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Comparative analysis of low-Earth orbit (TROPOMI) and geostationary (GeoCARB, GEO-CAPE) satellite instruments for constraining methane emissions on fine regional scales: application to the Southeast US

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Abstract. We conduct observing system simulation experiments (OSSEs) to compare the ability of future satellite measurements of atmospheric methane columns (TROPOMI, GeoCARB, GEO-CAPE) for constraining methane emissions down to the 25 km scale through inverse analyses. The OSSE uses the GEOS-Chem chemical transport model ($0.25° \times 0.3125°$ grid resolution) in a 1-week simulation for the Southeast US with 216 emission elements to be optimized through inversion of synthetic satellite observations. Clouds contaminate 73-91% of the viewing scenes depending on pixel size. Comparison of GEOS-Chem to TCCON surface-based methane column observations indicates a model transport error standard deviation of 12 ppb, larger than the instrument errors when aggregated on the 25 km model grid scale, and with a temporal error correlation of 6 hours. We find that TROPOMI (7×7 km^2 pixels, daily return time) can provide a coarse regional optimization of methane emissions, and is highly sensitive to cloud cover. The geostationary instruments can do much better and are less sensitive to cloud cover, reflecting both their finer pixel resolution and more frequent observations. The information content from GeoCARB toward constraining methane emissions increases by 20-25% for each doubling of the GeoCARB measurement frequency. Temporal error correlation in the transport model moderates but does not cancel the benefit of more frequent measurements for geostationary instruments. We find that GeoCARB observing twice a day would provide 70% of the information from the nominal GEO-CAPE mission considered by NASA in response to the Decadal Survey of the US National Research Council.

1 Introduction

Methane is the second most important anthropogenic greenhouse gas after CO_2 (Myhre et al., 2013), and plays a key role in tropospheric and stratospheric chemistry (Thompson, 1992; West and Fiore, 2005; Solomon et al., 2010). The contributions from different source sectors and regions to the atmospheric methane budget remain highly uncertain (Kirschke et al., 2013; Saunois et al., 2016; Turner et al., 2017). Satellite observations of atmospheric methane columns in the shortwave infrared (SWIR) are a promising resource for constraining emissions through inverse analyses (Jacob et al., 2016; Houweling et al., 2017) but can be limited by instrument precision, sampling frequency, pixel resolution, cloud cover, and model transport error. Here we apply an Observing System Simulation Experiment (OSSE) for the Southeast US to compare the ability of new

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Atmospheric Measurement Techniques Discussions

satellite instruments to characterize methane emissions down to the 25-km scale, using as reference results from the recent SEAC^{4}RS aircraft campaign in the region (Sheng et al., 2018).

SWIR methane observations from space have so far been mainly from the SCIAMACHY instrument (2003-2013; Frankenberg et al., 2006) and the GOSAT instrument (2009-2016; Kuze et al., 2009, 2016). These data have proven useful for optimizing methane emissions on regional scales down to ∼100 km when averaged over several years (Bergamaschi et al., 2013; Fraser et al., 2013; Monteil et al., 2013; Wecht et al., 2014b; Turner et al., 2015; Alexe et al., 2015; Feng et al., 2017), but they are too sparse to constrain methane emissions on finer spatial or temporal scales. Our ability to observe methane from space should be considerably improved with the recent launch (October 2017) of the SWIR TROPOMI instrument, providing daily global coverage with 0.6% precision and 7×7 km^2 nadir resolution (Butz et al., 2012). The GeoCARB geostationary mission to be launched in the 2020s will observe methane columns over North and South America with 0.6% precision and 3×3 km^2 resolution (Polonsky et al., 2014; O'Brien et al., 2016). The observing frequency of GeoCARB is not finalized yet and could be 1-4 times per day. Other geostationary instruments still at the proposal stage offer improved combinations of pixel size, precision, and observing frequency, including GEO-CAPE (Fishman et al., 2012), GeoFTS (Xi et al., 2015), G3E (Butz et al., 2015), and CHRONOS (Edwards et al., 2018). GEO-CAPE has been pre-formulated by NASA as a recommended mission from the US National Research Council (2007) Decadal Survey on Earth Science and Applications from Space.

