We thank the referees for their helpful comments that have improved our paper.

Anonymous Referee #1

Summary/General comments: Sheng et al. present a high resolution inversion of SEAC4RS aircraft methane data to find optimal methane emissions in that region in that time frame. They find the new, gridded EPA inventory is consistent with their observations, while WETCHIMP methane emissions are found to be too high. This paper is well placed in ACP. Overall the paper is well written, clear, and adds to our understanding of methane emissions in the US. I have only a couple of concerns – once these are addressed I would encourage publication.

Major comments: I have two larger concerns: The definition of region: The region studied is defined as the Southeast US and is illustrated in Figure 1. My problem is that the SEAC4RS data does not constrain emissions for this whole region. In fact, less than half the domain has a significant sensitivity (AK sensitivity) and thus is informed by the analysis. Important regions that have large fluxes in the prior inventories (fossil in TX, wetland in FL, GA, SC) are not constrained by the work but are included in the regional flux estimates. This should be corrected. At a minimum, the area defined should be compressed to not include large expanses with minimal surface sensitivity in the west. Even better, would be to use the AK sensitivity to filter only the domain where there is significant surface constraint. This could be illustrated in Figure 1, and that mask could be applied to the domain for flux estimation. This would be straightforward for the authors to do and would make the results more robust.
We have updated Figure 1, Figure 5, and the text accordingly.

Transport error/60 ppb: I'm a little unclear on how 60 ppb was settled on as the observation-model error. Transport error could be significant, and I would like to see more discussion/explanation of how that is accounted for. If the 60 ppb is derived from the observations. I wonder if it is more representative of atmospheric variability?
60 ppb is derived from the statistics of residual errors (differences between observed and simulated values after removing the mean model bias). We have expanded discussion.

Minor comments: Page 1 Line 3: The % will be lower once accounting for the above comment.
We have updated the text as: "The Southeast US is a major source region for US methane emissions including large contributions from oil/gas production and wetlands."

Page 1 Line 11: It's not clear you can state your work is in contrast with national scale work. Not only is the work on different scales, it is for different years, and, more importantly, different times of year. This study is focused on only Aug-Sept, whereas other studies have used 1+ year of data. This needs to be made clearer throughout that the results are for Aug.-Sept. only.
Little regional or interannual variability is expected for anthropogenic emissions and so our results have general applicability. This is now stated in the text with references. For wetlands we have added a few statements that our results are for Aug-Sep 2013.

Page 2 Line 9: Should indicate here at least once if Tg C or Tg CH4.
Done.

Page 2 Lines 15-20: This is not strictly accurate. Miller et al., 2013 did not rely on EDGAR, as a geostatistical approach was used. I would suggest changes phrasing here to correct this. (A side comment: the Miller work did have little data in the Southeast so it was essentially unconstrained).

We corrected this.

Page 4 Lines 5-6: I have some concerns about the way the flask data has been aggregated onto a grid here. It would be very helpful to see some continuous variables for the flights and where the flasks were sampled. This would enable the reader to assess if averaging the discrete flask samples is representative of 0.25 degree boxes, or if the samples are representative of smaller atmospheric features (such as plumes).

We now plot the continuous data in Fig. 2, and update the text accordingly.

Page 4 Line 17: I'd like more on the 60 ppb.

See the response in major comments.

Page 6 Lines 10-20. Care is needed here as mentioned before not all these studies relied to this level on EDGAR. Also, many of those studies were rather unsensitive to the SE, so they likely do not see the wetland emissions, as opposed to falsely attributing those emissions to anthropogenic sources. At the least, we cannot make the conclusion in lines 18-19.

Agree. We now omit this conclusion.

Page 6 lines 29-31: these fractions will come down when accounting for region of sensitivity.

Done.

Page 6 line 7: Should specify this finding of regional consistency is for Aug.-Sept. only.

See response above.

Figure 1: Zoom and add contours as described in major comment.

We have updated Fig. 1.

Figure 2: I struggle with the map figure as it is hard to interpret the methane concentration on a map like this where we don't know if it is when the mixing layer is deep or shallow or what the background value is on the given day. For example, are regions with blue and red adjacent indicative of high spatial variability or from different sampling?

See the response for "Page 4 Lines 5-6:…"

Figure 5: Update when updating domain definition.

Done.

Anonymous Referee #2
Received and published: 13 February 2018
General comments
This study uses observations of CH4 from aircraft campaigns in Aug-Sep 2013 to estimate
CH4 emissions in the southeast US, a region with importance to the US total
anthropogenic CH4 emission and with significant areas of wetlands. The authors use
a Bayesian inversion method to estimate the emissions and uncertainties. The method
is scientifically sound and the manuscript is well-written. However, there are a few
outstanding points that should be clarified before publication. In particular, I think the
results of the study would be strengthened by adding a sensitivity test to determine the
sensitivity of the results to the prior wetlands emission estimate used (see also specific
comments). In summary, I recommend publication after minor revisions.
See the response below in specific comment.

Specific comments
P1, L13: The authors state that the results of previous inversions finding higher emissions
than EPA estimates were owing to too low estimates for wetland emissions. What
estimate for the wetland emissions is the reference here?
We have removed this statement (see response to Referee #1). It's not relevant now.

P2, L19-20: While errors in the prior can bias the results, the extent to which this is a
problem is dependent on the constraint from the observations and on the uncertainty
assigned to the prior estimates, or in other words the degrees of freedom to adjust the
prior. This statement should be qualified bearing in mind these other factors too.
We now add: "… this depends on the constraint from the observations and on the uncertainty
assigned to the prior estimates."

P3, Eq.1: Strictly speaking, the RHS of this equation should be multiplied by ½
We have corrected the equation.

P3, L14: Here the authors say that inversions of methane are usually solved numerically,
however, that is not the case. While global inversions of CH4 over many years
usually use numerical adjoint methods, regional inversions (as in this study) also often
use the analytical solution.
We have updated the text as:
"…analytically or numerically using an adjoint method (Jacob et al., 2016). Unlike adjoint-based
inversions, analytical solution provides direct error characterization …".

P3, L15: While it is true that the analytical method allows the posterior error covariance

to be calculated directly, some numerical methods allow it to be estimated.
We have added 'direct'

P3, L26: Did the global simulation optimized with GOSAT also include surface observations? This should be mentioned. Also, it has been shown that satellite-only optimizations can lead to be biases due to errors in the retrievals. Have comparisons of the CH4 mixing ratios from the optimized simulation against surface and/or aircraft observations been made?
The GOSAT optimized emissions are evaluated with independent surface observations. We now mention this in the text.

