Mapping of North American methane emissions with high spatial resolution by inversion of SCIAMACHY satellite data

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Abstract

We estimate methane emissions from North America with high spatial resolution by inversion of SCIAMACHY satellite observations using the GEOS-Chem chemical transport model and its adjoint. The inversion focuses on summer 2004 when data from the INTEX-A aircraft campaign over the eastern US are available to validate the SCIAMACHY retrievals and evaluate the inversion. From the INTEX-A data we identify and correct a water vapor-dependent bias in the SCIAMACHY data. We conduct an initial inversion of emissions on the horizontal grid of GEOS-Chem (1/2°x2/3°) to identify correction tendencies relative to the EDGAR v4.2 emission inventory used as a priori. We then cluster these grid cells with a hierarchical algorithm to extract the maximum information from the SCIAMACHY observations. A 1000-cluster ensemble can be adequately constrained, providing ~100 km resolution across North America. Analysis of results indicates that the Canadian wetlands source is lower than the a priori but consistent with other recent estimates. Anthropogenic US emissions are 30.1 ± 1.3 Tg a⁻¹, compared to 25.8 Tg a⁻¹ and 28.3 Tg a⁻¹ in the EDGAR v4.2 and EPA inventories respectively. We find that US livestock emissions are 40% greater than in these two inventories. No such discrepancy is apparent for overall US oil and gas emissions, although this may reflect some compensation between overestimate of emissions from storage/distribution and underestimate from production. We find that US livestock emissions are 70% greater than the oil and gas emissions, in contrast to the EDGAR v4.2 and EPA inventories where these two sources are of comparable magnitude.

1. Introduction
Methane is the second most important anthropogenic greenhouse gas after carbon dioxide [Myhre et al., 2013]. Major anthropogenic sources include natural gas extraction and use, coal mining, landfills, livestock, rice cultivation, and biomass burning. Wetlands are the largest natural source. The magnitude of global methane emissions is constrained within ±15% by knowledge of the global sink from oxidation by OH, but the magnitudes and trends of emissions from different source types and source regions are highly uncertain [Myhre et al., 2013; Hartmann et al., 2013]. Reducing methane emissions has been identified as a low-cost priority in greenhouse gas emissions reduction strategies [IEA World Energy Outlook 2013; van Vuuren et al., 2006; Weyant et al., 2006] but this requires that the sources be quantified. The United States (US) Environmental Protection Agency (EPA) provides national emission inventories for methane [EPA, 2013]. However, a number of studies using atmospheric observations from surface and aircraft suggest that these inventories may underestimate total emissions or emissions from various source types by a factor of two or more [Katzenstein et al., 2002; Xiao et al., 2008; Kort et al., 2008; Petron et al., 2012; Miller et al., 2013a; Karion et al., 2013; Santoni et al., submitted].

Satellite observations of atmospheric methane provide a resource for constraining emissions, as first demonstrated by Bergamaschi et al. [2007], Satellites deliver dense spatial coverage unachievable by surface networks or aircraft campaigns. Methane has been retrieved from nadir satellite measurements of solar backscatter in the short-wave infrared (SWIR) and terrestrial radiation in the thermal infrared (TIR). SWIR retrievals are available from SCIAMACHY for 2003-2012 [Frankenberg et al., 2011] and GOSAT for 2009-present [Parker et al., 2011; Schepers et al., 2012]. TIR retrievals are available from AIRS for 2002-p resent [Xiong et al., 2008], TES for 2004-2011 [Worden et al., 2012], and IASI for 2007-present [Xiong et al., 2008].
SWIR retrievals provide total atmospheric columns. TIR retrievals provide vertical profiles but with low sensitivity to the lower troposphere due to lack of thermal contrast, and this limits their value for detecting regional sources [Wecht et al., 2012].

SCIAMACHY had full global coverage with a six-day return time. Current coverage by GOSAT is much sparser. Instrument degradation limited the value of the SCIAMACHY data after 2005 [Frankenberg et al., 2011]. The TROPOMI instrument to be launched in 2015 will provide SWIR methane data with global daily coverage and 7x7 km$^2$ nadir resolution [Veefkind et al., 2012].

Here we use SCIAMACHY observations for July-August 2004 in an inversion of methane sources in North America with the adjoint of the GEOS-Chem chemical transport model (CTM) at $1/2^\circ\times2/3^\circ$ (~50x50 km$^2$) resolution. This time window takes advantage of concurrent methane observations from the NASA INTEX-A aircraft mission over the eastern US [Singh et al., 2006] that offer extensive vertical profile information (for satellite validation) and boundary layer mapping (for complementary source characterization). The EPA [2013] emission inventory shows no significant change from 2005 to 2011, implying that constraints on 2004 emissions should be relevant to present-day.

A number of previous studies have used SCIAMACHY data for global inverse modeling of methane sources [Bergamaschi et al., 2007; Meirink et al., 2008; Bergamaschi et al., 2009; Bergamaschi et al., 2013; Monteil et al., 2013; Cressot et al., 2013; Houweling et al., 2013]. All have recognized the need for correcting bias in the SCIAMACHY data that otherwise propagates to the inverse solution. An early validation of SCIAMACHY using ground based Fourier transform spectrometers [Dils et al., 2006] failed to identify retrieval error related to inaccuracies in water vapor spectroscopic parameters [Frankenberg et al., 2008]. More recently, Houweling et al. [2013] show that bias in SCIAMACHY is correlated with tropospheric water vapor
concentrations. As we show below, a water vapor correction enables successful validation of the SCIAMACHY data with the INTEX-A vertical profiles.