OSSEs are standard approaches to assess the utility of future satellite instruments to deliver on a specific objective, here the mapping of methane emissions. OSSEs at 50 km spatial resolution have been conducted to evaluate the potential of future satellite observations for quantifying methane emissions over California (Wecht et al., 2014a) and North America (Bousserez et al., 2016). Bousserez et al. (2016) in particular assessed the benefit of geostationary multi-spectral (SWIR + thermal infrared) measurements. Turner et al. (2018) conducted a kilometer-resolution OSSE to explore the potential of different satellite observing configurations to resolve the distribution of methane emissions on the scale of an oil/gas field.

Here we conduct a comparative analysis of TROPOMI, GeoCARB, and GEO-CAPE for constraining the spatial distribution of methane emissions at a fine regional scale (25 km), and we investigate more generally how the information content from different satellite observing configurations depends on pixel size, observing frequency, and cloud contamination. Of particular interest is to define observing frequency requirements for GeoCARB to resolve regional-scale methane sources. We focus on the Southeast US, which is a major source region for methane including large contributions from oil/gas production and wetlands (Maasakkers et al., 2016; Bloom et al., 2017). Sheng et al. (2018) previously used boundary layer methane observations from the NASA SEAC^{4}RS aircraft campaign (Toon et al., 2016) in August-September 2013 to optimize methane emissions over the Southeast US. This offers an opportunity to directly compare the observing power of satellite instruments to that from a dedicated aircraft campaign.

2 Observing system simulation experiments

Our OSSE framework is shown in Figure 1. We build on the previous work of Sheng et al. (2018), who conducted a Bayesian inverse analysis of the SEAC^{4}RS aircraft observations with the GEOS-Chem chemical transport model (CTM) at $0.25° \times$

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Atmospheric Measurement Techniques Discussions

$0.3125°$ resolution. They used the SEAC^{4}RS data together with prior estimates and error statistics from the gridded EPA inventory of Maasakkers et al. (2016) and the WetCHARTs extended ensemble wetland inventory of Bloom et al. (2017), to optimize the spatial distribution of methane emissions in the Southeast US for August-September 2013. We follow the same analytical inversion framework as Sheng et al. (2018) for our OSSE. We first simulate a methane column concentration field using the GEOS-Chem CTM with prior emission estimates (base simulation). We then sample this field following the specifications of the different satellite instruments (Table 1), accounting for instrument random noise and cloud contamination (discussed below). Randomness in the noise of synthetic observations is a standard OSSE assumption (e.g., Wecht et al., 2014a; Bousserez et al., 2016) and may overestimate the absolute DOFS of an instrument but does not affect a comparative analysis of different instruments.

The sampled synthetic observations define the observation vector $\mathbf{y}$ for the inversion. The sensitivity of these observations to the distribution of methane emissions over the domain (arranged as a state vector $\mathbf{x}$) is defined by the Jacobian matrix $\mathbf{K} = \partial\mathbf{y}/\partial\mathbf{x}$, where the ith column of $\mathbf{K}$ ($\partial\mathbf{y}/\partial x_i$) is constructed from GEOS-Chem by perturbing individual state vector elements x_i to compute the resulting perturbation $\Delta\mathbf{y}$ (relative to the base simulation). We then use this Jacobian matrix together with prior and observational error statistics (error covariance matrices $\mathbf{S_A}$ and $\mathbf{S_O}$) to quantify the information content of observations toward constraining emissions. The OSSE is conducted for the one-week period of August 8-14, 2013. All observations use a mean SWIR averaging kernel from GOSAT with uniform near-unit sensitivity in the troposphere (Worden et al., 2015).

The state vector $\mathbf{x}$ of emissions, representing the spatial distribution of emissions to be resolved by the inversion, is the same as in Sheng et al. (2018). It includes 216 Gaussian mixture model (GMM) elements with radial basis functions (Turner and Jacob, 2015), representing the extent of information on emissions that we may hope to achieve in 1 week of observations. The GMM provides $\sim$25 km resolution to resolve emission hotspots and coarser resolution in areas of weak emissions.