P4, L12: I think it would be helpful if the authors would briefly explain the residual method, which is used to calculate the observation error variances.
We now do so. See response to Referee #1.

P4, L28-29: I'm not sure how the inversion can return information at the fine scale when it is performed at coarser scale. I think further explanation would be helpful here.
The statement is not strictly accurate. We have removed it.

P5, L20-25: There is a little bit of overlap in the locations of the anthropogenic and wetland sources (see Fig. 1). Has this been factored into the posterior emission estimates per source type?
The sources are in fact well separated on the 0.25°x0.3125° grid and we now say so.

P6, L8-9: The WetCHARTs ensemble mean was used as the prior for the inversion, therefore, it is not surprising that the ensemble mean has the lowest RMSE compared to the posterior emissions, as the two estimates are not independent from one another. This should be stated. Also, have the authors looked at the Gain matrix to determine how well constrained the wetland emissions are by the observations? It would be valuable to test how sensitive the results are to using a different prior for the wetland emissions.
We agree and now state this.
Results will be sensitive to different wetland ensemble members in particular those smallest or largest members, assuming the same prior uncertainty. We now also state this in the text.

Technical comments
P1, L5: I don't think the term "state-of-science" exists (or at least I've never heard it before). I suggest replacing with "state-of-the-art" or "up-to-date" (also elsewhere in the manuscript).
Done.

High-resolution inversion of methane emissions in the Southeast US using SEAC[4]RS aircraft observations of atmospheric methane: anthropogenic and wetland sources

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Abstract. We use observations of boundary layer methane from the SEAC[4]RS aircraft campaign over the Southeast US in August-September 2013 to estimate methane emissions in that region through an inverse analysis with up to $0.25° \times 0.3125°$ (25×25 km^2) resolution and with full error characterization. The Southeast US ~~accounts for about half of total US anthropogenic emissions according to the gridded EPA national inventory and also has extensive~~ is a major source region for

5   methane including large contributions from oil/gas production and wetlands. Our inversion uses ~~state-of-science~~ state-of-the-art emission inventories as prior estimates, including a gridded version of the anthropogenic EPA Greenhouse Gas Inventory and the mean of the WetCHARTs ensemble for wetlands. Inversion results are independently verified by comparison with surface (NOAA/ESRL) and column (TCCON) methane observations. Our posterior estimates for the Southeast US are 12.8±0.9 Tg a^{-1} for anthropogenic sources (no significant change from the gridded EPA inventory) and 9.4±0.8 Tg a^{-1} for wetlands

10 (27% decrease from the mean in the WetCHARTs ensemble). The largest source of error in the WetCHARTs wetlands ensemble is the landcover map specification of wetland areal extent. ~~We find no regional bias in the anthropogenic EPA inventory, including for different source sectors, in contrast with previous inverse analyses that found the EPA inventory to be too low at national scales. These previous inversions relied on prior anthropogenic source patterns from the EDGAR v4.2 inventory that have considerable error, and also assumed low wetland emissions. Despite the regional-scale consistency, we find significant~~

15 ~~local errors in the EPA inventory~~ Our results support the accuracy of the EPA anthropogenic inventory on a regional scale but there are significant local discrepancies for oil/gas production fields ~~,~~ suggesting that emission factors are more variable than assumed in the EPA inventory.

1 Introduction

Methane is an important greenhouse gas (Myhre et al., 2013)~~for which individual countries report~~. Individual countries must report their national emissions to the United Nations Framework Convention on Climate Change (UNFCCC; United Nation, 1992). Observations of atmospheric methane reviewed by Brandt et al. (2014) have implied that the US national inventory

5 reported by the Environmental Protection Agency (EPA) may be greatly underestimated. Here we use aircraft observations from the NASA SEAC[4]RS aircraft campaign over the Southeast US (Toon et al., 2016), together with a newly gridded version of the EPA inventory (Maasakkers et al., 2016), in a fine-resolution inversion with detailed error characterization to better quantify ~~the sources of~~ methane emissions over this major source region.

The EPA (2016) reports a national anthropogenic emission total of 29.2 Tg CH_4 a^{-1} for 2014, with no significant trend over

10 the past decade and less than $\pm 3\%$ interannual variability. Major contributors are livestock (32%) , the oil/gas industry (32%), waste (22%), and coal mining (8%). The EPA (2016) inventory is consistent with Lyon et al. (2015) for oil/gas systems and Wolf et al. (2017) for livestock, and 8% higher than the previous versions (EPA, 2013, 2014), largely due to updated oil/gas emissions. There is also a highly uncertain natural source from wetlands, estimated at 4.5-14 Tg a^{-1} for the contiguous US in the WETCHIMP compilation of inventories (Melton et al., 2013). Inverse analyses of atmospheric methane observations have

15 suggested that the EPA bottom-up inventory (EPA, 2013, 2014) is too low by about 30% (Miller et al., 2013; Turner et al., 2015; Alexe et al., 2015)~~, but they~~. However, Turner et al. (2015) and Alexe et al. (2015) relied on prior estimates from the global EDGAR v4.2 inventory (European Commission, 2011) that have large errors in source patterns particularly for oil/gas systems (Maasakkers et al., 2016; Sheng et al., 2017). ~~For example, EDGAR v4.2 does not account for the large source from oil/gas production in the Southeast US but the gridded EPA inventory does (Maasakkers et al., 2016).~~ Errors in source patterns

20 used as prior estimates can greatly bias inversion results (Jacob et al., 2016)~~.~~, though this depends on the constraint from observations and on the uncertainty assigned to the prior estimates. Miller et al. (2013) used a geostatistical inversion that did not rely on any prior estimates, but had little constraints in the Southeast US.

The SEAC[4]RS aircraft campaign conducted in August-September 2013 offers an opportunity for better estimating methane emissions in the Southeast US, a region that accounts for about half of total anthropogenic methane emissions in the US

25 according to the gridded EPA inventory (Maasakkers et al., 2016) and also has extensive wetlands. The aircraft flights provided extensive ~~boundary-layer~~ boundary layer measurements of methane across the region. We conduct an inverse analysis of the SEAC[4]RS data with the GEOS-Chem chemical transport model (CTM) at $0.25° \times 0.3125°$ resolution, using ~~state-of-science~~ state-of-the-art prior estimates from the gridded EPA inventory of Maasakkers et al. (2016) and the WetCHARTs extended ensemble wetlands inventory of Bloom et al. (2017). This allows us to evaluate the EPA inventory with better accuracy and

30 resolution than has been done before, and also to gain better understanding of US wetland emissions.