Our work goes beyond the above studies in using SCIAMACHY for a continental-scale optimization of methane sources with high resolution, including validation and verification with independent aircraft data. The adjoint-based approach allows us to exploit the density of the satellite observations to optimize emissions at the $1/2^\circ \times 2/3^\circ$ native resolution of GEOS-Chem, but we show that too fine a resolution can inhibit successful inversion by diluting the information from the observations. Previous studies have proposed methods for coarsening the discretization of model emissions in a way that optimizes the inversion [Bocquet 2005; Bocquet 2009; Bocquet et al., 2011; Wu et al., 2011]. These methods require laborious construction of the Jacobian of the CTM, which is precisely what we seek to avoid by using the adjoint method. Here we introduce a hierarchical clustering algorithm to optimize the discretization of emissions in the context of adjoint-based inverse modeling.

2. Observations

SCIAMACHY is in a sun-synchronous polar orbit with an equator overpass local time of $\sim 10:00$. It retrieves methane from nadir SWIR spectra at 1.66-1.67 $\mu$m with a nadir footprint of $30 \times 60$ km$^2$ and cross-track scanning. It achieves complete global coverage every 6 days. Observations are limited to daytime and land. We use the Iterative Maximum A Posteriori (IMAP) v5.5 retrieval from Frankenberg et al. [2011]. The retrieval first calculates the methane vertical column density $\Omega_{CH_4}$ [molecules cm$^{-2}$]:

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$\Omega_{CH4} = \Omega_a + a^T (\omega - \omega_A)$  \hspace{1cm} (1)

where $\omega$ is the true vertical profile of methane, consisting of 20 partial columns on a vertical grid, $\omega_A$ is the a priori profile provided by the TM5-4DVAR CTM [Meirink et al., 2008], $\Omega_A$ is the corresponding a priori column concentration, and $a$ is an averaging kernel vector that describes the sensitivity of the retrieved column to each partial column in $\omega$. The sensitivity measured by $a$ is nearly uniform throughout the troposphere and decreases with altitude in the stratosphere. To account for the impact of aerosols and partial cloud cover on the observed light path, $\Omega_{CH4}$ is normalized and converted to a column mixing ratio $X_{CH4}$ [v/v] using the CO$_2$ proxy method described in detail by Frankenberg et al. (2006):

$$X_{CH4} = \left( \frac{\Omega_{CH4}}{\Omega_{CO2}} \right) X_{CO2}$$  \hspace{1cm} (2)

where $\Omega_{CO2}$ is the vertical column density of CO$_2$ also retrieved by SCIAMACHY, and $X_{CO2}$ is a modeled column mixing ratio of CO$_2$. CO$_2$ is used for normalization because it is retrieved in a spectrally neighboring fitting window and its mixing ratio is known with much higher precision than methane.

The IMAP v5.5 product was previously validated by Houweling et al. [2013], who used coincident observations from the Total Carbon Column Observing Network (TCCON) to identify a seasonally-dependent bias that they attributed to water vapor. Here we use in situ vertical profiles from the INTEX-A aircraft during summer 2004. The aircraft flew over the eastern US with extensive boundary layer legs (Figure 1, right panel) and vertical profiles extending up to 12 km. Methane was measured using gas chromatography from whole air flask.
samples collected every 4 minutes with accuracy of 1.0 ppb and precision of 0.1 ppb [Colman et al., 2001; http://www-air.larc.nasa.gov/cgi-bin/arcstat]. For SCIAMACHY validation we require vertical profiles that span from at least 900 to 400 hPa and coincide with SCIAMACHY overpasses within ±150 km and ±6 h. We find 9 profiles satisfying these criteria, each corresponding to 7 to 29 satellite observations. We map the aircraft profiles on the 12 levels of the SCIAMACHY retrieval pressure grid, extrapolate above the DC-8 ceiling using the SCIAMACHY a priori profile, and apply equation (1) to simulate the SCIAMACHY retrieval. From there we derive $X_{CH4}$ by dividing by the local air column density. We average the coincident SCIAMACHY observations and compute the SCIAMACHY-INTEX difference

\[ \Delta X_{CH4} \]

Results indicate a mean bias $\Delta X_{CH4} = -14.2$ ppb (0.8%) and a residual standard deviation of 29.2 ppb (1.6%) for individual SCIAMACHY observations.

Previous studies have demonstrated the need for a latitudinally dependent SCIAMACHY bias correction [Bergamaschi et al. 2007; Meirink et al., 2008; Bergamaschi et al., 2009; Bergamaschi et al., 2013; Cressot et al., 2013]. Some have documented the interference of water vapor as the cause of the bias [Frankenberg et al., 2008; Houweling et al., 2013] and we seek such a relationship here. Figure 2 (left panel) shows the relationship of $\Delta X_{CH4}$ with the average pressure-weighted specific humidity in the 900-400 hPa column measured by the INTEX-A aircraft. There is a linear relationship (weighted $R^2 = 0.69$) that implies a negative bias under dry conditions and a positive dependence of the bias on humidity. We use this relationship to calculate a linear bias correction factor and apply it to the original IMAP v5.5 retrieval. After the correction we find an insignificant mean bias $\Delta X_{CH4} = 2.5$ ppb (0.1 %) and residual standard deviation of 28.2 ppb (1.6 %) for individual SCIAMACHY observations (Figure 2, right panel). The residual standard deviation (which we take to represent SCIAMACHY random
measurement error) is consistent with the average IMAP v5.5 theoretical error of 30.9 ppb (1.7 %) reported by Frankenberg et al. [2011]. All SCIAMACHY data shown here include the specific humidity correction applied with the local GEOS-5 meteorological data used to drive GEOS-Chem.

Figure 1 (left) shows mean SCIAMACHY methane column mixing ratios during the INTEX-A period. Values are highest over the central US where there are large sources from livestock and from natural gas and oil (oil and gas) production. Values are also high over the Canadian wetlands in northern Ontario. The low values in the West reflect elevated terrain so that the stratosphere (where methane is depleted) makes a relatively large contribution to the column mixing ratio. Also shown in Figure 1 are the individual INTEX-A observations in the boundary layer (below 850 hPa). These show areas of high concentrations in the Midwest and East but with fine-scale structure that must reflect in part day-to-day variability in meteorology. We do not use the INTEX-A data for the inversion but use them instead for validation (as described above) and as independent evaluation of the inversion results obtained from SCIAMACHY.