The analytical solution to the Bayesian inversion problem includes full characterization of the information content from the observations towards quantifying the state vector of emissions, as computed by the Degrees of Freedom For Signal (DOFS; Rodgers, 2000). Combining the Jacobian matrix $\mathbf{K}$ constructed from GEOS-Chem together with the prior error covariance matrix $\mathbf{S_A}$ and the observation error covariance matrix $\mathbf{S_O}$, we compute the averaging kernel matrix $\mathbf{A} = \partial\hat{\mathbf{x}}/\partial\mathbf{x}$ that represents the sensitivity of the optimization ($\hat{\mathbf{x}}$) to the true state ($\mathbf{x}$):

$$\mathbf{A} = \mathbf{S_A}\mathbf{K}^T(\mathbf{K}\mathbf{S_A}\mathbf{K}^T + \mathbf{S_O})^{-1}\,\mathbf{K}. \quad (1)$$

The DOFS is then the trace of the averaging kernel matrix:

$$\mathrm{DOFS} = \mathrm{tr}(\mathbf{A}). \quad (2)$$

The DOFS represents the number of pieces of information provided by the observing system for quantifying the state vector. DOFS = 216 would represent perfect constraints on our state vector. The SEAC^{4}RS aircraft inversion of Sheng et al. (2018) achieved DOFS = 10.

The prior error covariance matrix $\mathbf{S_A}$ for our problem is taken from the emission inventory error estimates of Maasakkers et al. (2016) and Bloom et al. (2017), as described by Sheng et al. (2018). The observational error covariance matrix $\mathbf{S_O}$ is

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Atmospheric Measurement Techniques Discussions

specific to the observing configuration, and includes contributions from model transport error in simulating the observations as well as instrument errors.

We estimate the model transport error variance by the residual error method (Heald et al., 2004) applied to the GEOS-Chem simulation with prior emissions of hourly observed Total Carbon Column Observing Network (TCCON) methane columns in Lamont, Oklahoma for August-September 2013 (Wunch et al., 2011; Wennberg et al., 2017). In that method, the mean bias in the model compared to the observations is attributed to error in the prior emissions (to be corrected in the inversion) and the residual characterizes the observation error including contributions from model transport error and instrument error. In our case, the TCCON measurements are highly precise (precision is $<$4 ppb), so that the residual characterizes the model transport error. The residual error distribution is shown in Figure 2, with an error standard deviation of 12 ppb. This is consistent with previous GEOS-Chem transport error estimates (Wecht et al., 2014a; Turner et al., 2015).

Temporal correlation in the model transport error may limit the benefit of high-frequency observations, because repeated observations of the same scene may produce the same model-observation differences. Here we estimate this error correlation from the autocorrelation vs. time lag of the difference between GEOS-Chem and TCCON observations. Results in Figure 2 show an e-folding error correlation time scale of 6 hours which we apply as off-diagonal elements in the observational error covariance matrices for the different satellite observing configurations. Also shown in Figure 2 is the persistence (e-folding) time scale for cloud cover, which affects the extent to which the temporal error correlation limits the information content of high-frequency observations; this will be discussed in the next Section.

The instrument error for individual observations is given by the precision values in Table 1, taken from the original references. The observations are averaged over $0.25^\circ \times 0.3125^\circ$ GEOS-Chem grid cells for the purpose of the inversion, and the instrument error standard deviation is decreased by the square root of the number of successful retrievals averaged over each grid cell for individual retrieval time.

Any cloud contamination within an observation pixel will cause an unsuccessful SWIR retrieval for methane (Butz et al., 2012). Remer et al. (2012) used high-resolution cloud data (0.5-1.0 km) over the US for different regions and seasons to infer probabilities for satellites to view clear-sky pixels. They focused on aerosol retrievals but the same statistics can be used for methane retrievals. The retrieval of methane has no tolerance for any cloud in the scene. For the Southeast US in summer with an average cloud fraction of 0.7, they find that cloud contamination would invalidate 91% of retrievals for TROPOMI (7×7 km^2 pixels), 73% for GeoCARB (3×3 km^2 pixels), and 79% for GEO-CAPE (4×4 km^2 pixels). The invalid retrieval rate of 91% for TROPOMI in the Southeast US is in the global mean range of 83% for GOSAT (proxy retrieval, Parker et al., 2011) and 97% expected for TROPOMI (full physics retrieval, Butz et al., 2012).

