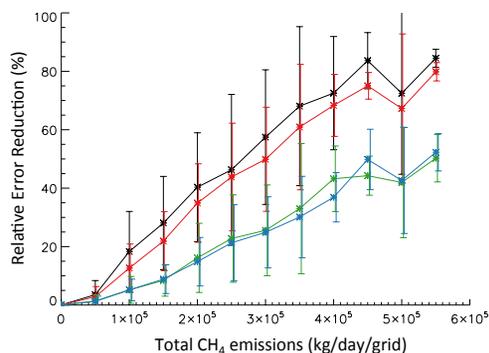
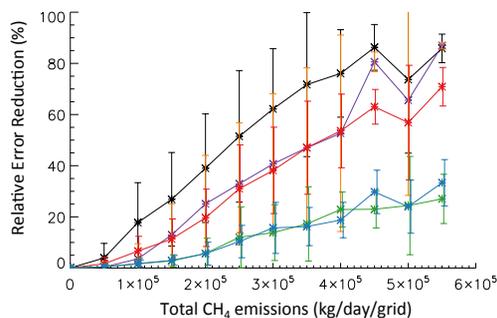


**Figure 4.** Reduction (%) in methane emission standard errors for a 30 day inversion (1–30 July 2008) using: **(a)** GOSAT low-earth orbit observations (GOSAT\_LEO); **(b)** GEO-CAPE observations with a TES-like instrument (TES\_GEO); **(c)** GEO-CAPE observations with a GOSAT-like instrument (GOSAT\_GEO); **(d)** GEO-CAPE observations with a multi-spectral instrument (MULTI\_GEO). The degree of freedom for signal (DOFs) of the inversion is also indicated.

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**Figure 5.** Reduction (%) in methane emission standard errors as a function of emission magnitude for a 7 day (1–8 July 2008) (top) and a 30 day (1–30 July 2008) (bottom) inversion. Blue: GOSAT low-earth orbit observations (GOSAT\_LEO); green: GEO-CAPE observations with a TES-like instrument (TES\_GEO); red: GEO-CAPE observations with a GOSAT-like instrument (GOSAT\_GEO); black: GEO-CAPE observations with a multi-spectral instrument (MULTI\_GEO). Results for a 3 day MULTI\_GEO inversion are also shown in purple (top). The vertical bars indicate the standard deviation of the relative error reduction within each bin.

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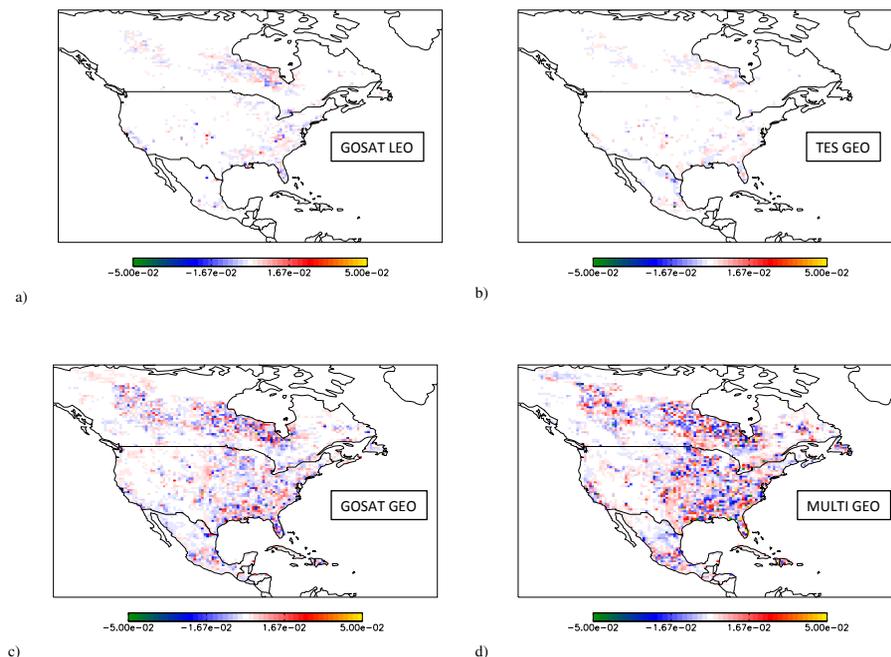
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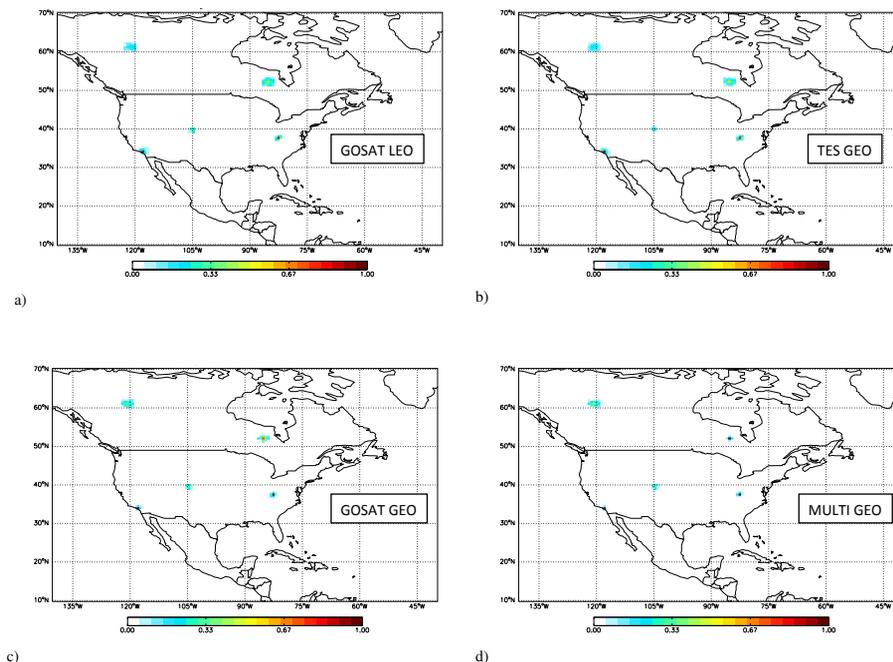


**Figure 6.** Sensitivity of optimized emission scaling factors (unitless) to a 2% perturbation of boundary condition methane concentrations for a 30 day inversion (1–30 July 2008), using: **(a)** GOSAT low-earth orbit observations (GOSAT\_LEO); **(b)** GEO-CAPE observations with a TES-like instrument (TES\_GEO); **(c)** GEO-CAPE observations with a GOSAT-like instrument (GOSAT\_GEO); **(d)** GEO-CAPE observations with a multi-spectral instrument (MULTI\_GEO).

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**Figure 7.** Rows of the model resolution matrix (unitless) for five locations for a 30 day inversion (1–30 July 2008), using: **(a)** GOSAT low-earth orbit observations (GOSAT\_LEO); **(b)** GEO-CAPE observations with a TES-like instrument (TES\_GEO); **(c)** GEO-CAPE observations with a GOSAT-like instrument (GOSAT\_GEO); **(d)** GEO-CAPE observations with a multi-spectral instrument (MULTI\_GEO). Coordinates of the five locations considered are reported in Table 1 and correspond to the center of each structure on the maps.

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