

Author Response to Review of Referee #3 for “Seasonal Variability of Stratospheric Methane: Implications for Constraining Tropospheric Methane Budgets Using Total Column Observations”

K. M. Saad¹, D. Wunch^{1,2}, N. M. Deutscher^{3,4}, D. W. T. Griffith³, F. Hase⁵, M. De Mazière⁶, J. Notholt⁴, D. F. Pollard⁷, C. M. Roehl¹, M. Schneider⁵, R. Sussmann⁸, T. Warneke⁴, and P. O. Wennberg¹

¹California Institute of Technology, Pasadena, California, USA

²University of Toronto, Toronto, Ontario, Canada

³University of Wollongong, Wollongong, NSW, Australia

⁴University of Bremen, Bremen, Germany

⁵Karlsruhe Institute of Technology, IMK-ASF, Karlsruhe, Germany

⁶Royal Belgian Institute for Space Aeronomy, Brussels, Belgium

⁷National Institute of Water and Atmospheric Research, Omakau, New Zealand

⁸Karlsruhe Institute of Technology, IMK-IFU, Garmisch-Partenkirchen, Germany

Correspondence to: K. M. Saad (katsaad@caltech.edu)

We thank Referee #3 for their comments.

The primary change in the updated manuscript is a reprocessing of the TCCON tropospheric methane (CH₄) column-averaged dry-air mole fractions (DMFs), which is described in detail in added supplement, “Updates to Tropospheric Methane Data” (Appendix A). Although some of the regression statistics and comparisons have changed as a result of measurement updates, the main conclusions, the mismatch in tropospheric seasonality and the dependence of the stratospheric contribution error on tropopause height, remain the same.

In our responses below, page and line numbers included refer to the previous discussion draft. Appendices are referred to based on their order in the revised manuscript, and their headings are noted to avoid ambiguity.

10 **My first major concern is that the fluxes used for the "Base" case do not actually match the total column TCCON measurements all that well. This can be seen somewhat by the top row of scatter plots in Figure 4. The correlation between the the total column simulated by GEOS-Chem has a correlation with the TCCON measurements of 0.86, which is even a bit lower than the correlation of the tropospheric columns, which are arguably more relevant for flux inversions. But more worrying, in Figure 7 it can be seen that the seasonality of the total column across the TCCON**
15 **northern hemisphere sites considered is completely wrong. This inability to capture the seasonal cycle in the total column means that only limited conclusions can be drawn from assessing the (slightly different) mismatch in the two parts of the column. Thus I think the main weakness of this paper is the choice of fluxes used for the forward simulation.**

We chose to use the default emissions provided for the GEOS-Chem offline CH₄ simulation to demonstrate how systematic errors in the vertical profile of CH₄ (which are caused by parameters that do not vary interannually, namely OH fields and transport schemes) can alias into the optimized emissions resulting from an assimilation of total column measurements into an
20

atmospheric inversion. This analysis is a sensitivity study on how model biases can alias into emissions optimization. Thus, the choice of emissions would not drive results unless those emissions are somehow causing the systematic biases. The aseasonal simulation was set up as an experiment to determine if the seasonality of emissions was causing the tropospheric phase lag observed in the base simulation. As Fig. 7 illustrates, the seasonal phase was consistent between simulations even as the amplitude changed, which demonstrates that the chosen emissions fields do not drive the main result of this analysis.

These fluxes are only listed in terms of categories, with no itemization of which anthropogenic inventory (I guess EDGAR4.X?), which "other natural emissions", or which model was used for the very important seasonal wetland and rice fluxes. At very least this needs to be amended and clarified.

References were cited for the "default" offline CH₄ simulation, which included a description of these fluxes. We have since added details and references for each of the emissions categories have been added for the reader's convenience.

It's fine that the fluxes are added to the model at 60 second increments, but I guess that aside from fires and wetlands/rice the fluxes are constant throughout the year? Or did you employ a diurnal or weekly or annual cycle?

The list of emissions, which were grouped by time evolution (annual, monthly, and daily), now includes additional details that should make the time scales of their variability more apparent to the reader.

And what about the OH fields? Is there a reference for where these came from? Have they been optimized via methyl chloroform or similar?

Optimized OH fields were not available for GEOS-Chem, which led to the OH sensitivity experiments to test the dependency of CH₄ DMFs on the magnitude, seasonality, and distribution of tropospheric OH. These experiments are described in Appendix B1, "Equilibrium Sensitivity Experiments". The Northern to Southern Hemisphere ratio of 1.0 (monthly range of 0.975 – 1.02, applying a six month lag in the Southern Hemisphere) is consistent with the ratio of 0.97 ± 0.12 found by Patra et al. (2014). The tropospheric OH are monthly-averaged output from a GEOS-Chem tropospheric chemistry simulation (Park et al., 2004). The description of tropospheric OH and stratospheric loss parameterization fields now include references.

What would have been a more relevant choice for this type of study would be to use optimized fluxes, resulting from an atmospheric inversion using the same model. There are a few groups working on methane inversions with GEOS-Chem, so such fluxes should not have been difficult to find through collaboration. Then you would have been able to start with a seasonal cycle in the column that is actually consistent at the TCCON sites, assuming that the TCCON sites were assimilated in the inversion. This would have made the analysis more relevant, and it would be my strongest recommendation for improving this study.