Here we first remove observations for cloudy regions over the inversion domain and period following the three-hour $0.25^\circ \times 0.3125^\circ$ fractional cloud cover information in the GEOS-FP meteorological data used to drive GEOS-Chem (Lucchesi, 2013), and then scale the removal rate to match the clear-sky probabilities as a function of satellite pixel resolution (Remer et al., 2012). Although the satellite data loss from cloud cover is severe, the $0.25^\circ \times 0.3125^\circ$ resolution of our inversion allows aggregation of data from a large number of observation pixels for comparison to the model. This does not help when there is solid cloud cover on the 25 km scale in the GEOS-FP data (as in the white areas for the GeoCARB pseudo-observations in Figure 1) but it

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Atmospheric Measurement Techniques Discussions

helps for fractional cloud cover. The median number of aggregated successful pixel retrievals for a given $0.25° \times 0.3125°$ grid cell at a given observation time is 3, 30, and 15 for TROPOMI, GeoCARB, and GEO-CAPE, respectively. Thus the median instrument error standard deviation on the $0.25° \times 0.3125°$ grid scale over our inversion domain is 6 ppb for TROPOMI, and 2-4 ppb for the geostationary instruments. This is smaller than the 12 ppb model transport error standard deviation (Figure 2), so that most of the observational error is contributed by model transport. This is an important result as it implies that the instrument precisions in Table 1 are sufficiently good at the 25 km scale that the inversion results are relatively insensitive to them, while at kilometer scales Turner et al. (2018) found much more pronounced benefit for finer instrument precision.

3 Results and discussion

The information content from different satellite observing configurations is diagnosed by the DOFs, as described in the Methods section, representing the number of pieces of information on emissions that can be retrieved by inversion of synthetic observations. Figure 3 shows a contour plot of the DOFS as a function of observing frequency and pixel resolution, assuming a fixed instrument precision of 0.6%. As discussed in the previous Section, results are relatively insensitive to instrument precision since most of the observational error is contributed by model transport. The DOFS increase as measurement frequency increases (more independent observations) and as pixel size decreases (more observations aggregated in a $0.25° \times 0.3125°$ grid cell). The benefit of increasing measurement frequency eventually weakens at high values because of temporal correlation in the GEOS-Chem model transport error. The benefit of increasing pixel resolution also weakens below 4 km because the inversion does not try to resolve emissions to resolution finer than $0.25° \times 0.3125°$. Even so, the maximum DOFS of 70 in Figure 3 could be achieved by a very high-resolution system (1 km pixel size and hourly observations) is much less than the ideal value of 216. A longer observation period or a lower model transport error would be needed to approach that ideal value.

DOFS for TROPOMI, GeoCARB (1-4 measurements per day) and GEO-CAPE are indicated on the contour map. The TROPOMI inversion has 18 DOFS, higher than the SEAC^{4}RS aircraft campaign (DOFS = 10; Sheng et al., 2018). The geostationary GeoCARB and GEO-CAPE observations achieve higher DOFS, reflecting their higher observing frequency and pixel resolution (greater density of observations). The GeoCARB information content increases by about 20% when going from 1 to 2 measurements for day, and another 20% when going from 2 to 4 measurements per day. GEO-CAPE provides higher DOFS than GeoCARB, despite coarser pixels, because it measures hourly. We see from Figure 3 that an instrument measuring hourly with 7×7 km^2 pixels would provide the same information as GeoCARB measuring 4 times per day with 3×3 km^2 pixels, and GeoCARB measuring twice a day would provide about 70% of information content obtained from GEO-CAPE hourly measurements. Again, this result depends on the spatial resolution of the inverse problem (here $\sim$25 km). A focus on resolving emissions on finer scales would place a larger premium on decreasing pixel size.