2 Methods

We derive an optimized estimate of spatially resolved methane emissions in the Southeast US (domain of Fig. 1) by Bayesian inverse analysis of atmospheric methane observations from the SEAC[4]RS aircraft campaign. Let the vector $\mathbf{x}$ represent a

gridded ensemble of methane emissions in the region (state vector for the inversion).The inversion minimizes the cost function $J(\mathbf{x})$ by solving $\nabla_{\mathbf{x}} J(\mathbf{x}) = \mathbf{0}$:

$$J(\mathbf{x}) = (\mathbf{x} - \mathbf{x_A})(\mathbf{x}) = \frac{1}{2}(\mathbf{x} - \mathbf{x_A})^T \mathbf{S_A^{-1}}(\mathbf{x} - \mathbf{x_A}) + (\mathbf{y} - \mathbf{Kx})\mathbf{S_A^{-1}}(\mathbf{x} - \mathbf{x_A}) + \frac{1}{2}(\mathbf{y} - \mathbf{Kx})^T \mathbf{S_O^{-1}}(\mathbf{y} - \mathbf{Kx}). \tag{1}$$

Here the methane observations are assembled as a vector $\mathbf{y}$, $\mathbf{x_A}$ is the prior emission estimate, $\mathbf{K}$ is the Jacobian matrix describing the sensitivity of concentrations to emissions, and $\mathbf{S_A}$ and $\mathbf{S_O}$ are the prior and observational error covariance matrices, respectively. The observational error includes contributions from both the instrument error and the model transport error.

Analytical solution of $\nabla_{\mathbf{x}} J(\mathbf{x}) = \mathbf{0}$ yields the optimal estimate $\hat{\mathbf{x}}$, the posterior error covariance matrix $\hat{\mathbf{S}}$, and the associated averaging kernel matrix $\mathbf{A}$ (Rodgers, 2000; Brasseur and Jacob, 2017)

$$\hat{\mathbf{x}} = \mathbf{x_A} + \mathbf{S_A}\mathbf{K}^T(\mathbf{K}\mathbf{S_A}\mathbf{K}^T + \mathbf{S_O})^{-1}(\mathbf{y} - \mathbf{Kx_A}), \tag{2}$$

$$\hat{\mathbf{S}}^{-1} = \mathbf{K}^T\mathbf{S_O^{-1}}\mathbf{K} + \mathbf{S_A^{-1}}, \tag{3}$$

$$\mathbf{A} = \mathbf{I}_n - \hat{\mathbf{S}}\mathbf{S_A^{-1}}. \tag{4}$$

where $\mathbf{I}_n$ is the identity matrix with n being the dimension of the state vector $\mathbf{x}$. Inversions of atmospheric methane observations ~~usually~~ may solve $\nabla_{\mathbf{x}} J(\mathbf{x}) = \mathbf{0}$ either analytically, or numerically using an adjoint method ~~(Henze et al., 2007). The analytical solution has the advantage of providing complete~~ (Jacob et al., 2016). Unlike adjoint-based inversions, analytical solution provides direct error characterization of the optimal estimate $\hat{\mathbf{x}}$ through its error covariance matrix $\hat{\mathbf{S}}$. The related averaging kernel matrix $\mathbf{A}$ describes the sensitivity of the optimal estimate $\hat{\mathbf{x}}$ to the true emissions $\mathbf{x}$. The trace of $\mathbf{A}$ quantifies the Degrees of Freedom For Signal (DOFS), i.e., the number of pieces of information in the observing system for constraining the methane emissions (DOFS $\leq n$).

The Jacobian matrix $\mathbf{K}$ for the inversion is constructed with the GEOS-Chem CTM (http://www.geos-chem.org), which relates methane emissions to atmospheric concentrations through simulation of atmospheric transport. We use a nested version of GEOS-Chem as described by Kim et al. (2015) with $0.25° \times 0.3125°$ horizontal resolution over the North America window and adjacent oceans (9.75°-60°N, 130°-60°W), driven by GEOS-FP assimilated meteorological data from the NASA Global Modeling and Assimilation Office (GMAO). The same version of the GEOS-Chem has been applied to simulation of other chemical observations from the SEAC[4]RS campaign (Kim et al., 2015; Fisher et al., 2016; Marais et al., 2016; Travis et al., 2016; Zhu et al., 2016; Yu et al., 2016; Chan Miller et al., 2017). The boundary conditions for the nested-grid simulation are from a $4° \times 5°$ global simulation by Turner et al. (2015) using methane emissions optimized with three years of GOSAT satellite data. The ~~model~~ GOSAT-optimized emissions have been independently evaluated with atmospheric methane observations from the NOAA surface network (Turner et al., 2015). The GEOS-Chem uses a 3-D archive of monthly average OH concentrations from Park et al. (2004) to compute the methane sink, with a lifetime of 8.9 years in the troposphere consistent with observational constraints (Prather et al., 2012; Turner et al., 2017). ~~Loss by OH~~ The sink is irrelevant for our North American simulation since

ventilation of the domain is much faster(Wecht et al., 2014). Since we treat OH concentrations as decoupled from methane in the inversion, the relationship between emissions and concentrations is strictly linear, so that **K** fully describes the GEOS-Chem model ~~as applied to our problem~~for the purpose of the inversion.

The prior emission estimates for the inversion are taken from the $0.1° \times 0.1°$ gridded version of the EPA anthropogenic greenhouse gas emission inventory for 2012 (Maasakkers et al., 2016) and the mean wetland emissions from the $0.5° \times 0.5°$ monthly WetCHARTs extended ensemble for 2013 (Bloom et al., 2017). Figure 1 (top panels) shows the distribution of these prior methane emissions over the inversion domain for August-September 2013. Emissions total 13.3 Tg a^{-1} for anthropogenic sources and 13.0 Tg a^{-1} for wetlands over these two months (expressed on an annual basis). Anthropogenic emissions in the EPA inventory have little seasonal or interannual variability (Turner et al., 2015; Maasakkers et al., 2016), while wetland emissions have a large seasonal variation.

The SEAC[4]RS DC-8 aircraft conducted 21 flights over the Southeast between August 6 and September 21, 2013. Methane was measured by gas chromatography from whole air flask samples and calibrated to the NOAA standard. Figure 2 (left panel) shows the SEAC[4]RS flight tracks and the spatial distribution of the methane flask measurements below 2 km altitude~~averaged over the $0.25° \times 0.3125°$ model grid~~. The mean observed vertical profile is shown in the right panel of Figure 2, and compared to the GEOS-Chem profile using the prior emissions. The model is unbiased in the free troposphere above 2 km, implying a successful representation of background methane by the boundary conditions. Model overestimation in the boundary layer below 2 km suggests that the prior US emissions are too high. ~~In what follows we will~~ For the inversion, we use the SEAC[4]RS observations ~~over the Southeast US~~ below 2 km altitude ~~for the inversion. This represents a total data set of $m = 652$ methane observations~~ averaged over the $0.25° \times 0.3125°$ GEOS-Chem grid for individual flights. This represents 652 individual methane observations.