3. Optimization of methane emissions

We optimize methane emissions in North America on the basis of SCIAMACHY observations by Bayesian inference, adjusting an initial (a priori) emission field in order to minimize the difference in $X_{CH_4}$ between SCIAMACHY and GEOS-Chem with error weighting. We describe the procedure and its evaluation below.
3.1 GEOS-Chem model and a priori emissions

We use the GEOS-Chem CTM v9-01-02 (http://acmg.seas.harvard.edu/geos/index.html) as the forward model for the inversion. GEOS-Chem is driven by GEOS-5 meteorological data from the NASA Global Modeling and Assimilation Office (GMAO). The GEOS-5 data have 1/2° latitude x 2/3° longitude horizontal resolution and 6-h temporal resolution (3-h for surface variables and mixing depths). Here we use the native 1/2°x2/3° resolution for GEOS-Chem over North America and adjacent oceans (10-70°N, 40-140°W), with 3-h dynamic boundary conditions from a global simulation with 4° x 5° resolution. This nested North American functionality of GEOS-Chem has been used previously in a number of air quality studies including extensive evaluation with observations [Park et al., 2004, 2006; L. Zhang et al., 2011, 2012; Y. Zhang et al., 2011; van Donkelaar et al., 2012]. These show a good simulation of regional transport with no apparent biases.

The GEOS-Chem methane simulation was originally described by Wang et al. [2004] and updated by Pickett-Heaps et al. [2011]. The main methane sink is tropospheric oxidation by OH, computed using a 3-D archive of monthly average OH concentrations from a GEOS-Chem simulation of tropospheric chemistry [Park et al., 2004]. The mean mass-weighted tropospheric OH concentration is 10.8 x 10^5 molecules cm^-3. Additional minor sinks for methane are soil absorption (from Fung et al. [1991]) and oxidation in the stratosphere. We use stratospheric methane loss frequencies archived from the NASA Global Modeling Initiative (GMI) model [Considine et al., 2008; Allen et al., 2010] as described by Murray et al. [2012]. The resulting global mean atmospheric lifetime of methane is 8.9 years and the lifetime against oxidation by tropospheric OH is 9.9 years. Model intercomparisons in the literature give corresponding values
of 8.6 ± 1.2 years and 9.8 ± 1.6 years [Voulgarakis et al., 2013]. Prather et al. [2012] estimate corresponding values of 9.1 ± 0.9 years and 11.2 ± 1.3 years from observational constraints.

For the a priori emissions we use the 2004 anthropogenic inventory from EDGAR v4.2 with 0.1°x0.1° resolution and no seasonality [EC-JRC/PBL 2009]. Natural sources include temperature-dependent emissions from wetlands [Kaplan et al., 2002; Pickett-Heaps et al., 2011], termites [Fung et al., 1991], and daily GFED3 open fire emissions [van der Werf et al., 2010; Mu et al., 2010]. Figure 3 shows total methane emissions for North America and the contributions from the five largest source types.

Table 1 lists US anthropogenic emission totals by source type in the EDGAR v4.2 and EPA inventories (the EPA inventory is available only as a national total). Total US anthropogenic emissions from EDGAR v4.2 and EPA are 25.8 and 28.3 Tg a⁻¹, respectively.

EDGAR v4.2 and EPA give similar estimates for emissions by source type, except for oil and gas and coal mining. EDGAR reports oil and gas emissions of 6.3 Tg a⁻¹, 30% lower than the EPA [2013] estimate of 9.0 Tg a⁻¹. It reports US coal mining emissions of 3.9 Tg a⁻¹, 40% higher than the EPA [2013] estimate of 2.7 Tg a⁻¹.

Figure 4 shows surface air methane concentrations from the global and nested GEOS-Chem simulations with a priori emissions as described above, compared to observations from the NOAA Global Monitoring Division (GMD) network (http://www.esrl.noaa.gov/gmd/). Boundary concentrations for the nested grid are archived at the edge of the North America domain.

Comparison of GEOS-Chem with the NOAA data over the oceans show that the model simulates realistic latitudinal gradients. This conclusion is supported by comparison of GEOS-Chem to observations from the HIPPO campaign [Wofsy et al., 2012], which shows that GEOS-Chem
simulates northern hemispheric latitudinal gradients without significant error [Turner et al., 2013].

3.2 Inversion Method

We seek to use the SCIAMACHY observations over North America during the INTEX-A period to optimize methane emissions on the 1/2°x2/3° GEOS-Chem grid. Consider the ensemble of SCIAMACHY observations (column mean methane mixing ratios) assembled into an observation vector $y$. Simulation of these observations over the North American domain of GEOS-Chem depends on the gridded emissions within the domain as well as on the boundary conditions (methane background concentrations) at the edges of the domain. We assemble the gridded emissions and the gridded boundary conditions into a state vector $x$. Let $F$ represent the GEOS-Chem model serving as forward model for the inversion. We have

$$y = F(x) + \varepsilon \quad (3)$$

where $\varepsilon$ is the observational error and includes contributions from forward model error, representation error (sampling mismatch between observations and the model), and measurement error. Error statistics are represented by the observational error covariance matrix $S_O = E[\varepsilon \varepsilon^T]$ where $E[\cdot]$ is the expected value operator.

Bayesian optimization weighs the constraints on $x$ from the SCIAMACHY observations with the a priori estimates $x_A$ (error covariance matrix $S_A$). Applying Bayes’ theorem and assuming Gaussian errors leads to an optimized estimate for $x$ by minimizing the cost function $J(x)$ [Rodgers 2000]:
Minimization of $J(x)$ is done with the GEOS-Chem adjoint model, developed by Henze et al. [2007] with application to methane source optimization by Wecht et al. [2012]. The adjoint calculates $\nabla_x J(x_A)$, passes it to a steepest-descent algorithm that returns an improved estimate of $x$, calculates $\nabla_x J(x)$, and iterates until convergence to find $\nabla_x J(x) = 0$. We describe below in more detail the different components of the inversion.