Figure 4 (left panel) examines further the sensitivity of the DOFS to observing frequency for GeoCARB, and the role of the model transport error correlation in limiting the gains from increasing measurement frequency. Without model transport error correlation the DOFS increase roughly as the square root of the measurement frequency (about 40% for each doubling), as would be expected from the central limit theorem. Temporal error correlation significantly reduces but does not eliminate the

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Atmospheric Measurement Techniques Discussions

gain from increasing observing frequency. Thus we find that the DOFS increase by 20-25% instead of 40% for each doubling of the measurement frequency when temporal error correlation is taken into account. Beyond increasing data density, an advantage of more frequent measurements for a region is to increase the opportunity for observing clear-sky scenes ("cloud clearing"), particularly if clouds are more transient than the 6-hour error correlation time scale (in which case multiple observations over that time scale would increase the chance of obtaining a clear-sky value). Cloud cover in the GEOS-FP meteorological data used to drive GEOS-Chem has a persistence time scale typically longer than 6 hours (Figure 2, right panel), which moderates this cloud-clearing benefit of high-frequency observations.

All satellite observing configurations considered in our work have low retrieval success rates because of cloud contamination of individual pixels (Table 1), as determined from the GEOS-FP cloud cover information scaled to match the Remer et al. (2012) clear-sky probability statistics for the Southeast US. These statistics are for summer (regional cloud cover of 70%), but Remer et al. (2012) also give statistics for other seasons with regional cloud cover for the Southeast ranging from 55 to 81%. Figure 4 (right panel) shows the effects of these different cloud statistics on the DOFS for the TROPOMI, GeoCARB, and GEO-CAPE configuration. TROPOMI is strongly sensitive to regional cloud cover because of its coarse pixel size and (to a lesser extent) its infrequent return time. The geostationary systems are far less sensitive.

4 Conclusions

We performed observing system simulation experiments (OSSEs) to compare the ability of low-Earth orbit (TROPOMI) and geostationary (GeoCARB, GEO-CAPE) satellite instruments for constraining methane emissions through inverse analyses. The OSSEs use the GEOS-Chem chemical transport model ($0.25° \times 0.3125°$ grid resolution) in a 1-week simulation for the Southeast US with 216 emission state vector elements. The information content from the different satellite instrument configurations towards quantifying the state vector of emissions is computed as the degrees of freedom for signal (DOFS) using a Bayesian analytical inversion framework.

We find that inverse analysis of TROPOMI observations of atmospheric methane columns should provide a successful regional characterization of methane emissions, though with limited spatial resolution. The information content from TROPOMI is strongly dependent on cloud cover, due to limited cloud-clearing capability (coarse pixels, infrequent return time). Geostationary observations can perform much better, with less dependence on cloud cover, due to a combination of finer pixel resolution and more frequent returns. GeoCARB gains 20-25% in information content for each doubling of its measurement frequency from once to eight times per day. GeoCARB measuring twice a day can deliver 70% of information content from the GEO-CAPE configuration (hourly observations). The benefit of increasing observation frequency is moderated by the 6-h temporal error correlation in the transport model.

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Atmospheric Measurement Techniques Discussions

Table 1. Specifications of satellite instruments.[a]

Instrument	Observing frequency [b]	Pixel size (km^2)	Precision	Cloud contamination [c]	Reference
TROPOMI	once a day	7×7	0.6%	91%	Butz et al. (2012)
GeoCARB	1-4 times a day	3×3	0.6%	73%	Polonsky et al. (2014); O'Brien et al. (2016)
GEO-CAPE	once an hour	4×4	1%	79%	Fishman et al. (2012)

[a] All instruments measure atmospheric methane columns with near-uniform sensitivity in the troposphere, specified here with a typical SWIR averaging kernel (Worden et al., 2015).

[b] All observations are daytime only (SWIR solar back-scatter instruments) and limited to the 9:00-16:00 local time (LT) window. TROPOMI observes at 13:00 LT once a day. GeoCARB observes at 13:00 LT (once a day), 11:00 and 13:00 LT (twice a day), or 9:00, 11:00, 13:00, and 15:00 LT (four times a day). GEO-CAPE observes every hour in the 9:00-16:00 LT window (8 times a day).

