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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Seasonal Variability of Stratospheric Methane: Implications for Constraining Tropospheric Methane Budgets Using Total Column Observations" *by* K. M. Saad et al.

Anonymous Referee #3

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The manuscript presents a study looking at forward simulations of methane in the GEOS-Chem model, and compares the column-integrated tropospheric/stratospheric column in the model with that derived from tropospheric/stratospheric retrieval of XCH_4 from TCCON employing the HF method outlined in Saad et al. (AMT, 2014). The idea, as outlined in the abstract, is compelling: models might match the total column methane mixing ratio measurements of TCCON (and, by extension, satellites), while having compensating errors in the tropospheric and stratospheric components, which would, in turn, bias flux inversions based upon such a model. However it is not clear to me that this study demonstrates this.

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

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. Without this, I am not sure that the conclusions are clear enough to warrant publication in ACP.

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 showns 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.

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 statospheric 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 CH_4 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.

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.

More minor comments:

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.

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.

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

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

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?

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.

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 hte stratosphere than in the troposphere for the Base case. This doesn't make sense to me. Please explain.

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

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?

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 figures presented. Could you comment on this? How do ACE-FTS and TCCON agree?

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? 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? 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?

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.

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? Also, is the temporal sampling of the model consistent with that of the rather sparse flasks? 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 9: Please label the plots (especially upper panel).

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. I was also not quite sure about the units here. 10^6 kg is 0.001 Tg, so the bright yellow (10 10^6 kg CH_4) is 0.01 Tg CH_4. But then in Figure 11 the increments between the seasonal and aseasonal run seem to be rather on the order of 1 Tg CH_4 mo^{-1}, which is two orders of magnitude higher. Or have I missed something here?

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 CH_4 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 neibouring 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.

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.

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. 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.

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.

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"

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

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