The ability of the inversion to constrain methane emissions over North America is contingent on the model variability being driven by these emissions. Starting from initial conditions, we find that it takes about a week for variability of methane columns over North America in the nested model to be driven by fresh emissions and boundary conditions (as opposed to the initial conditions). We therefore initialize our simulation on 22 June, 9 days prior to assimilating the first observations on 1 July. The lifetime of methane against oxidation by OH is sufficiently long to play no significant role in the variability of methane concentrations over the North America domain.

We initially attempted to optimize North American emissions and boundary conditions as a single state vector in the inversion. This was not successful because boundary conditions have a much larger impact in determining methane concentrations, even if they are less important for determining variability. We therefore iteratively minimize two separate cost functions, $J(x_b)$ and $J(x_e)$, to optimize boundary concentrations and emissions, respectively:

$$J(x_e) = (F(x_e) - y)^T S_o^{-1} (F(x_e) - y) + (x_e - x_{e,A})^T S_{A}^{-1} (x_e - x_{A})$$  

$$J(x_b) = (F(x_b) - y)^T S_o^{-1} (F(x_b) - y) + (x_b - x_{b,A})^T S_{B,A}^{-1} (x_b - x_{B,A})$$  

(4)
\[ J(x_E) = (F(x_E) - y)^T S_o^{-1} (F(x_E) - y) + (x_E - x_{E,A})^T S_{E,A}^{-1} (x_E - x_{E,A}) \]  

(6)

Here the state vectors are \( x_B \), scale factors of boundary concentrations at the edge of the domain relative to the a priori, and \( x_E \), logarithms of scale factors of methane emissions relative to the a priori within the North America domain. We optimize the logarithms of the emission scale factors to ensure positivity in the optimized emissions. A priori values for \( x_B \) and \( x_E \) are labeled \( x_{B,A} \) and \( x_{E,A} \), respectively, and the corresponding a priori error covariance matrices are \( S_{B,A} \) and \( S_{E,A} \).

Each element of \( x_B \) represents a temporally averaged scale factor applied to a 4° x 5° grid cell on the North American boundary (47 vertical levels) for a total of 3290 elements. A priori boundary concentrations are specified from the global GEOS-Chem simulation with a priori emissions (shown in figure 4). The a priori error covariance matrix \( S_{B,A} \) is constructed using error statistics from HIPPO-GEOS-Chem comparisons over the central Pacific presented by Turner et al. [2013]. The diagonal is populated with a model error standard deviation of 16 ppb (0.9%), and off-diagonal terms are parameterized with exponential error correlation length scales of 275 km in the horizontal and 78 hPa in the vertical [Wecht et al., 2012]. We assume that the above error statistics apply to all four boundaries.

Each element \( x_{E,i,j} \) of \( x_E \) represents a temporal average applied to each 1/2° x 2/3° emitting grid cell \((i,j)\) in North America for a total of 7906 elements. It is expressed as follows:

\[ x_{E,i,j} = \ln \left( \frac{E_{i,j}}{E_{A,i,j}} \right) \]  

(7)
where $E_{ij}$ is the true emission flux and $E_{A,ij}$ is the a priori described above.

The emissions a priori error covariance matrix, $S_{E,A}$ is constructed by assuming a uniform relative error standard deviation of 30% for emissions from each model grid cell and no a priori error correlations so that $S_{E,A}$ is diagonal. The sensitivity of the optimized solution to the specification of a priori error will be discussed later by considering an inversion without a priori constraints.

The observational error covariance matrix $S_O$ includes contributions from representation error, measurement error, and GEOS-Chem model error [Heald et al., 2004]. Representation error is assumed to be negligible because SCIAMACHY $X_{CH4}$ observations have horizontal footprints (30 km x 60-120 km) comparable to the size of GEOS-Chem grid cells. We use reported IMAP v5.5 values for the measurement error (standard deviation 30.2 ppb or 1.7%) since these are consistent with our INTEX-A validation (section 2). GEOS-Chem comparison to HIPPO vertical profiles across the Pacific indicates a model error standard deviation of 16 ppb for methane column mixing ratios and we assume that this holds for North America too. All errors are assumed to be Gaussian and are added in quadrature to calculate the observational error for each observation. We do not include error correlation between observations since the overall observational error variance is dominated by the measurement error for which no correlation is expected.

The iterative optimization works as follows. First, we perform five adjoint iterations to reduce $J(x_B)$. We then use the updated values of $x_B$ to calculate $J(x_E)$ and perform five iterations to reduce $J(x_E)$. We use the updated values of $x_E$ to recalculate $J(x_B)$ and repeat. When the reduction of the cost function at each iteration becomes small, after 40 iterations, we hold $x_B$ constant, and iteratively solve $\nabla_{x_E} J(x_E) = 0$. Optimization of $x_B$ corrects background methane
for the inversion and is of peripheral interest here. We focus our discussion on the optimization of emissions in North America.

3.3 Clustering

Figure 5 shows the results from the inversion described above, with optimized correction factors of methane emissions at the 1/2°x2/3° horizontal resolution of the model over North America. Optimized emissions in 93% of grid cells have inversion corrections of less than 30%, as compared to 68% of grid cells that would be expected from the a priori error. This is because the observations have insufficient information to constrain emissions at the native GEOS-Chem resolution.