As you note, most of the recent optimized emissions that result from atmospheric inversions, especially those using GEOS-Chem as the forward model, assimilate TCCON total column measurements. Using these fluxes would make the measurement to model comparisons, and thus their correlations, no longer independent, and the statistics would be less meaningful.

Moreover, using optimized fluxes may not improve the seasonality of the mismatch. Fraser et al. (2011) compared TCCON total columns to GEOS-Chem run with posterior fluxes, which were derived from an inversion using GOSAT total columns and surface measurements, and found a seasonally-varying measurement-model mismatch that fell between ± 20 ppb (Fig. 6 of that paper). We agree that work that compares optimized fluxes from atmospheric inversions that assimilate data at various vertical levels would be very informative, and this approach would be an important next step.

Another concern related to the choice of fluxes relates to the method used for the aseasonal simulation. The manuscript describes that the seasonal fluxes (fires, wetlands, and rice) were "disabled" (I assume this means set to zero?), and then the other fluxes were scaled up to maintain the fluxes and the approximate (but certainly not exact, as shown in Figure 11) latitudinal distribution. Why not simply use an annual mean of the variable fluxes? Then you are not changing two things at once (geographic distribution and temporal variability) and attempting to attribute the changes to only one of the factors.

We agree that producing aseasonal emissions by changing the seasonally varying fluxes to be constant throughout each year for each grid box would be ideal. Unfortunately, the model infrastructure made such a simulation difficult to execute as it required the emissions code to be re-written, risking differences due to compiling changes. Thus the scaling technique was developed as an alternative to assess first-order impacts of emissions seasonality. We have added this explicitly as a limitation that should be improved on in the future. However, most of the notable results, especially the phase lag in the tropospheric seasonality, are consistent between the model runs despite any differences in the spatial distribution of emissions. This demonstrates the robustness of our conclusions regardless of the emissions fields used. Additionally, the analyses comparing the base and aseasonal simulations are aggregated on zonal or hemispheric scales and therefore should not vary because of the spatial differences of their emissions at smaller scales.

My next major concern is related to the numerics of how the stratospheric and tropospheric model columns are divided. I do not understand how the the stratospheric column-integrated dry air mole fractions have values around 30-100 ppb (from Figure 4). This seems very, very low. Looking at the prior profiles from Wunch et al. (2011), Figure 2, the stratospheric values of CH₄ range from 500-1800 ppm. I am not sure if this can be explained by the weighting with the pressure-weighted averaging kernel, as the methane column averaging kernel is actually rather flat (from Figure 4, Wunch et al., 2011). Also from Figure 1 of Saad et al. (2014), the only mixing ratios of stratospheric methane less than even 500 ppb seem to be over 40 km or so, which is far above even the highest tropopause. I had postulated that perhaps you had calculated the mixing ratio not in parts per million molecules of stratospheric air but rather of total column air (in which case it should have been explained). Although I would not advocate for such an approach, in that case the stratospheric partial column dry air mole fraction could be added directly to the tropospheric dry air mole fraction to get the total column dry air mole fraction. Looking again at Figure 4, this is clearly not the case: the tropospheric column is clearly larger than the total column. This needs to be clarified.

You correctly postulated that the stratospheric contribution is calculated in reference to the total column of air. This was done for both practical and conceptual reasons. TCCON X_{CH_4} and $X_{\text{CH}_4}^t$ are processed to remove various spectroscopic biases and calibrated to in situ aircraft profiles, now described in Appendix A, “Updates to Tropospheric Methane Data.” Thus, using these column-averaged DMFs instead of the CH_4 columns in our proxy for stratospheric air ensures measurement biases are not the cause of any measurement-model mismatch.

Conceptually, because this paper focuses on how the model’s stratospheric contribution to the total column can alter the conclusions made about tropospheric trends, we determined that stratospheric CH_4 over the total column of air would be more relevant than the stratospheric partial column of CH_4 . We agree that if the purpose of this work was to assess modeled stratospheric profiles, the stratospheric partial column would be more appropriate. Because the stratosphere has less CH_4 , the stratospheric contribution depresses the total column value, so the tropospheric column average should be larger. We frame the stratospheric contribution as positive number to make the value more intuitive: a larger stratospheric contribution indicates the influence of the stratosphere on the total column is greater. The stratospheric contribution is also represented as a positive number for visual clarity; applying a sign change to the stratospheric contribution in Fig. 4 and adding it to $X_{\text{CH}_4}^t$ does reproduce X_{CH_4} .

We have updated the wording of the definition of the stratospheric contribution on p.5 l.21 to remove the ambiguity of how the stratospheric contribution is calculated. Additionally, we have added an appendix with the derivation of the stratospheric contribution, “Derivation of Stratospheric Contribution” (Appendix C). We have also changed usage of “stratosphere” to “stratospheric contribution” throughout the text for contexts in which the ambiguity could be confusing.

One other concern was the consistency of the model tropopause with that from the TCCON retrievals. You mention testing the impact of moving the tropopause model layer up one or two levels, but this does not allow for potential seasonal or regional variability in the match between the two. At very least the (latitude- and seasonal-dependent) correlation between the model and retrieval tropopause heights should be presented in some way.