[c] Percentage of observing scenes with unsuccessful retrievals due to cloud contamination (Remer et al., 2012).

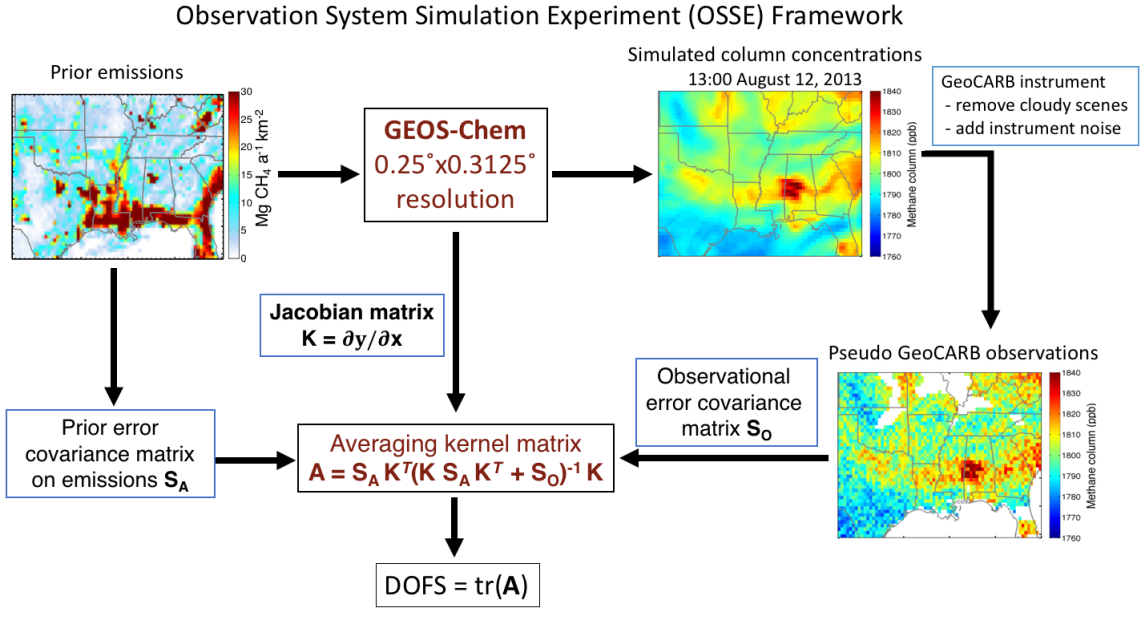

Figure 1. Observing System Simulation Experiment (OSSE) framework for the Southeast US to compare the ability of new satellite instruments to constrain methane emissions on the 25 km ($0.25^\circ \times 0.3125^\circ$) scale. GeoCARB is used here as an example. The right panels show illustrative column concentrations and corresponding GeoCARB observations for a particular time. The column concentrations are in unit of dry molar mixing ratio (ppb). White areas indicate full cloud cover or oceans preventing GeoCARB from making any observations on the 25 km scale.