As the discretization of emissions becomes finer, the observations become less sensitive to emissions from each grid cell. The inversion therefore has less ability to pull emissions in each grid cell away from their a priori value, and the optimal solution will be more tightly constrained by the a priori. This can be seen quantitatively from the minimization of (6):

\[
\nabla_{x_E} J(x_E) = 2(\nabla_{x_E} F)^{T} S_{O}^{-1}(F(x_E) - y) + 2S_{E,A}^{-1}(x_E - x_{E,A}) = 0
\]

where \( \nabla_{x_E} F \) is the Jacobian matrix of the forward model. As the dimension of \( x_E \) increases, the Jacobian matrix values become smaller and thus the individual terms of \( (\nabla_{x_E} F)^{T} S_{O}^{-1}(F(x_E) - y) \) decrease in magnitude as \( (\nabla_{x_E} F)^{T} \) distributes \( S_{O}^{-1}(F(x_E) - y) \) over a larger number of state vector elements. By contrast, the magnitude of individual terms of \( S_{E,A}^{-1}(x_E - x_{E,A}) \) does not change. Thus the a priori increases in importance relative to the observations.
The problem could be mitigated by accurately specifying error correlations in the a priori or by imposing them in the solution, as is done in geostatistical inversions (Michalak et al., 2004). But there is little confidence to be had in the specification of error correlations for methane sources.

We opted therefore to reduce the dimension of our emission state vector by clustering of grid cells, taking advantage of the results from the native-resolution inversion (Figure 5) to group together neighboring grid cells with similar emission scale factors and thus minimize the aggregation error associated with clustering. We tried successively smaller numbers of clusters and repeated the inversion in the same manner described above for the native-resolution inversion, seeking to find the best number of clusters for the inversion as measured by the fit to observations. As we initially decrease the number of clusters starting from the native resolution, we can expect an improved fit of the inversion results to the observations for the reasons discussed above. However, as the spatial resolution of the state vector becomes too coarse (too few clusters), the fit to observations degrades because of aggregation error.

We use a hierarchical clustering algorithm [Johnson, 1967] as a data-driven aggregation technique to optimally define clusters from the native resolution emissions grid. The algorithm initially assigns each 1/2°x2/3° grid cell to its own region, calculates the “distance” to all other regions, and joins the two most similar. Distance is calculated as follows. We define the location for a region $l$ by the vector $v_l=(p, 0.05\times s)^T$ where $p$ is the location of the region centroid on a sphere and $s$ is the mean value of the optimized scale factor from the native resolution inversion presented in Figure 5. All variables are normalized to unit variance and zero mean. The factor 0.05 was selected to adjust the weight of scale factors relative to geographic distance. The distance between two regions $l$ and $m$ is calculated as the norm $||v_l - v_m||$. The process of joining
the two most similar regions proceeds iteratively, reducing the number of regions by one during each step. The algorithm can be stopped at any stage so that any number of clusters can be constructed.

Figure 6 (black) shows the contribution of the model-observation term, 

$$(F(\hat{x}_e) - y)^T S_o^{-1} (F(\hat{x}_e) - y),$$

to the optimized cost function for inversions performed using different numbers of clustered regions. Here $\hat{x}_e$ is the optimal estimate from the inversion. We do not include the a priori term since it depends on the number of clusters used. The best results are achieved for 300-1000 clusters. As the number of clusters decreases from 7906 (native resolution) to 1000, the observations become more sensitive to elements in the state vector, producing a better model fit. As the number of clusters decreases below 300, aggregation error degrades the model fit. The range in the cost function for the different inversions is relatively small because the measurement error dominates for any individual data point. We see from Figure 6 that our optimal estimate for total US anthropogenic emissions is only weakly sensitive to the number of clusters used. We will use the inversion with 1000 clusters as our best estimate in terms of optimization and spatial detail.

3.4 Evaluation with SCIAMACHY and INTEX-A data

The right panel of Figure 5 shows the correction factors to the a priori methane emissions from the 1000-cluster inversion. Figure 7 shows optimized emissions, calculated as the product of optimized correction factors and prior emissions in each grid cell. We checked for improvement of the model fit to the SCIAMACHY data by comparing GEOS-Chem simulations with optimized and a priori emissions and boundary conditions. For this we calculated the GEOS-Chem – SCIAMACHY root-mean-square difference (RMSD) and correlation coefficient.
(R) for the ensemble of 1/2°x2/3° grid cells with SCIAMACHY data, averaged over the July 1 – August 14, 2004 period and weighted by the number of SCIAMACHY observations in each grid cell. We find that the inversion reduces the model-observation RMSD from 11.6 to 9.7 ppb, while R increases from 0.65 to 0.76. This demonstrates improvement, limited by the random noise in the SCIAMACHY measurements.

We further used the boundary layer observations from INTEX-A (Figure 1) to provide independent verification of the inversion results. The model-observation RMSD for individual observations decreases from 33.5 to 28.5 ppb, while R increases from 0.73 to 0.74. Here the improvement appears to be limited by small-scale model and representation error for individual observations.

Averaging of the data allows us to reduce that error and is a more useful comparison. Figure 8 shows boundary layer (>850 hPa) GEOS-Chem – INTEX-A differences averaged on an 8°x10° horizontal grid and for the INTEX-A period. The resulting model-observation RMSD weighted by the number of INTEX-A observations in each 8°x10° grid cell decreases 23.2 to 12.3 ppb when using optimized instead of a priori emissions. The correlation coefficient R increases from 0.69 to 0.88.

We performed sensitivity inversions to investigate the effects of a priori constraints on emissions and model bias. A native-resolution inversion without a priori constraints on emissions shows similar signs and patterns of emission corrections to the inversion with a priori constraints, but the magnitudes of corrections are larger. Evaluation using INTEX-A data averaged into 8°x10° regions as above does not show as good a fit to observations, with an RMSD of 14.5 ppb and R of 0.77. This indicates that the a priori inventory contributes useful information. A sensitivity inversion including a uniform positive bias correction of 15 ppb in
GEOS-Chem on the basis of INTEX-A free tropospheric data shows negligible effect on the correction factors to emissions because most of the bias is absorbed by correction to the boundary conditions.

