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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Constraints and biases in a tropospheric two-box model of OH" *by* Stijn Naus et al.

Anonymous Referee #2

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Review of Naus et al., 2018

This paper investigates the use of tropospheric two-box models for estimating global tropospheric OH concentrations and methane (CH4) emissions. Output from a 3D atmospheric model (TM5) was used to investigate a range of potential biases brought about through the simplifying assumptions inherent in a box model framework. In particular, the influence simple parameterisations of inter-hemispheric transport, stratospheric loss and inter-hemispheric differences in OH are investigated, along with the ability of sampling networks to represent the hemispheric averages used in two-box models. This work follows from some recent publications that have used such models to propose that changes in OH, whilst being highly uncertain, may have contributed to some of the recent variability in CH4. The findings of Naus et al broadly agree with the

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inter annual changes inferred in these studies. However, this paper highlights the limits of such analysis and gives an indication of the impact of the above-mentioned biases on the outcome of such inversions.

The paper is topical and makes a valuable contribution to the recent literature on methane and OH variations and should be suitable for publication in ACP, following some changes.

General comments:

1. While the paper is generally well written, it is quite long and at times a little too verbose. This is partly because some of the issues are technical and, for the most part, are rightly discussed in depth. I've made a few suggestions where the text could be clarified or shortened below. However, I would encourage the author team to take another look over the manuscript as a whole and try to re-structure and re-focus parts of the text and shorten some bits. 2. Wouldn't the site-specific mole fractions from the 3D model be biased high because of the presence of "local pollution" in the model? As I understand it, NOAA conditionally sample data to select background mole fractions where possible, and then additional filtering is applied to remove obviously "polluted" samples. In contrast, the 3D model, particularly at the coarse resolution used (and I'm assuming for monthly mean output?), will almost always be influenced by nearby sources at most NOAA sites. The authors should at least mention this. Ideally, local pollution could be removed in the model mole fractions. 3. In Section 2.4, I think it might be worth clarifying how the "combined biases" run was done. I assume this was from a single model run, outputting all time-dependent quantities, rather than summing the individual biases? 4. Am I wrong to be left with the impression that, in order to avoid these biases, rather than throwing away 2-box models entirely, we could just re-parameterize them based on the outputs of 3D models (as was done in the inversions in this paper)? There are still good reasons why we might want to do this. For example, Rigby et al. (2017) used an MCMC approach which required many thousands of model evaluations. This is challenging with a 3D model, but fairly trivial with a box

model. Therefore, if we could use a small number of 3D model runs to derive better parameters, we could still use the advantages of a Monte Carlo inverse method (e.g. non-Gaussian distributions, non-linear models, etc).

Specific comments:

Title: "tropospheric two-box model of OH". Perhaps this should be OH, CH4, and methyl chloroform? Or just "in a tropospheric two-box model".

P1 L2 and several other places ("In two recent studies…"). A box model was also used in McNorton et al., 2016. However, I believe this was a 1-box model. Do you also want to discuss this? Or limit your discussion strictly to two-box models?

P1 L8: It's not quite accurate to say that two-box model studies use "fixed model parameters for interhemispheric transport, chemical loss rates and loss to the stratosphere". E.g. in Rigby et al., 2018, interhemispheric transport was allowed to vary each year (even though it didn't have any a priori interannual variability), and stratospheric loss depended on the stratospheric MCF or CH4 concentration, etc.

P1 L16 ("However, …"). This sentence is somewhat of a non sequitur. What is it in comparison to? E.g. are you saying that there is no overall trend without all biases?

P1 L17 ("Moreover, the magnitude… "). Perhaps say "absolute magnitude" to make this sentence more clear?

P1 L20 – L25: Given that the main results on OH and CH4 anomalies aren't too dissimilar from the two-box model studies compared to the other uncertainties (Figure 6), is it fair to say that it is "crucial" to use a 3D model? (Also see general comment 4).

P2 L3: The use of "resorts" makes it sound as if models are only used as a last resort. They are essential to understanding the data.

P2 L4: "most involving 3D transport model". Perhaps change to "more complex 3D transport models".

P2 L10: What does "explicitly contain much information on a species' distribution" mean? Does this mean that there is no information on longitudinal gradients in mole fractions, etc. If so, what is this trying to imply?

P2 L14: suggest change to ", which is largely determined by the TROPOSPHERIC hydroxyl radical CONCENTRATION"

P2 L18: "consequences this has had in the past" (grammar). However, perhaps this sentence can be cut as it doesn't really say anything.

P2 L22: suggest "more robust observational constraints on OH on the large scales are THOUGHT TO BE derived indirectly. . ." because we don't really know this for sure.

P2 L25 (e.g. Montzka et al., 2000; Bousquet et al (2005)). I suggest referencing some of the earlier papers here (e.g. Lovelock, Prinn)

P2 L30: Not sure why Montzka et al is described as the "most important" work here? Seems unnecessary. Also, note that the results of Montzka et al are consistent with Rigby et al., 2017.

P3 L10: "do not significantly affect the outcome". I don't think you know this for sure, so suggest changing to "are not thought to significantly affect the outcome"

P3 L12: Perhaps change "approximating interhemispheric (IH) exchange using SF_6" to "approximating the possible range of interhemispheric exchange rates using SF_6" or similar, because Rigby et al. only used SF_6 to estimate the a priori value of IH exchange rate (although, not surprisingly, these values weren't strongly updated in their inversion).