Accurately representing GEOS-Chem’s tropospheric column for the purpose of comparison to measurements depends on setting the tropopause so that the calculation from model output is consistent with the way the model defines the troposphere. Shifting the tropopause level allowed us to test the degree to which calculating $X_{\text{CH}_4}^t$ using the daily average tropopause could bias the comparison. Because the vertical gradient of CH_4 is steepest across the UTLS, choosing a lower tropopause level would change the vertical integration much less than choosing a higher level. Thus, integrating to higher pressure levels would provide a better measure of sensitivity to the integration tropopause height chosen.

Additionally, GEOS-Chem sets the top of the troposphere one level below the vertical pressure level below the tropopause, which we thought could also introduce a bias. We ran a simulation setting the top of the troposphere at the level in which the tropopause exists (now listed in Appendix B1, “Equilibrium Sensitivity Experiments”), essentially shifting the tropopause up two levels, to determine if the choice of the definition of the tropopause changed the distribution of CH_4 concentrations. This change did not improve measurement-model agreement and, as the newly added figure demonstrates, had almost no impact on the seasonality of $X_{\text{CH}_4}^t$ (Fig. 15 in the updated manuscript).

We consider other inconsistencies in the model tropopause, such as seasonal or zonal variability, as one of the model errors that can alias into X_{CH_4} comparisons. The calibration of TCCON measurements to in situ aircraft profiles (Wunch et al., 2015) limits any bias that errors in the TCCON tropopause heights could induce in the comparisons with the model. Moreover, the difference between calibrated TCCON and integrated aircraft X_{CH_4} and $X_{\text{CH}_4}^t$ values have no correlation to the tropopause heights used to generate the TCCON priors or computed from the aircraft temperature and pressure profiles (uncertainty-weighted $R^2 = 0$).

P3, second paragraph: This sounds like you're describing atmospheric inversion while going out of your way not to call it "inversion". Or are you referring to optimization only by processed-based scaling of set spatial fields? Please clarify, and if you mean inversion, please say so.

We infer that the referee meant p.2 second paragraph and have added the term “atmospheric inversion” for clarity.

P2, L10: The reference to Stephens et al. (2007) here seems not to fit so well - this study was looking at aircraft profiles rather than column-integrated information.

The reference of Stephens et al. (2007) was included to illustrate the importance of assimilating observations that provide information about the vertical profile to accurately constrain chemical transport models. We agree that p.2 1.10 is not the appropriate location for this point and have moved the reference to the paragraph on p.2 1.22.

P4, L14: Although I mentioned it already above, there needs to be some citations to describe the model and fluxes used.

Citations and additional details for the GEOS-Chem offline CH_4 emissions, tropospheric OH, and stratospheric loss fields have been added.

P5, L9-10: In Appendix A1 I couldn't find any real description of the OH sensitivity runs. Do your OH fields have seasonality? This experiment is insufficiently described.

We have added the following description of the “Updated OH” simulation (Table 2, Fig. 3) to Appendix B1, “Equilibrium Sensitivity Experiments”:

“The updated OH simulation used OH output from a 2012 GEOS-Chem standard chemistry simulation with extensive updates to the photochemical oxidation mechanisms of biogenic volatile organic compounds (VOCs), described in Bates et al. (2016) and references therein. These were converted to 3D monthly mean OH concentrations to conform to the infrastructure of the GEOS-Chem offline CH_4 tropospheric loss mechanism. The OH was then scaled by 90% to keep the lifetime above 8 years, and emissions were scaled by 112% to maintain the same balance between sources and sinks in the base simulation. Figure 14 provides zonal averages of the difference between the base and updated OH columns.”

We also ran several sensitivity experiments on different OH fields, which included scaling the default OH fields and using different scalings of the “Standard Chemistry + Biogenic VOCs” OH output. A table delineating these simulations has now been added to Appendix B1.

5 **P5, L15: I was a bit confused here: are the means and medians for all values over the day, over just over those where TCCON measurements were made?**

GEOS-Chem smoothed column-averaged DMFs were only calculated for days in which TCCON measurements were made and were smoothed using solar zenith angles, vertical scaling factors, and surface pressures for TCCON measurements used in the comparisons. The added discussion of the TCCON retrieval in Section 2.1 provides a description of the vertical scaling factor that clarifies subsequent references. To further lessen confusion, this sentence has been changed to, “For comparisons with column measurements, model vertical profiles were smoothed with corresponding TCCON CH₄ averaging kernels, interpolated for the daily mean solar zenith angles, and prior profiles, scaled with daily median vertical scaling factors and interpolated to the daily mean surface pressures measured at each site, following the methodology in Rodgers and Connor (2003) and Wunch et al. (2010).”

15

In general I found the use of "DMF" to mean "column-integrated dry air mole fraction" to be rather confusing. Flask measurements also measure dry air mole fraction, so DMF on its own does not tell the reader that an integrated column is being discussed. This is found throughout the manuscript and should be clarified.

The modifier “column-average” now precedes “DMF” unless referring to a surface or profile measurement to maintain consistency and avoid ambiguity.