4. Optimized Methane Emissions

The optimized correction factors in Figure 5 show patterns of increases and decreases relative to the a priori emissions in Figure 3. There is a large decrease in emissions from natural wetlands in northern Ontario and western Canada. There is also a broad decrease in the eastern US, particularly in Appalachia, suggesting an overestimate of emissions from coal mining and waste management. Emissions in the central US increase, suggesting an underestimate of livestock emissions and also possibly from natural gas and oil extraction. We elaborate on source attribution below.

Table 1 shows US anthropogenic emission estimates from EPA, EDGAR v4.2, and this work. Optimized US anthropogenic emissions are 30.1 ± 1.3 Tg a⁻¹, where our best estimate is from the 1000-cluster inversion and our error standard deviation is from the ensemble of inversions with different numbers of clusters in Figure 6 excluding the 3-cluster inversion. Our best estimate is 17% higher than EDGAR v4.2, and 6% higher than EPA (2013), within the stated EPA 95% confidence interval of 14%.

Our inversion optimizes the geographic distribution of emissions without a priori information on source type. It is of interest to determine whether the corrections can be attributed to the particular source types of Figure 3. To do so, we multiply the optimized emission correction factors by the a priori source estimates for each source type and grid cell. This
approach assumes that the relative a priori source distribution in each grid cell is correct. It does 
not assume that a priori spatial distributions of sources are correct because spatial patterns will 
change after applying the correction factors. We estimate uncertainties on the basis of the 
ensemble of values for the inversions with different clusters, recognizing that they are likely 
underestimates as all inversions make the same assumption about correctness of the a priori 
source distribution.

To calculate annual emissions from our inversion results, we must assume knowledge of 
the seasonality of emissions. Most anthropogenic emissions are assumed to be aseasonal, as in 
the EDGAR v4.2 inventory. The EPA [2013] inventory, however, estimates livestock emissions 
during the summer (JJA) to be 16% higher than the annual average, driven by temperature 
sensitivity of emissions from manure management. We account for this seasonality when 
estimating annual emission rates in this study.

Results in Table 1 show that our emission estimate for livestock is 40% higher than the 
EDGAR v4.2 and EPA inventories and represents the largest US source. Our oil and gas source 
is intermediate between EDGAR v4.2 and EPA and is smaller than the livestock source source. 
Other sources are also smaller, across all inventories. Our landfills source is consistent with both 
EDGAR v4.2 and EPA while our coal mining source is smaller than EDGAR v4.2 but consistent 
with EPA.

However, the patterns of correction factors from our inversion in Figure 5 reveal structure 
that cannot be simply explained by the EDGAR v4.2 source types. A multiple linear regression 
of absolute corrections on the distributions of individual EDGARv4.2 and natural a priori 
emissions yields an $R^2$ of only 0.21. For example, our correction factors in Figure 5 indicate a 
large EDGAR underestimate of livestock emissions in Iowa, where hog manure is important, but
a decrease in eastern North Carolina where hog manure is important too. This could reflect differences in manure management practices. Our inversion also calls for a large increase in emissions from the Permian Basin in western Texas, a major oil and gas production region, but the EDGAR v4.2 inventory is very low there. This suggests that oil and gas emissions in EDGAR are too heavily weighted by the distribution and end use sectors relative to the production sector.

5. Comparison to previous studies

A number of previous studies have used methane observations from surface sites and aircraft as top-down constraints on methane emissions in North America. We discuss here the consistency of our results.

There has been much interest in quantifying wetland emissions from in the Hudson Bay Lowlands (HBL) of northern Ontario, as this is the second largest area of boreal wetlands in the world after western Siberia. Pickett-Heaps et al. [2011] reviewed previous studies and estimated an HBL source of 2.3 Tg a\(^{-1}\) from aircraft and surface observations. Miller et al. [2013b] estimated a source of 2.4 Tg a\(^{-1}\) using tall tower observations. We find here a consistent estimate of 2.1 Tg a\(^{-1}\), accounting for the seasonality given by Pickett-Heaps et al. [2011] with strong peak in June-August. This calls for downward revision of the Kaplan et al. [2002] source used as a priori in GEOS-Chem.

The CalNex aircraft campaign in May-June 2010 provided constraints on methane emissions from California through a series of boundary layer flights across the state. Inverse analyses of the CalNex data by Santoni et al. [submitted] and Wecht et al. [submitted] indicate
statewide emissions of 2.4-2.8 Tg a\(^{-1}\), Los Angeles Basin emissions of 0.3-0.4 Tg a\(^{-1}\), and a factor of 2-4 underestimate of livestock emissions in the EDGAR v4.2 inventory for the Central Valley. Our inversion of the SCIAMACHY data is closely consistent with these results indicating a statewide emission of 2.1 Tg a\(^{-1}\) in California, 0.2 Tg a\(^{-1}\) in the Los Angeles Basin, and a factor of 2.6 underestimate in livestock emissions relative to EDGAR v4.2. The livestock underestimate is larger for California than the national underestimate of 40% reported earlier, and provides further evidence of spatial errors in emission factors in the EDGAR inventory.

Miller et al. [2013a] estimated methane emissions across the US using a network of surface and aircraft data from 2007-2008. Their optimal estimate for US anthropogenic emissions is 44.5 Tg a\(^{-1}\), much higher than our value of 30.1 Tg a\(^{-1}\). They increase emissions in the central US relative to EDGAR and decrease emissions in Appalachia, similar to the spatial patterns reported here. Their observations, however, are relatively sparse east of the Great Plains and may not adequately characterize the emissions reductions throughout Appalachia and the northeast that are required by SCIAMACHY and consistent with the INTEX-A data. Miller et al. [2013a] estimate a factor of 2.3 increase relative to EDGAR v4.2 for the northern plains (Nebraska, Iowa, Wisconsin, Minnesota, and South Dakota), a region of high livestock density and few other sources of methane. This compares well to our factor of 2.2 increase for the region, again higher than the national average for livestock emissions.