We use the residual error method (Heald et al., 2004) to estimate the diagonal elements of the observational error covariance matrix **S$_O$**. The method assumes that the mean model bias between the observations and the model ~~with prior emissions is~~ is due to error in prior emissions to be corrected by the inversion~~, while the residual error represents~~. The residuals are the differences between observed and modeled values after removing the mean model bias. The residual error standard deviation (RSD) is taken to represent the observational error including contributions from the instrument and the transport model. Figure 3 shows the vertical profile of the ~~residual error standard deviation (RSD)~~RSD for the ensemble of the SEAC[4]RS data over the Southeast US. The RSD is about 60 ppb below 2 km and 20 ppb in the free troposphere above. Subsetting the data by latitudinal bands gives similar results. We thus use 60 ppb for the standard deviation of the observational error (diagonal elements in **S$_O$**). The instrument precision is better than 2 ppb (Simpson et al., 2002), thus most of that observational error is from the transport model~~(including representation error)~~. We take **S$_O$** to be diagonal since error correlations between ~~boundary-layer~~ boundary layer observations on the GEOS-Chem grid are not significant (Wecht et al., 2014).

The inversion can in principle optimize emissions at the $0.25° \times 0.3125°$ grid resolution of the GEOS-Chem model, representing 3004 grid cells over the inversion domain. ~~However,~~ But the aircraft observations do not have sufficient information to constrain emissions at that resolution. In order to reduce the dimensionality of the state vector, we project the 3004 grid cells onto 216 elements of a Gaussian mixture model (GMM) with radial basis functions based on spatial proximity and source type

patterns (Turner and Jacob, 2015). The use of the GMM allows us to retain high resolution of up to 25 km for major localized sources while degrading resolution in areas of weak or broadly distributed sources. Areas dominated by wetlands have resolution of 100-200 km in the GMM because they are broadly distributed. ~~Individual state vector elements in the GMM have weighted influence functions over the $0.25° \times 0.3125°$ GEOS-Chem grid, so that the inversion effectively returns information on that fine grid even though the actual resolution of the inversion is coarser (Turner and Jacob, 2015).~~

The anthropogenic inventory of Maasakkers et al. (2016) and the wetlands inventory of Bloom et al. (2017) both include gridded error estimates that serve as the diagonal elements of the prior error covariance matrix $\mathbf{S_A}$. Maasakkers et al. (2016) found no significant spatial error correlation at $0.1° \times 0.1°$ resolution in their inventory while a variogram analysis across the elements of the WetCHARTS ensemble indicates a spatial error correlation length scale of 130 km. ~~Therefore we~~ Here we ignore this correlation and take $\mathbf{S_A}$ to be diagonal.

3 Results and discussion

Figure 1 (bottom panels) shows the results of the inversion including the optimized posterior emissions, the corrections to the prior emissions, and the DOFS as measured by the diagonal elements of the averaging kernel matrix. Figure 4 (top panels) compare the observed boundary layer methane concentrations to the values simulated by GEOS-Chem with prior and posterior emissions (Figure 1). The simulation with prior emissions has a positive bias that is effectively corrected when using posterior emissions. The coefficient of determination (R^2) between model and observations increases from 0.30 to 0.50 when using posterior emissions. Figure 4 also evaluates the SEAC^{4}RS inversion results with independent surface air observations from the three NOAA/ESRL surface network sites in the region (Andrews et al., 2014) and with methane column observations ~~to~~ from the TCCON site in Lamont, Oklahoma (Wunch et al., 2011; Wennberg et al., 2017). The posterior emissions improve the simulation of these independent data sets. GOSAT satellite observations are another source of independent data but the 2-month period is too sparse for useful evaluation (Wecht et al., 2014).

Total posterior emissions over the SEAC^{4}RS domain are 15% (4 Tg a^{-1}) lower than the prior estimate (Figure 1). The inversion is able to constrain about 10 pieces of information in the spatial distribution of methane emissions as measured by the DOFS. It is strongly sensitive to the Gulf Coast and to large anthropogenic source areas such as the Floyd Shale in central Alabama. For the regions with averaging kernel sensitivity larger than 0.05, posterior emissions are 35% lower than the prior estimate. The posterior errors are 18%-30% over these regions. The scaling factors show large downward corrections of prior emissions in Louisiana and Mississippi, and along the Gulf Coast, where wetlands are the dominant sources. There are also downward corrections in southern West Virginia, where coal mines are dominant, and in the Haynesville Shale gas production region of northern Louisiana and southern Arkansas. On the other hand, there are significant upward corrections for the coal mines of southern Illinois and for the Floyd Shale in central Alabama.

We can attribute the $0.25° \times 0.3125°$ scaling factors from the inversion to specific methane source sectors by using the sector-resolved spatial patterns in the prior emission inventories, as described by Turner et al. (2015) but here with the improved anthropogenic source patterns from Maasakkers et al. (2016) and wetland emissions from (Bloom et al., 2017). Anthropogenic

and wetland sources are well separated spatially in these inventories. Figure 5 compares our results with the prior emission totals for the different sectors in the Southeast US. We find a significant 27% ~~(3.6 Tg a^{-1})~~ reduction in regional wetland emissions relative to the prior estimate (mean of the WetCHARTs extended ensemble). For the subdomain with averaging kernel sensitivity larger than 0.05 that reduction is 42%. By contrast, we find no significant regional bias in the EPA anthropogenic inventory for any of the major source sectors ~~.~~ for the period of August-September 2013. However, there are large local biases that tend to cancel each other on a regional scale (e.g., Haynesville Shale vs. Floyd Shale for natural gas). This suggests that methane emission factors for the oil/gas sector are more variable than assumed in the EPA (2016) inventory.