20

Figure 3: The caption says that the stratosphere shows a seasonal cycle of 15 ppb at Park Falls, but in the figure looks like more like 30 ppb. Please explain. I was also surprised to see that Park Falls appears to have a larger seasonal cycle in the stratosphere than in the troposphere for the Base case. This doesn't make sense to me. Please explain.

The text cites a seasonal amplitude of 15 ppb, referring to the peak amplitude of the seasonal cycle (i.e. the difference between the peak and the mean). The peak-to-trough amplitude, which is twice the peak amplitude, would indeed be 30 ppb. We have changed the word “amplitude” to “range” and updated the values accordingly to reduce confusion.

The model's larger seasonal cycle of the stratospheric contribution compared to that of the tropospheric column does not agree with the measurements, as illustrated by Fig. 7. The stratospheric contribution is a function of the gradient across the tropopause and CH₄ loss in the stratosphere (Appendix C, “Derivation of Stratospheric Contribution”); thus, model errors in prescribed tropopause height, stratospheric chemistry, and stratospheric transport will impact the seasonal cycle of the stratospheric contribution.

P7, L13-14: What about the significant figures on the slopes (e.g. 1.1±0.020).

The extra significant figures on the slope errors were unintended and have been removed.

35

P8, L5-7: I'm not sure that Figure 4 shows a good agreement between the stratospheric columns of TCCON and GEOS-Chem. Yes, the clump of points is closer to the 1:1 line, but it hardly forms a line at all. Is the correlation coefficient for this one station really notably higher?

5 The wording indicating good agreement has been changed to, "fall most closely to the one-to-one line." The spread across the one-to-one line seen at Lauder is partly due to seasonal variability, as the stratospheric loss parameterization in the model is monthly. Averaging GEOS-Chem daily values to correspond to the ACE-FTS and GEOS-Chem climatologies would make the relationship more compact.

10 **Figure 5: Again I'm confused about the calculation of the stratopsheric column. For instance, we can see from Figure 4 that the stratospheric column simulated by GEOS-Chem is around 50 ppb. Then looking at Figure 5, ACE-FTS minus GEOS-Chem seems to show a difference of approximately -50 ppb around 45 degrees south. Does this mean that the ACE-FTS measurements are showing close to zero methane? In general there seems to be better agreement between TCCON and GEOS-Chem (Figure 4) than ACE-FTS and GEOS-Chem (Figure 5), but it is difficult to tell from the**
15 **figures presented. Could you comment on this? How do ACE-FTS and TCCON agree?**

Figure 5 illustrates differences between the CH₄ profiles given by ACE-FTS and GEOS-Chem climatologies, not pressure-weighted column averages as in the TCCON comparison. As a point of reference, they correspond to the prior profiles from Wunch et al. (2011) you mentioned in previous comments. Thus, the ±150 ppb range appertains to the difference of the mean CH₄ mole fractions at each pressure level. The ACE-FTS climatology used in Fig. 5 is an older version of the measurements
20 (v. 2.2, Jones et al., 2012), which could impact some of individual grid box differences. However, a comparison to the v.3.5 (which are used in the $X_{\text{CH}_4}^t$ calculation) monthly mean CH₄ DMFs indicate that the data version likely would not change main features illustrated in Fig. 5.

Because the comparisons between TCCON and GEOS-Chem are for pressure-weighted column averages, the agreement is therefore not directly comparable to ACE-FTS mole fraction differences at individual pressure levels. Agreement between
25 TCCON and ACE-FTS is difficult to quantify because ACE-FTS retrievals provide vertical information solely in the upper atmosphere, and TCCON retrievals provide column averages that, due to the pressure weighting, are dominated by the troposphere. However, ACE-FTS is one of the various platforms used in the development of the empirical model that generates TCCON priors (Wunch et al., 2015), and the stratospheric CH₄ profiles it measures are used in the calculation of the TCCON tropospheric CH₄ product (Saad et al., 2014).

30

Figure 7: I am very surprised to see that the aseasonal simulations have higher seasonal cycles in both the stratosphere and the stratosphere than the base case. Are you sure of this result? What role does the (potential) seasonality of the OH sink have here?

The larger seasonal amplitude of the aseasonal $X_{\text{CH}_4}^t$ is indeed a notable result. The greatest differences, from August
35 through October, result from dampening the large summer wetland fluxes that balance high summer OH concentrations in the

base simulation. The larger variance across sites that we note is also indicative that the seasonal amplitude does not increase as drastically at the sub-tropical sites. (We did not include the figure with all site seasonalities because it was visually chaotic, given the many Northern Hemisphere sites.)

The second largest difference, during the spring, could also be a result of the source/sink balance: the aseasonal simulation introduces fluxes in the winter, when the OH concentrations are lowest. As we mention on p.10 1.18, the model may also have an error in phase with the seasonal emissions that produces the reasonable seasonal cycle amplitude in the base simulation troposphere (Fig. 7). We have added to that paragraph a discussion of the interaction between emissions and OH loss.

While the seasonal amplitude of the mean Northern Hemisphere stratospheric contribution is larger for the aseasonal versus base simulation, the maximum difference of their means is only about 2 ppb, which is within the 1σ standard deviations across sites. This similarity further demonstrates the insensitivity of the model's stratosphere to chosen emissions.