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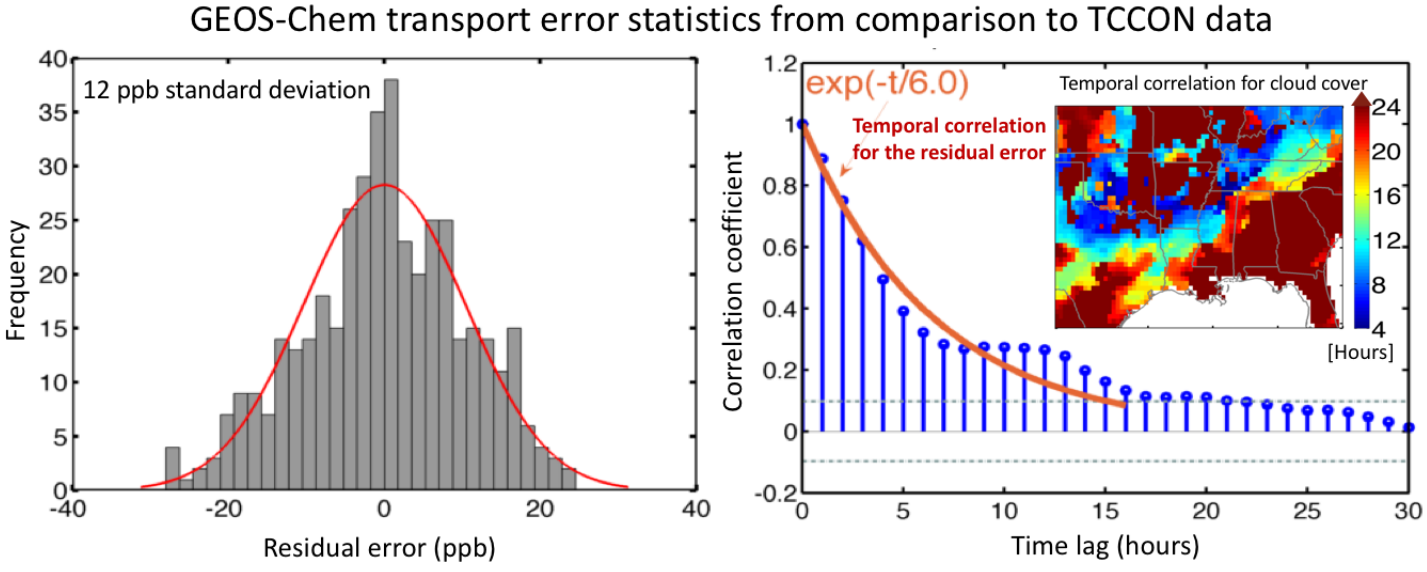

Figure 2. GEOS-Chem model transport error statistics derived from the residual error method (Heald et al., 2004) applied to hourly TCCON ground-based observations in Lamont, Oklahoma, in August-September 2013. Residuals are the differences between hourly simulated and observed values after removal of the mean bias. The left panel shows the frequency distribution of residual error (GEOS-Chem minus TCCON) and a Gaussian fit to that distribution with standard deviation 12 ppb. The right panel panel shows autocorrelation coefficients of the residual error plotted against time lag and an exponential fit with a temporal error correlation e-folding scale of 6 hours. Significance levels ($p < 0.05$) are shown as dashed lines. The correlation becomes insignificant past a time lag of 16 hours. Also shown inset are the temporal e-folding correlation time scales for cloud cover fraction in the GEOS-FP meteorological data for August-September 2013.

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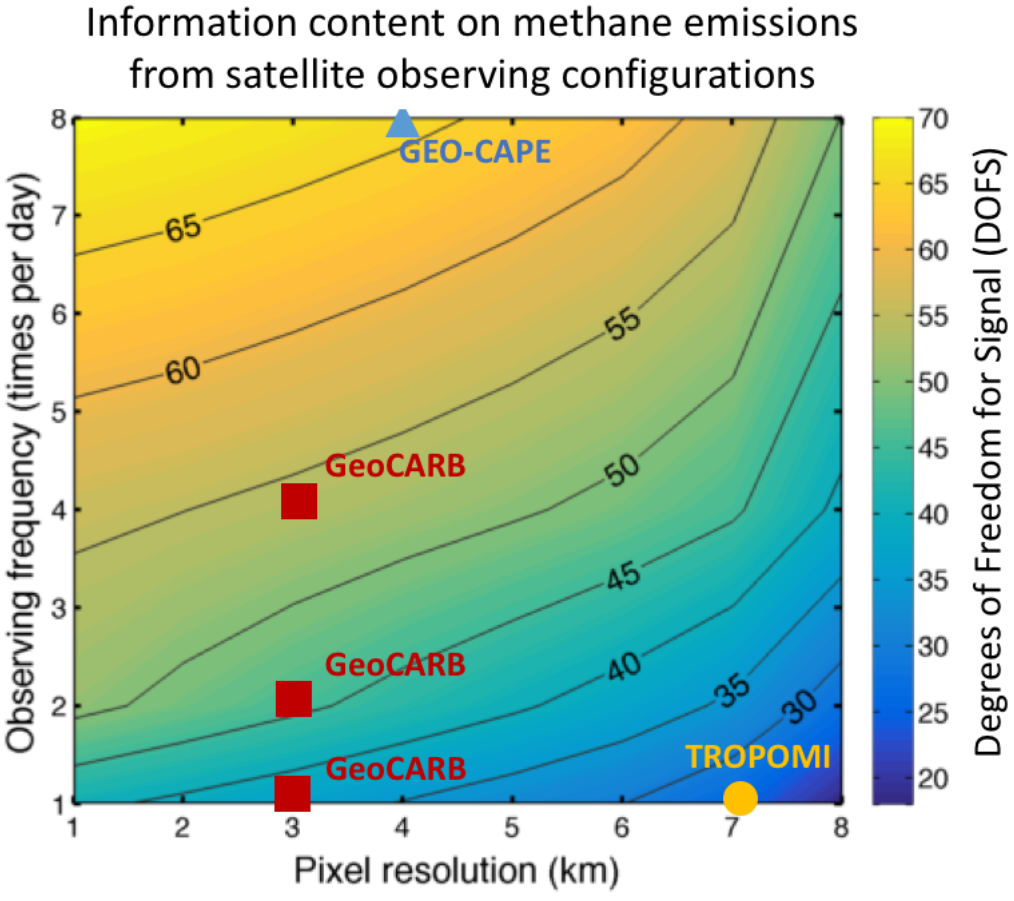