The WetCHARTs extended ensemble includes 18 wetland methane emission models intended to encompass the uncertainties in estimating wetland emissions (Bloom et al., 2017). The different models (ensemble members) use different datasets for wetland extent fraction A [m^2 wetlands per m^2 surface area], heterotrophic respiration rate R [mg C day^{-1} per m^2 of wetland ~~areas~~area], temperature-dependent factor $q_{10}^{T/10}$ of C respired as CH4 [mg CH$_4$ per mg C] where T is the surface skin temperature, and global scaling factors s. The wetland methane emission flux E [mg CH$_4$ m^{-2} day^{-1}] at a time t and location x for each of these members is given by

$$E(t,x) = sA(t,x)R(t,x)q_{10}^{\underline{T(x,t)}T(t,x)/10}. \tag{5}$$

The 18-member ensemble consists of three temperature dependence factors ($q_{10} = 1, 2, 3$), three global scale factors ($s = 125, 166, 208$), and two wetland extent maps (A) from the Global Lakes and Wetlands Database (GLWD; Lehner and Dölla, 2004) and GLOBCOVER (Bontemps et al., 2011). The heterotrophic respiration rate (R) is the median output from the carbon data model framework (CARDAMOM; Bloom et al., 2016), and is not varied across that ensemble.

Figure 6 shows the Southeast US wetland emissions for each WetCHARTs member, along with the root-mean-square error (RMSE) of its spatial distribution relative to our optimized posterior estimate on the $0.25° \times 0.3125°$ grid. Consistency in spatial distribution with our optimized estimate is indicated by a low RMSE. We find that the specification of wetland extent is the most systematic source of error in wetland emission estimates; all GLOBCOVER-based models underestimate wetland emissions, while all GLWD-based models overestimate emissions. Estimates using $q_{10} = 1$ (no temperature dependence in the CH4:C respiration ratio) exhibit the lowest RMSE values. The WetCHARTs ensemble mean used as prior for our inversion ~~performs better than any individual member.~~

~~For anthropogenic emissions, Figure 5 shows that the inversion is consistent on the regional scale with the EPA sectoral inventory gridded by Maasakkers et al. (2016). Previous inversions using the EDGAR v4.2 inventory as prior found large underestimates over the Southcentral US that they attributed to a combination of oil/gas and livestock sources (Miller et al., 2013; Alexe et a This is in contrast with our finding in particular for East Texas and Louisiana. The EDGAR v4.2 inventory has large errors in its source patterns (see Fig. 3 of Maasakkers et al. (2016)). It places almost all oil/gas emissions in distribution centers instead of in production fields, and the resulting methane concentration underestimate over production fields (e.g., Floyd shale in East Texas) would be interpreted as a model error. These previous inverse studies also used low prior estimates of wetland emissions (2.7-5.9 Tg a^{-1} for the contiguous US, 1.6-3.5 Tg a^{-1} for the Southeast US, at the low end of the WetCHARTs ensemble in Figure 5), leading them to erroneously attribute methane underestimates to nearby anthropogenic sources.~~

~~Despite the good regional agreement of our inversion with the EPA (2016) inventory for the Southeast US for different sectors, there are large local biases that tend to cancel each other on a regional scale (e.g., Haynesville Shale vs. Floyd Shale for natural gas). This suggests that methane emission factors for the oil/gas sector are more variable than assumed in the EPA (2016) inventory~~has the lowest RMSE, although this may reflect its influence on the posterior solution.

4 Conclusions

We used extensive boundary layer methane observations from the SEAC[4]RS aircraft campaign over the Southeast US in August-September 2013 to optimize methane emissions in that region with up to $0.25° \times 0.3125°$ spatial resolution and with detailed error characterization. The inversion used new ~~state-of-science~~ state-of-the-art inventories as prior information, including the gridded version of the EPA (2016) national anthropogenic inventory from Maasakkers et al. (2016) and the WetCHARTs wetlands extended ensemble from Bloom et al. (2017). The inversion domain over the Southeast US accounts for 45% of national methane emissions in the EPA inventory, and for 56% of wetland emissions over the contiguous US in the mean WetCHARTs estimate.