And what about the sampling throughout the year? Are there enough measurements at Bremen in December and January, or is part of this seasonality a question of shifting sampling throughout the year? Related to this: I assume you are only considering days on which there are TCCON measurements in the model analysis?

The seasonality of GEOS-Chem is computed from the smoothed pressure-weighted column-averaged DMFs, which incorporate the TCCON scaled prior profiles (see Appendix B3, "Model Smoothing for Measurement Comparisons") and thus require us to consider days on which TCCON measurements exist. While the number of measurements per month is variable throughout the year, all high latitude sites have a time series long enough to extract detrended monthly mean information. Moreover, the sites that are most susceptible to low winter sampling are the five in Europe, which are located in adjacent GEOS-Chem grid boxes. Because we average the seasonality across the Northern Hemisphere, the aggregate of these high-latitude sites would remove any impact that fewer winter measurements have. Figure 1 plots the Northern Hemispheric seasonality without the sites north of 50°N , Bialystok and Bremen, for comparison. The only sites that are not included in the Northern Hemisphere seasonality are those which began taking measurements less than a year before the end of the model run: Saga and Réunion Island. We have rectified this omission in the text.

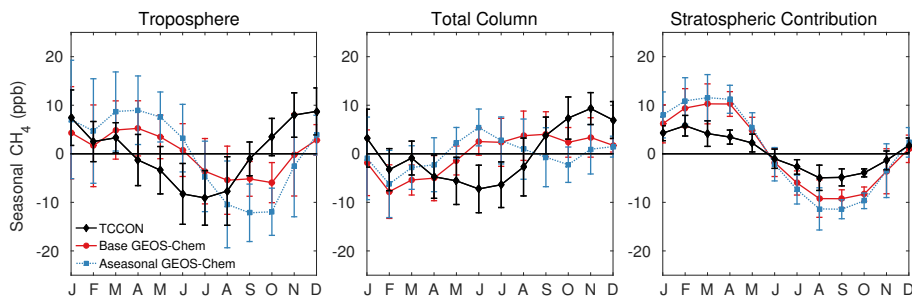


Figure 1. Detrended seasonality of TCCON (black diamonds), GEOS-Chem base (red circles), and GEOS-Chem aseasonal (blue squares) CH_4 column-averaged DMFs, averaged across Northern Hemisphere sites, except Bialystok, Bremen, Saga, and Réunion Island. Error bars denote the 1σ standard deviation across sites.

Another surprise here is that that seasonal cycle of the tropospheric and stratospheric columns in the aseasonal case are essentially in phase, yet when the total column is considered, a bimodal seasonal cycle is found. How can this be?

The stratospheric contribution is the amount by which the stratosphere decreases the total column average (via stratospheric loss and transport). Thus, the stratospheric contribution has an inverse effect on X_{CH_4} relative to $X_{\text{CH}_4}^t$, and the balance between the stratospheric contribution and $X_{\text{CH}_4}^t$ causes the seasonality in X_{CH_4} . We define the stratospheric contribution more explicitly and include its derivation in Appendix C to prevent confusion.

P10, L11: I disagree with this statement: it seems that the seasonal cycle of the modelled stratospheric columns precede the seasonal cycle of TCCON by a good month.

The comparison of the stratospheric seasonality is difficult to assess by eye, but the stratospheric contributions of TCCON and GEOS-Chem are in phase, which is illustrated by the shared inflections point in June and December.

Figure 8: The smoothing carried out here is not informative. Why not a box and whiskers plot to show how variable the data really are? Also, Park Falls is rather a tricky station with quite a lot of local influence and not a clear seasonal cycle. Perhaps another station would be more informative?

Park Falls was chosen because of the TCCON sites that also have surface observations, the $X_{\text{CH}_4}^t$ seasonality most closely matches the Northern Hemisphere mean shown in Fig. 7; thus Fig. 8 provides a good basis to compare surface and tropospheric column measurements. While the site does have a complicated seasonality near the surface, we find it notable that GEOS-Chem is able to capture several of those features, especially the local minimum in October, but still deviates from the observations, as we note on p.101.8. The box and whisker plots with superimposed observations and model data were difficult to follow visually. Instead, to show the variability, we have added to Fig. 8 lower and upper bounds denoting the 25th and 75th percentiles, respectively, of detrended data for each month.

Also, is the temporal sampling of the model consistent with that of the rather sparse flasks?

We had compared more frequent "Programmable Flask Package" (PFP) measurements, which have been measured at Park Falls since 2006, and found only slight differences in the seasonal cycle. Because we could not find equivalent in situ NOAA measurements, which we chose because they are on the same calibration scale as TCCON (Wunch et al., 2010), in the Southern Hemisphere, we only plot the flask measurements. Figure 2 plots Fig. 8 with the higher resolution flask data included for your reference.

In a broader sense I'm not sure what the real message here is. We see already in Figure 7 that the GEOS-Chem run does a very poor job of representing the seasonal cycle in the NH column: would you expect it to be better at the surface?