Katzenstein et al. [2003] measured methane concentrations on a road survey across Texas, Oklahoma, and Kansas. Assuming a mean boundary layer height and a characteristic ventilation time for the region, they estimated a methane emission of 4-6 Tg a\(^{-1}\) for that tri-state region. Assuming that these emissions are mainly from natural gas and oil, they concluded that EPA emission estimates are too low by a factor of 2.5. Our inversion indicates a methane
emission of 8.7 Tg a\(^{-1}\) for the region, greater than the Katzenstein et al. [2003] estimate and possibly reflecting their oversimplified ventilation model. In any case, their assumption that oil and gas dominate sources in the region may not be valid as the EDGAR v4.2 inventory for the region (4.0 Tg a\(^{-1}\)) assigns 52% of methane emissions to livestock and only 29% to oil and gas.

Miller et al. [2013a] estimate emissions of 10.8 Tg a\(^{-1}\) for the region and attribute the underestimate to both livestock and oil and gas. Most of the emission correction for the region in our inversion is from livestock.

Xiao et al. [2008] derived a US ethane emission of 2.4 Tg a\(^{-1}\) from analysis of INTEX-A observations and combined this with independent estimates of ethane-to-methane emission ratios to deduce a US fossil fuel methane emission of 16 Tg a\(^{-1}\). This would include contributions from natural gas, oil, and coal mining. By comparison, EPA and EDGAR v4.2 estimate US fossil fuel emissions of 11.7 and 10.1 Tg a\(^{-1}\), respectively. Using the source-type distributions provided by EDGAR, we calculate a fossil fuel methane source of 9.6 Tg a\(^{-1}\). Uncertainty on ethane-to-methane emission ratios may have affected the Xiao et al. [2008] estimate.

Kort et al. [2008] used Lagrangian modeling of observations from the COBRA-NA aircraft campaign across North America in 2003 to evaluate US and Canada emissions. From a single linear regression of modeled vs. observed methane they estimated US anthropogenic emissions of 41±6 Tg a\(^{-1}\), larger than our best estimate of 30.1 Tg a\(^{-1}\). Their observations, however, are only sensitive to emissions from a relatively small fraction of the US and Canada.

Petron et al. [2012] and Karion et al. [2013] used in situ observations to estimate methane leak rates of 4% and 6-12% of total natural gas production Weld County, CO and Uintah County, UT, respectively. In contrast, the EDGAR v4.2 and EPA inventories assume a national average leak rate of 1.0-1.4%. Our inversion of the SCIAMACHY data does not indicate higher-than-
expected natural gas emissions from these two counties but this would not account for post-2004 growth. Methane leakage rates can vary considerably by basin [US Government Accountability Office, 2010].

6 Conclusions

We used SCIAMACHY satellite observations in a high-resolution continental-scale inversion of methane emissions in North America driven by the GEOS-Chem chemical transport model (CTM) and its adjoint at 1/2°x2/3° horizontal resolution. The inversion focused on summer 2004, when concurrent observations from the INTEX-A aircraft observations are available to both validate the SCIAMACHY data and independently evaluate the inversion. The high density of observations available from SCIAMACHY enables higher spatial detail in constraining methane emissions on the continental scale than had been achievable before.

Removal of observational bias is essential for a successful inversion. Our validation of the SCIAMACHY observations (IMAP v5.5) with INTEX-A vertical profiles identified a systematic bias correlated with water vapor, consistent with previous studies. We found that we could successfully correct for this bias, and the residual error is consistent with the theoretical error estimate from the IMAP v5.5 retrieval.

Continental-scale inversion for methane required accurate specification of the boundary conditions in GEOS-Chem. This was accomplished by optimizing both the North American emissions and the boundary conditions as part of the inversion. We found that the information content from the SCIAMACHY data was insufficient to constrain emissions at the native 1/2°x2/3° horizontal resolution of GEOS-Chem. We solved this problem by using a hierarchical...
clustering algorithm to identify 1000 geographical clusters for which the inversion provides optimal results. The optimized emissions obtained from the 1000-cluster inversion were independently evaluated by GEOS-Chem simulation of the INTEX-A aircraft data. This demonstrated a major improvement over the simulation driven by a priori emissions.

Our optimized methane emissions for the Canadian wetlands are lower than the a priori but consistent with recent studies. Our optimized methane emissions for the US are lower than the EDGAR v4.2 inventory for the eastern US but higher for the central US. Our best estimate of US anthropogenic emissions is $30.1 \pm 1.3$ Tg a$^{-1}$, compared to $25.8$ Tg a$^{-1}$ and $28.3$ Tg a$^{-1}$ in the EDGAR v4.2 and EPA inventories respectively. Source attribution of our optimized methane emissions on the basis of the EDGAR patterns suggests that the above inventories underestimate livestock emissions by 40%, with smaller discrepancies for other sources. We find that livestock emissions in the US are 70% higher than oil and gas emissions, whereas the EPA inventory reports these two sources to be of comparable magnitude. However, we find in a regression analysis that the EDGAR patterns can account for only 21% of the variability in the source correction from the inversion. This implies large inventory errors in the geographic variability of emission factors (e.g., livestock management practices) and activity rates (e.g., oil and gas production in the West).

Our finding that US livestock emissions are underestimated in current inventories is consistent with previous regional studies. The degree of underestimate seems highly variable for different parts of the country, suggesting large variability in emission factors. Our finding that oil and gas emissions are not underestimated in current inventories is at odds with previous studies and may partly reflect variability in leakage rates and assumptions made in all studies to perform source type attribution.
Emissions of methane in North America may be rapidly changing in the future as a result of increasing oil and gas production, changes in recovery practices, evolving regulations, and climate change affecting wetlands. The GOSAT satellite observations (2009-present) may be useful to track recent trends but are relatively sparse. The TROPOMI instrument to be launched in 2015 will provide global daily coverage with 7x7 km² nadir spatial resolution and precision of 0.6% [Veefkind et al., 2012; Butz et al., 2012]. This will provide a tremendous boost to monitoring methane emissions from space.