Our inversion results suggest that the EPA emission inventory has no significant bias on the regional scale for the major anthropogenic source sectors (livestock, oil/gas, waste, coal), while the mean of the WetCHARTs wetland ensemble needs to be reduced by 27% over the inversion domain. These results are supported by independent methane observations from the NOAA/ESRL surface network and from the TCCON site in Lamont, Oklahoma. The specification of wetland areal extent is the dominant source of error in the WetCHARTs ensemble. Results also indicate that a low temperature dependence for the CH4:C heterotrophic respiration ratio best explains the spatial variability of the posterior emissions. ~~The mean of the WetCHARTs ensemble performs better than any individual ensemble member. Our finding of regional consistency~~ Despite regional agreement with the EPA anthropogenic inventory~~is in contrast with previous inverse studies that found large underestimates. These inversions relied on EDGAR v4.2 anthropogenic source patterns that have large errors and also assumed low wetland emissions. Despite regional agreement~~, we still find significant local discrepancies with the EPA inventory for the oil/gas sector, suggesting that methane emission factors are more variable than assumed in the inventory.
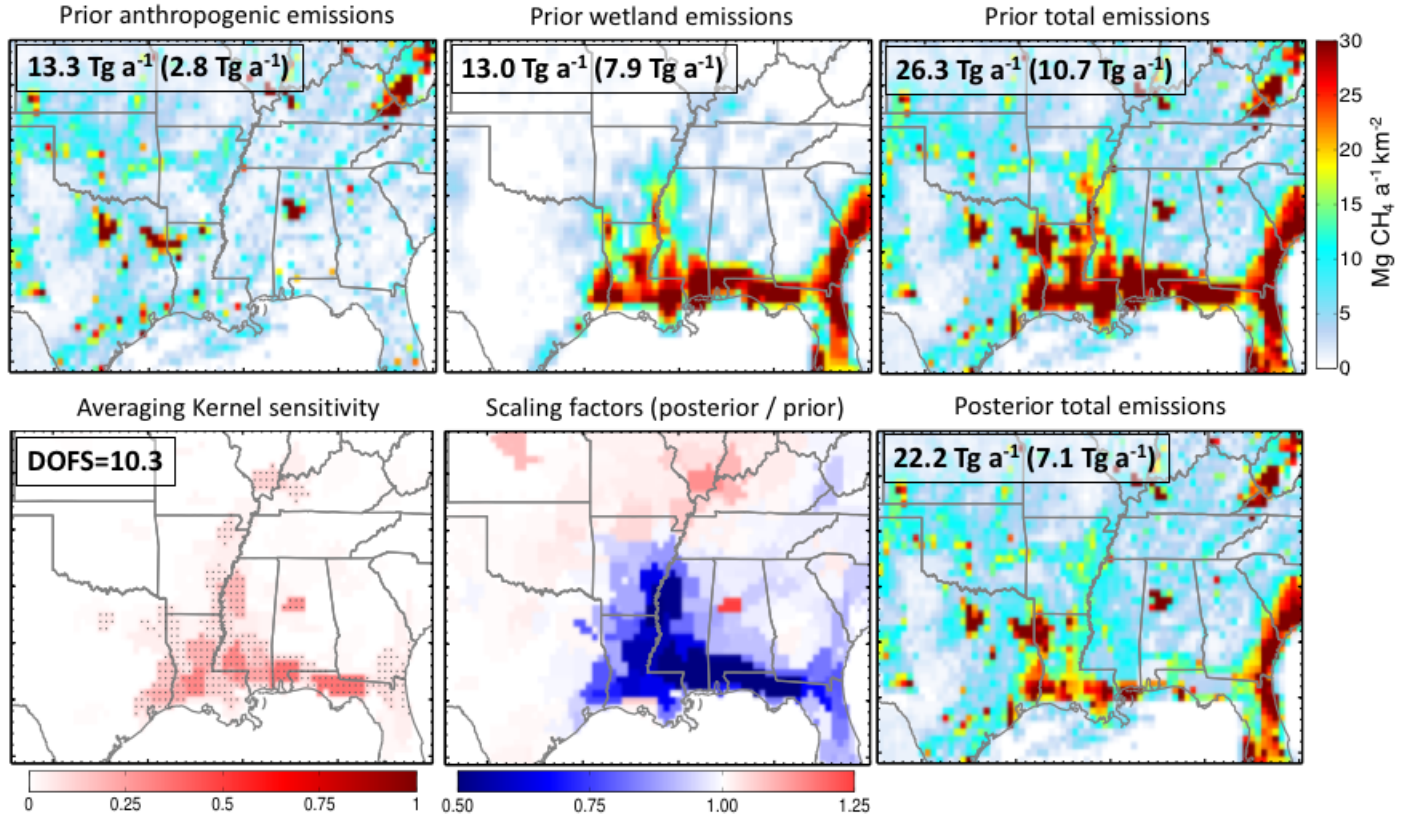
Figure 1. Methane emissions in the Southeast US in August-September 2013. The top panels show the prior anthropogenic and wetland methane emissions, and the bottom panels show the inversion results including posterior emissions, scaling factors (posterior/prior emission ratios), and the diagonal elements of the averaging kernel matrix ~~for~~ representing the sensitivity of the inversion results to the observations. The sum of these diagonal elements over the domain (trace of the averaging kernel matrix) quantifies the degrees of freedom for signal (DOFS) of the inversion. ~~Annual~~ Numbers inset in the emission panels are the regional totals expressed as annual means for clarity (i.e., assuming that August-September emission rates hold for the ~~SEAC^{4}RS period~~ rest of the year). Values in parentheses are ~~shown inset~~ the totals for the region with averaging kernel sensitivities larger than 0.05 (stippled areas in lower left panel).