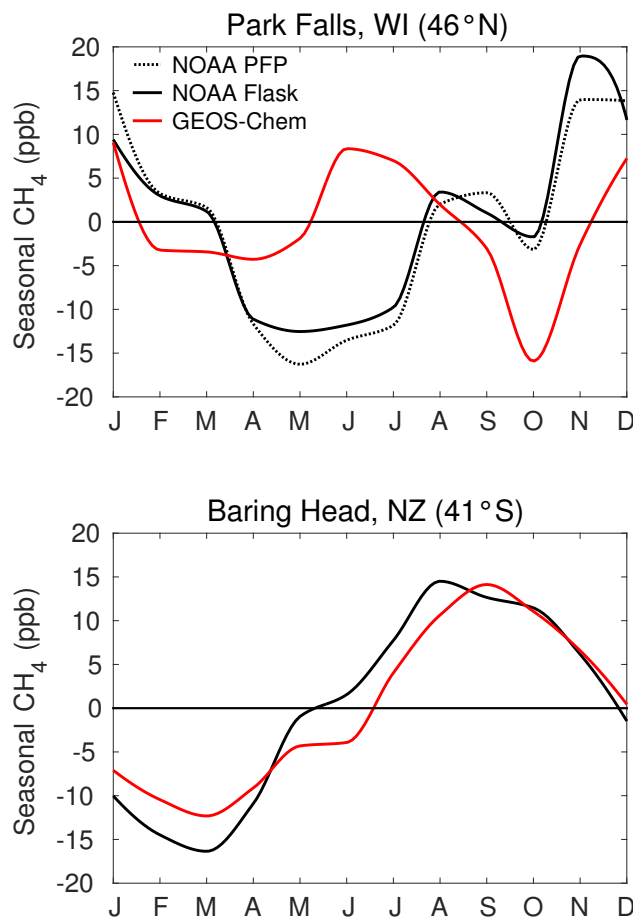


Figure 2. NOAA tall tower PFP flask (black dashed line), NOAA surface flask (black solid line), and GEOS-Chem surface level (red solid line) seasonality of CH₄ DMFs over 2005-2011 at Park Falls, WI, USA and Baring Head, NZ. PFP data is courtesy of Arlyn Andrews (NOAA): Andrews, A.E., E. Dlugokencky, and P.M. Lang (2008), Methane Dry Air Mole Fractions from the NOAA ESRL Surface Network using Programmable Flask Packages (PFP), 1992-2008, Version: 2013-07-03.

We included surface measurements (a) to demonstrate that the seasonality that we see is not due to some unknown bias in the $X_{\text{CH}_4}^t$ measurements and (b) to test whether the phase shift could be due to vertical transport, which would create a smaller lag at the surface, or horizontal transport, which is our hypothesis.

5 **Figure 9: Please label the plots (especially upper panel).**

The upper panel of Fig. 9 is now labeled.

Figure 10: The y-axis should have the same scale for the top and bottom figures, even if only part of the range is shown.

The y-axes of the two subfigures in Fig. 10 have been scaled so that the latitude grid boxes are equal.

I was also not quite sure about the units here. 10^6 kg is 0.001 Tg, so the bright yellow ($10 \cdot 10^6$ kg CH₄) is 0.01 Tg CH₄. But then in Figure 11 the increments between the seasonal and aseasonal run seem to be rather on the order of 1 Tg CH₄ mo⁻¹, which is two orders of magnitude higher. Or have I missed something here?

5 Figure 10 shows the zonally averaged wetland emissions, while Fig. 11 displays the total difference in emissions. The units in Fig. 10a have been changed to Gg, and the description of units in the caption of Fig. 11 has been changed from “Tg” to “summed over each zonal band, in Tg·mo⁻¹” to prevent confusion.

P13, L5-6: I did not quite understand the description of what you did here. You write "derived by calculating the total emissions resulting from an increase of 1 ppb of CH₄ in each surface grid box". Do you mean by calculating the emissions required to cause a 1 ppb increase in each surface grid box? How often were you adding this increment? Monthly? Do you consider the effect that these emissions have on the concentrations of neighbouring grid boxes? Is there a reference that explains this procedure in a bit more detail? Based on what is written here, I could not reproduce the experiment.

15 To show the true change in posterior emissions associated with a phase lag, the gain matrix would need to be derived for all grid boxes in the model. Because we did not have the actual sensitivity of CH₄ to wetland concentrations, which varies spatially depending on proximity to sources, we estimated that sensitivity as the mass of CH₄ associated with a 1 ppb increase in CH₄ in the surface grid box. The change in posterior emissions was then calculated as the product of this sensitivity and the fraction of the monthly mean emissions from wetlands in each surface grid box. Figure 10b mapped the difference between
20 this change in posterior wetland emissions and the value in the same grid box three months prior, summed for each zonal band. Because this approach does not include any of the information about transport (as would exist in the linear operator that transforms model emissions to concentrations), we are not able to consider neighboring grid boxes. We have since updated the calculation as the sensitivity to 1 ppb increase in CH₄ over the tropospheric column, as the focus on this analysis is the assimilation of column data.

25

P14, L8: I don't think you have convincingly shown that the seasonal lag is a function of transport, and not, say, your sink, or the spatial distribution of the fluxes.