Figure 3. Information content of different satellite observing configurations for constraining the distribution of methane emissions in the Southeast US. The figure shows the Degrees of Freedom for Signal (DOFS) for a 1-week inversion aiming to constrain 216 emission elements in the Gaussian mixture model characterizing the distribution of emissions at up to 25 km resolution. An ideal inversion would have DOFS = 216. The configurations are defined by their observing frequency and pixel resolution. The DOFS for the TROPOMI, GeoCARB (1, 2, and 4 measurements per day), and GEO-CAPE inversions are indicated.

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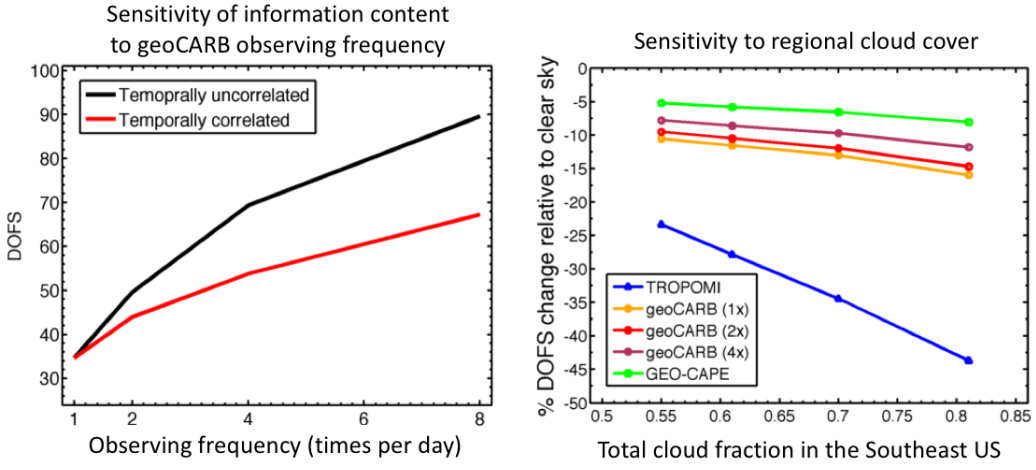

Figure 4. Effects of observing frequency and regional cloud cover on the information content (DOFS) on methane emissions from different satellite observing configurations. The left panel shows the sensitivity of the DOFS to observing frequency for the GeoCARB instrument, with and without accounting for temporal correlation in the model transport error (e-folding time scale of 6 hours).The right panel shows the sensitivity of the DOFS to regional cloud fraction, using the combination of the GEOS-FP cloud cover data and clear-sky probabilities as a function of pixel size (Remer et al., 2012).

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