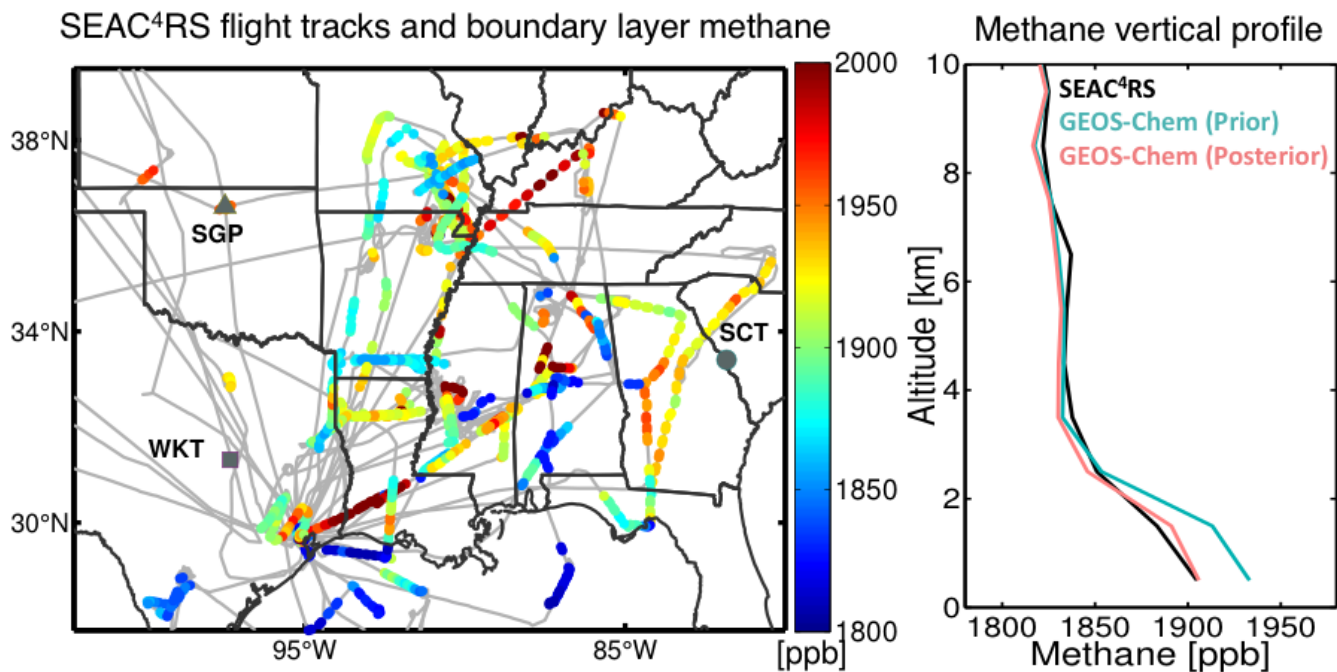
Figure 2. Boundary layer methane concentrations over the Southeast US measured during the SEAC[4]RS aircraft campaign (August ~~6-September~~ 6-September 21, 2013). The left panel shows the flight tracks in grey and the methane measurements at 0-2 km altitude~~averaged over the 0.25° × 0.3125° GEOS-Chem grid~~. The three NOAA/ESRL sites at SGP (Southern Great Plains, Oklahoma; 36.6°N, 97.5°W) , WKT (Moody, Texas; 31.3°N, 97.3°W), and SCT (Beech Island, South Carolina; 33.4°, 81.8°W) are indicated. SGP is co-located with the TCCON site at Lamont, Oklahoma. The right panel shows the mean methane vertical profiles over the Southeast US domain measured from the aircraft and simulated by GEOS-Chem using the prior and posterior emissions.
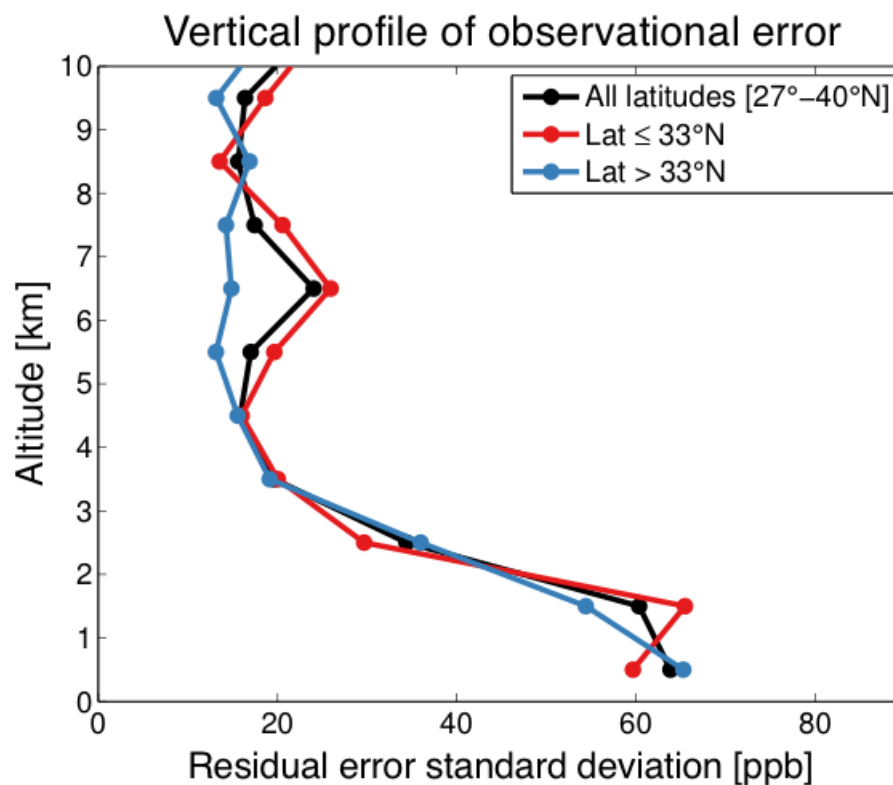
Figure 3. Residual standard ~~deviations~~ deviation (~~RSDs~~RSD) of the difference between SEAC[4]RS methane observations and the GEOS-Chem model with prior emissions, for 1-km altitude bins. ~~These RSDs are used to define~~ The RSD defines the observational error standard deviations for the inversion as described in the text. ~~The observational error is mainly from GEOS-Chem (see text).~~ Values are shown for two latitudinal ~~ranges~~bands.
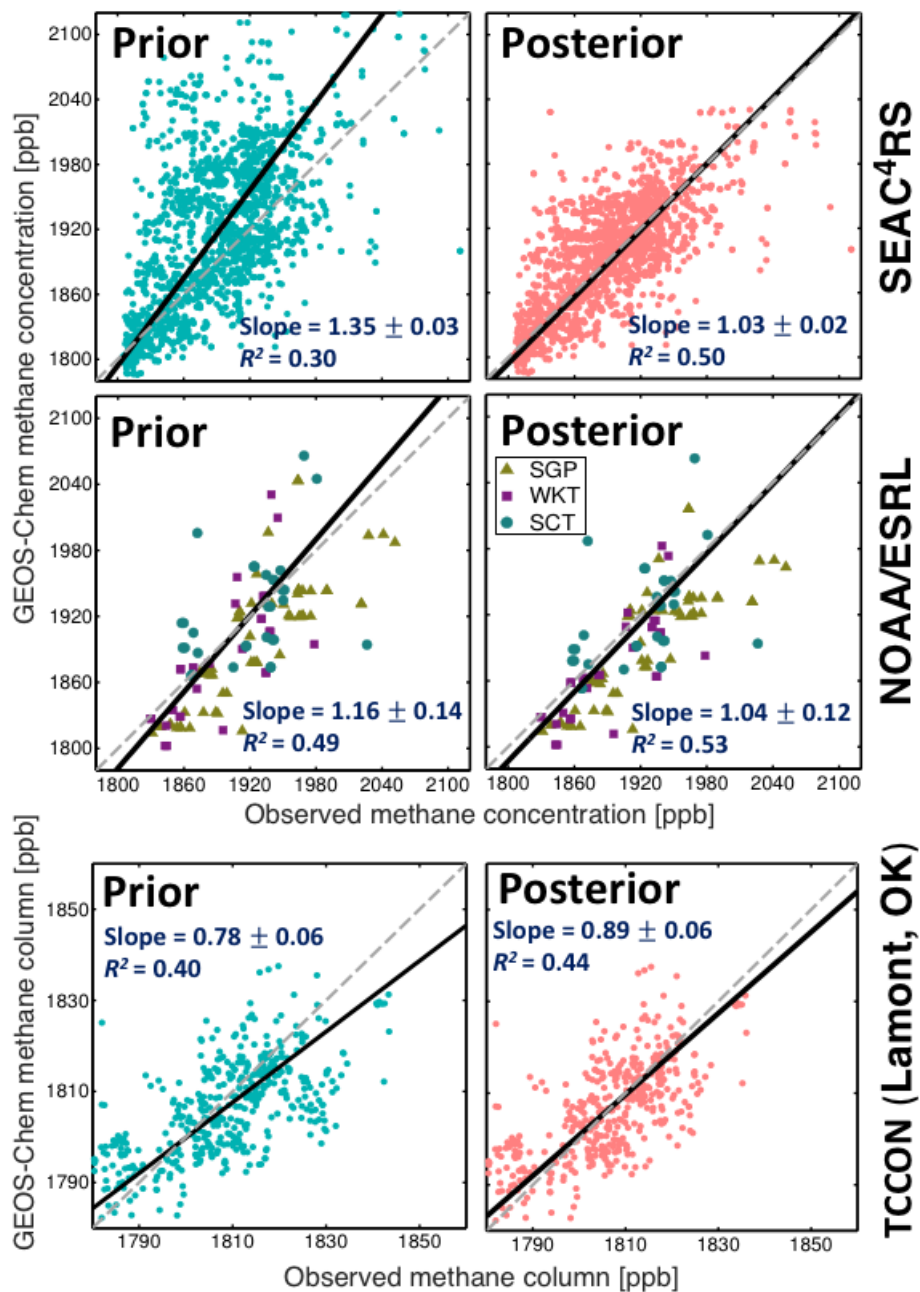
Figure 4. Evaluation of the SEAC⁴RS inversion of methane emissions in the Southeast US for the August 6 - September 21, 2013 period. The top panels compare GEOS-Chem methane concentrations with the SEAC⁴RS observations, using prior emissions (left) and posterior emissions (right). The middle panels compare GEOS-Chem methane concentrations with independent observations from the three NOAA/ESRL surface sites in the inversion domain (see Fig. 2 and caption). The bottom panels compare GEOS-Chem methane columns with TCCON hourly column observations at