The sensitivity experiments we ran tested the model's response to a number of different emissions, OH, and meteorology fields. The seasonal phase shift in the tropospheric column appeared in all simulations, although the seasonal cycle amplitude
30 and the shape of the springtime maximum varies. We have added a table describing these simulations and a figure that plots the tropospheric seasonality, as well as deviations from the base simulation, of each of these simulations to Appendix B1, “Equilibrium Sensitivity Experiments.” We have also removed the sentence in the conclusion referred to by this comment.

P15, L1-2: While I agree that prescribing the stratospheric CH₄ fields based on satellite observations might help, this will lead to transport that is not mass conserving, which is a problem for flux inversion. Please comment.

The insensitivity of the stratosphere to perturbations in tropospheric CH₄ suggest that prescribed stratospheric CH₄ would not need complicated adjustment to enforce mass conservation. We agree that the mechanism by which a model would set these CH₄ fields in the stratosphere would require careful consideration of how best to ensure the conservation of mass. For example, the stratospheric fields could be scaled according to the mass flux from the troposphere. As models develop their representation of stratosphere-troposphere exchange, however, the conservation of mass will become a more complicated problem. In addition to the UCX mechanism we suggested, a variety of linear schemes for stratospheric CH₄ have been tested for other models, such as Slimcat (Monge-Sanz et al., 2013).

Perhaps also mention that MIPAS and ACE-FTS are both good candidates for such an approach, but the former is not flying right now, and the latter has already been flying for 11 years and there is no replacement in sight.

This sentence now reads, “satellite observations from ACE-FTS, MIPAS (von Clarmann et al., 2009), or a compilation of remote sensing instruments (Buchwitz et al., 2015).” While stratospheric CH₄ fields for specific years would be ideal, even a monthly climatology with a secular increase applied would be an improvement on the current loss parameterization, which are monthly fields that do not vary interannually.

15

Typographical/language comments:

P3, L9: add "the" before "assimilation"

Table 1: The sign on the latitude of Darwin is wrong in this table.

P4, L11: Add degree symbol on both 4 and 5.

20 **P5, L5: "data WERE available" (plural)**

P5, L13: "and initial conditions" -> "and used as initial conditions"

There is no reference to Appendix A2 in the text.

P5, L18: "test the dependence of our results ON the"

25 **p6, L1, L5, and a few other places: "emissions seasonality" isn't quite right. It should either be "the emissions' sensitivity" or "the seasonality of the emissions".**

p11, L6: emissions -> emission

The above changes were made, and Appendix A2 (now B2) is now referenced in Section 2.2.1