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Figure 1 – Methane over North America during the INTEX-A aircraft campaign (1 July – 14 August 2004): SCIAMACHY mean column mixing ratios (left) and INTEX-A mixing ratios below 850 hPa (right). Observations are averaged spatially on the 1/2°x2/3° GEOS-Chem grid and temporally over the INTEX-A campaign duration. Note the difference in scales between the two panels.
Figure 2. Validation of the SCIAMACHY IMAP v5.5 retrieval of methane column mixing ratio ($X_{CH4}$) with coincident INTEX-A aircraft vertical profiles (see text). Left: difference $\Delta X_{CH4}$ between SCIAMACHY and INTEX-A plotted as a function of the mean pressure-weighted specific humidity in the 900-400 hPa column measured by the INTEX-A aircraft. The red line shows a linear regression weighted by the number of SCIAMACHY observations. Regression parameters and weighted $R^2$ are shown inset. Right: comparison of SCIAMACHY and INTEX-A $X_{CH4}$ after applying the water vapor correction from the linear regression. Weighted $R^2$ is shown inset and the 1:1 line is also shown. Colors represent the number of SCIAMACHY observations averaged around each INTEX-A profile.
Figure 3 – North American methane emissions used as a priori for the inversion: total emissions (top left panel) and contributions from the major source types. Inventories are from Kaplan et al. (2002) and Pickett-Heaps et al. (2011) for wetlands and from GEIA v 4.2 for all other (anthropogenic) sources. Values are averages for 22 June – 15 August 2004. Annual emission rates for 2004 (Tg a\(^{-1}\)) are shown inset for the North America domain as encompassed by the figure.
Figure 4 – Methane concentrations in surface air averaged over the inversion period (22 June – 14 August 2004). The GEOS-Chem simulation with a priori sources (background) is compared to NOAA GMD observations (circles). Left: global simulation at 4°x5° resolution used to archive a priori boundary concentrations for the nested simulation. Right: nested simulation at 1/2°x2/3° resolution for the North America domain. Note difference in scale between panels. The NOAA GMD data were obtained from http://www.esrl.noaa.gov/gmd/.
Figure 5 – Emission scale factors relative to the a priori (top left panel of Figure 3) from inversions optimizing emissions on the $1/2^\circ \times 2/3^\circ$ native resolution of GEOS-Chem (left) and for 1000 clustered regions (right). Gray areas (ocean/ice) are not included in the state vector for the inversion.
Figure 6 – Sensitivity of inversion results to the resolution with which North American methane emissions are optimized from the SCIAMACHY data for 1 July – 14 August 2004. Resolution is expressed as the number of spatial clusters used in the inversion. The maximum of 7906 clusters represents the native 1/2°x2/3° grid of GEOS-Chem. Optimal aggregation of grid cells based on proximity and emission correction tendencies yields successively smaller numbers of clusters. Black points show the model-observation term of the cost function \((F(\hat{x}_E) - y)^T S_o^{-1} (F(\hat{x}_E) - y)\) describing the ability of the cost function to fit the SCIAMACHY observations. Red points show optimized US anthropogenic emissions for each inversion.
Figure 7 – Optimized North American methane emissions from the 1000-cluster inversion. The annual emission rate for 2004 (Tg a\(^{-1}\)) is shown inset for the North America domain as encompassed by the figure.
Figure 8 – Independent evaluation of the SCIAMACHY inversion of methane emissions using INTEX-A aircraft data. The panels show the mean differences between GEOS-Chem and INTEX-A observations below 850 hPa and for 8°x10° grid squares in the simulation with a priori emissions (left) and with optimized emissions from the 1000-cluster inversion (right). A priori and optimized emission maps are shown in Figures 2 and 7. The model-observation root mean square difference (RMSD) and weighted correlation coefficient (R) are inset.
Table 1. – US sources of methane in 2004 [Tg a⁻¹].

<table>
<thead>
<tr>
<th>Source type</th>
<th>EPA (2013)¹</th>
<th>EDGAR v4.2²</th>
<th>Miller et al. (2013)³</th>
<th>This Work⁴</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total</td>
<td></td>
<td>47.2±1.9</td>
<td>38.9±1.4</td>
<td></td>
</tr>
<tr>
<td>Anthropogenic</td>
<td>28.3 (24.6, 32.3)</td>
<td>25.8</td>
<td>44.5±1.9</td>
<td>30.1±1.3</td>
</tr>
<tr>
<td>Livestock</td>
<td>8.8 (7.7, 10.4)</td>
<td>8.5</td>
<td>16.9±6.7</td>
<td>12.2±1.3</td>
</tr>
<tr>
<td>Natural Gas and Oil</td>
<td>9.0 (7.2, 13.4)</td>
<td>6.3</td>
<td></td>
<td>7.2±0.6</td>
</tr>
<tr>
<td>Landfills</td>
<td>5.4 (2.5, 7.9)</td>
<td>5.3</td>
<td></td>
<td>5.8±0.3</td>
</tr>
<tr>
<td>Coal Mining</td>
<td>2.7 (2.3, 3.2)</td>
<td>3.9</td>
<td></td>
<td>2.4±0.3</td>
</tr>
<tr>
<td>Other⁵</td>
<td>2.4 (1.4, 4.2)</td>
<td>1.9</td>
<td></td>
<td>2.5±0.2</td>
</tr>
<tr>
<td>Natural⁶</td>
<td></td>
<td></td>
<td>2.7</td>
<td>6.9±0.5</td>
</tr>
</tbody>
</table>

¹Values in parentheses represent lower and upper ends of 95% uncertainty ranges.
²Used as a priori for the inversion.
⁴Uncertainties on emissions from individual source types assume that the relative a priori source distribution in each grid cell is correct (see text).
⁵Including waste water treatment, rice cultivation, biofuel use, and other small sources.
⁶Including natural wetlands, termites, open fires, and soil absorption.

According to EPA (2013), none of these sources account for more than 3% of total US anthropogenic emissions.