Lamont, Oklahoma (Wennberg et al., 2017), after correction for stratospheric bias in the model (Turner et al., 2015). The 1:1 lines (dashed) and the reduced-major-axis (black solid line) linear regressions are also shown, along with the coefficients of determination (R^2) and the slopes ($\pm 1\sigma$) derived from the bootstrap method.
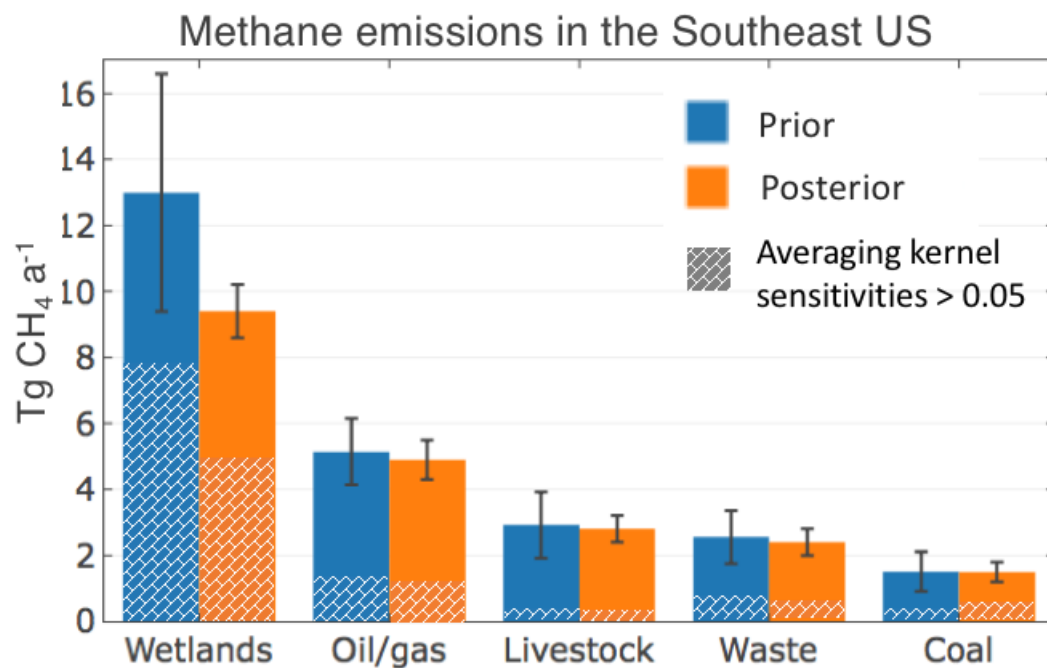
Figure 5. Prior and posterior methane emissions ~~for~~ in the Southeast US (domain of Fig. ~~1.~~ 1) for August-September 2013. The prior anthropogenic emissions are from the EPA national inventory for 2012 (EPA, 2016; Maasakkers et al., 2016) and the prior wetland emissions are the means of the WetCHARTs extended ensemble (Bloom et al., 2017). Error bars (one standard deviation) on sectoral emissions are from the prior and posterior error variances of our inversion. Methane emissions in the subdomain with averaging kernel sensitivities larger than 0.05 (Figure 1) are also indicated.
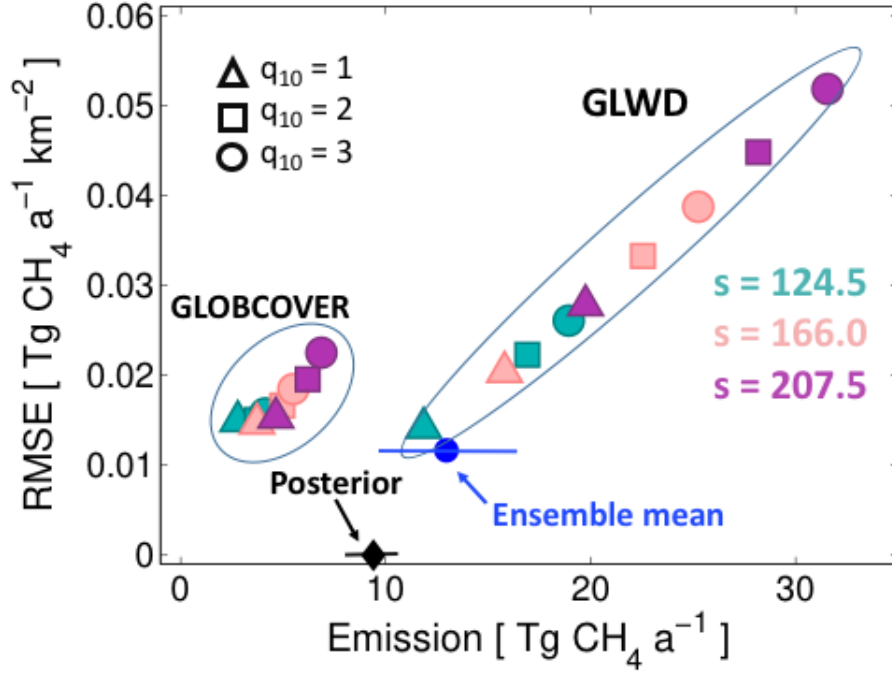
Figure 6. Range of wetland emission estimates ~~for~~ in the Southeast US (domain of Figure 1) ~~.~~ for August-September 2013. The figure shows the spread of the WetCHARTs extended ensemble and compares with the posterior emission estimate from our inversion in terms of emission total and root-mean-square error (RMSE) on the $0.25° \times 0.3125°$ spatial grid. The RMSE of the posterior emission estimate with itself is zero by definition. WetCHARTs ensemble members use wetland areal extent data from either the GLOBCOVER (Bontemps et al., 2011) or GLWD (Lehner and Dölla, 2004) databases, as well as different estimates of temperature sensitivity q_{10} and global scaling factors s (see equation (5) and text). The posterior wetland emission estimate from our inversion is shown as ~~dashed line~~ black solid diamond with its associated error standard deviation~~shaded~~. The mean of the WetCHARTs ensemble used as prior for our inversion is shown as blue solid circle with its associated error standard deviation.

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