References

- Bates, K. H., Nguyen, T. B., Teng, A. P., Crouse, J. D., Kjaergaard, H. G., Stoltz, B. M., Seinfeld, J. H., and Wennberg, P. O.: Production and Fate of C4 Dihydroxycarbonyl Compounds from Isoprene Oxidation, *The Journal of Physical Chemistry A*, 120, 106–117, doi:10.1021/acs.jpca.5b10335, <http://dx.doi.org/10.1021/acs.jpca.5b10335>, 2016.
- 5 Buchwitz, M., Reuter, M., Schneising, O., Boesch, H., Guerlet, S., Dils, B., Aben, I., Armante, R., Bergamaschi, P., Blumenstock, T., Bovensmann, H., Brunner, D., Buchmann, B., Burrows, J. P., Butz, A., Chédin, A., Chevallier, F., Crevoisier, C. D., Deutscher, N. M., Frankenberg, C., Hase, F., Hasekamp, O. P., Heymann, J., Kaminski, T., Laeng, A., Lichtenberg, G., De Mazière, M., Noël, S., Notholt, J., Orphal, J., Popp, C., Parker, R., Scholze, M., Sussmann, R., Stiller, G. P., Warneke, T., Zehner, C., Bril, A., Crisp, D., Griffith, D. W. T., Kuze, A., O'Dell, C., Oshchepkov, S., Sherlock, V., Suto, H., Wennberg, P., Wunch, D., Yokota, T., and Yoshida, Y.: The Greenhouse
- 10 Gas Climate Change Initiative (GHG-CCI): Comparison and quality assessment of near-surface-sensitive satellite-derived CO₂ and CH₄ global data sets, *Remote Sensing of Environment*, 162, 344–362, 2015.
- Fraser, A., Chan Miller, C., Palmer, P. I., Deutscher, N. M., Jones, N. B., and Griffith, D. W. T.: The Australian methane budget: Interpreting surface and train-borne measurements using a chemistry transport model, *Journal of Geophysical Research: Atmospheres*, 116, n/a–n/a, doi:10.1029/2011JD015964, <http://dx.doi.org/10.1029/2011JD015964>, d20306, 2011.
- 15 Jones, A., Walker, K. A., Jin, J. J., Taylor, J. R., Boone, C. D., Bernath, P. F., Brohede, S., Manney, G. L., McLeod, S., Hughes, R., and Daffer, W. H.: Technical Note: A trace gas climatology derived from the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) data set, *Atmospheric Chemistry and Physics*, 12, 5207–5220, doi:10.5194/acp-12-5207-2012, <http://www.atmos-chem-phys.net/12/5207/2012/>, 2012.
- Monge-Sanz, B. M., Chipperfield, M. P., Untch, A., Morcrette, J.-J., Rap, A., and Simmons, A. J.: On the uses of a new linear scheme for
- 20 stratospheric methane in global models: water source, transport tracer and radiative forcing, *Atmospheric Chemistry and Physics*, 13, 9641–9660, doi:10.5194/acp-13-9641-2013, <http://www.atmos-chem-phys.net/13/9641/2013/>, 2013.
- Park, R. J., Jacob, D. J., Field, B. D., Yantosca, R. M., and Chin, M.: Natural and transboundary pollution influences on sulfate-nitrate-ammonium aerosols in the United States: Implications for policy, *Journal of Geophysical Research: Atmospheres*, 109, 2004.
- Patra, P., Krol, M., Montzka, S., Arnold, T., Atlas, E. L., Lintner, B., Stephens, B., Xiang, B., Elkins, J., Fraser, P., et al.: Observational
- 25 evidence for interhemispheric hydroxyl-radical parity, *Nature*, 513, 219–223, 2014.
- Rodgers, C. D. and Connor, B. J.: Intercomparison of remote sounding instruments, *Journal of Geophysical Research*, 108, 4116, doi:10.1029/2002JD002299, 2003.
- Saad, K. M., Wunch, D., Toon, G. C., Bernath, P., Boone, C., Connor, B., Deutscher, N. M., Griffith, D. W. T., Kivi, R., Notholt, J., Roehl, C., Schneider, M., Sherlock, V., and Wennberg, P. O.: Derivation of tropospheric methane from TCCON CH₄ and HF total column
- 30 observations, *Atmospheric Measurement Techniques*, 7, 2907–2918, doi:10.5194/amt-7-2907-2014, <http://www.atmos-meas-tech.net/7/2907/2014/>, 2014.
- Stephens, B. B., Gurney, K. R., Tans, P. P., Sweeney, C., Peters, W., Bruhwiler, L., Ciais, P., Ramonet, M., Bousquet, P., Nakazawa, T., Aoki, S., Machida, T., Inoue, G., Vinnichenko, N., Lloyd, J., Jordan, A., Heimann, M., Shibistova, O., Langenfelds, R. L., Steele, L. P., Francey, R. J., and Denning, A. S.: Weak Northern and Strong Tropical Land Carbon Uptake from Vertical Profiles of Atmospheric CO₂, *Science*,
- 35 316, 1732–1735, doi:10.1126/science.1137004, <http://science.sciencemag.org/content/316/5832/1732>, 2007.
- von Clarmann, T., Höpfner, M., Kellmann, S., Linden, A., Chauhan, S., Funke, B., Grabowski, U., Glatthor, N., Kiefer, M., Schieferdecker, T., Stiller, G. P., and Versick, S.: Retrieval of temperature, H₂O, O₃, HNO₃, CH₄,

- N₂O, ClONO₂ and ClO from MIPAS reduced resolution nominal mode limb emission measurements, *Atmospheric Measurement Techniques*, 2, 159–175, doi:10.5194/amt-2-159-2009, <http://www.atmos-meas-tech.net/2/159/2009/>, 2009.
- Wunch, D., Toon, G. C., Wennberg, P. O., Wofsy, S. C., Stephens, B. B., Fischer, M. L., Uchino, O., Abshire, J. B., Bernath, P., Biraud, S. C., Blavier, J.-F. L., Boone, C., Bowman, K. P., Browell, E. V., Campos, T., Connor, B. J., Daube, B. C., Deutscher, N. M., Diao, M., Elkins, J. W., Gerbig, C., Gottlieb, E., Griffith, D. W. T., Hurst, D. F., Jiménez, R., Keppel-Aleks, G., Kort, E. a., Macatangay, R., Machida, T., Matsueda, H., Moore, F., Morino, I., Park, S., Robinson, J., Roehl, C. M., Sawa, Y., Sherlock, V., Sweeney, C., Tanaka, T., and Zondlo, M. a.: Calibration of the Total Carbon Column Observing Network using aircraft profile data, *Atmospheric Measurement Techniques*, 3, 1351–1362, doi:10.5194/amt-3-1351-2010, <http://www.atmos-meas-tech.net/3/1351/2010/>, 2010.
- Wunch, D., Toon, G. C., Blavier, J.-F. L., Washenfelder, R. a., Notholt, J., Connor, B. J., Griffith, D. W. T., Sherlock, V., and Wennberg, P. O.: The Total Carbon Column Observing Network, *Philosophical transactions. Series A, Mathematical, physical, and engineering sciences*, 369, 2087–2112, doi:10.1098/rsta.2010.0240, <http://www.ncbi.nlm.nih.gov/pubmed/21502178>, 2011.
- Wunch, D., Toon, G. C., Sherlock, V., Deutscher, N. M., Liu, C., Feist, D. G., and Wennberg, P. O.: The Total Carbon Column Observing Network’s GGG2014 Data Version, Tech. rep., Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, Oak Ridge, Tennessee, U.S.A. doi: 10.14291/tccon.ggg2014.documentation.R0/1221662, doi:10.14291/tccon.ggg2014.documentation.R0/1221662, <http://dx.doi.org/10.14291/tccon.ggg2014.documentation.R0/1221662>, 2015.