P3 L7 – L30: I believe these paragraphs are trying to state the reasons why you'd want AT LEAST a two-box model (i.e. so that you can examine the trend and IH gradient). Is that right? Otherwise, it's not clear why these two criteria are described in detail, rather than other factors (e.g. computational efficiency)

P3 34: “reversely” suggest change to “conversely”

P4 L5 – L20: This paragraph is a little hard to follow as it concerns two different parts of the study, the first of which is subdivided into four sub-sections. For the latter, I suggest using “firstly”, “secondly”, “thirdly”, “fourthly” to make it clear where this section ends and the next begins. Or restructure in some other way.

P5 L3: HˆT. The T should not be bold.

P6 L2: suggest change to “leave more freedom with respect to the timing of emissions”. However, I’m not sure what this assertion is based on? Is this justified? If so, how?

P7 Table captions: suggest change “perturbed in Monte Carlo” to “perturbed in Monte Carlo ensemble” or something more descriptive.

P8 L18: The nudged simulation needs to be described in a little more detail here (but still succinctly). It’s not obvious what distinguishes it from the other analysis from the description in this paragraph.

P8 L29: Perhaps not obvious what is meant by “budget” in this context. The sum of the sources minus the sinks in each box? If so, perhaps just say so.

P9 L2: “Note that we have do not” (delete “have”?)

P9 L31: “that deviated significantly from what is generally expected”. This is a very vague sentence. I’m not sure what the authors are trying to say here. Are they saying that this is based on their intuition?

P10 L10: “has undergone” instead of “undergoes”, which suggests that MCF is redistributed repeatedly?

P10 L18: “. . . surface measurements do not inform much on vertical gradients”. Not entirely sure what this means (seems obvious), but re-wording is at least required. Perhaps delete.

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P10 L20: "but this information is difficult to correctly incorporate. . ." Why would this be the case?

P10 L32: suggest "We explore this bias. . ."

P11 Figure 1 caption: "but adjusted by correction factors". I suggest being more specific. What exactly has been done.

P11 L5: I'm not sure that it's fair to say that Rigby et al assumed a "constant stratospheric lifetime". The local lifetime in their stratospheric box was time invariant. However, this would mean that the transient lifetime varied as the distribution of MCF changed. Perhaps say that the local stratospheric loss rate was constant (could also say that the value of this loss was consistent with a steady-state lifetime of X years)?

P11 L13: "As mentioned in . . . would obscure the impact of the bias correction." I'm not sure what this sentence means. Needs to be more specific.

P12 L4: "Equations 1 and 7". Perhaps it's better to reference Section 2.2.2, which actually describes how this calculation was done?

P12 L8: perhaps "SF6 has no SIGNIFICANT sink" or similar (not significant over these timescales, but it does have a sink)

P12 L13: "changes sign (NOT SIGNS) in the tropics". I'm not sure what is meant by this. Do you mean that the gradient changes sign seasonally now? If so, why just in the tropics?

P12 L18: Does SF6 have annually repeating sources in the model runs too? Would be good to remind us here.

P12 L20: What does "the treatment of this data in TM5" mean?

P13 L21 – L26: This seems relatively important. Worth showing as dotted lines in Figure 2?

P13 L28: Reference Figure 1 as well as Figure 3?

P13 L30: should this be “tropospheric hemisphere” instead of “troposphere”?

P14 L8: “The shift in the bias for MCF is driven by the latitudinal dimension “. What does this mean?

P14 L8: “combined with simple latitudinal interpolation.“ Not sure what this interpolation refers to. EDIT: After reading the Supplement, I think I understand a bit better. However, it still needs briefly explaining in the main text.

P15 L9 – P16 L8: Is this long discussion of (random?) uncertainties really necessary? To me it seems that this is a side issue that distracts from the main message (the biases). Perhaps cut this, or shorten substantially.

P16 Figure 4: I suggest using a more descriptive y-axis label.

P16 L7: Should this be Rigby et al.. 2017, rather than 2013?

P18 L1: This is an interesting discussion. However, I'm interested in the relative magnitudes. Would treating the stratospheric loss as a function of the strat-trop gradient get us most of the way to removing the “bias” (or would the addition of the stratospheric box be a pretty good way forward)? Or do you really need resolved stratospheric transport in detail to address most of this issue?

P18 L15: I assume this change would happen over a period of years? The sentences makes it sound rather instantaneous.

P19 L14: “visible” rather than “visibly”

P22 L11: Is the use of “persistent” correct here? Aren't you referring to a transient effect. Also, similarly to comment on P18 above, do we know how much of this drop might be expected with a model that attempts to resolve the stratosphere (e.g. Rigby et al., 2017).

P22 L22: I'm not entirely sure what "persistent interannual variations in transport of MCF of up to 20%" means. Can you clarify?

P22 L22: I don't like the use of the word "tricky". Furthermore, couldn't the reasons for this due to more than just the use of box models? E.g. changes in magnitude and timing of emissions reduction?

P22 L25: Rigby et al. didn't assume a fixed emissions uncertainty of 10Gg/yr. They used an emissions model that had a lot of unknown parameters (including the possibility up to 10Gg/yr of unreported production).

P22 L34: Perhaps "gross overestimate" is too strong!

P23 L3 – 4: "where information is weighted very differently". This could do with clarification as I'm not entirely sure what is meant by this.

P24 L2: "However, the potential and the magnitude of the biases are real". Please clarify.

Supplement:

S1: Wouldn't it be safer to omit months where there was a significant fraction of missing data, rather than interpolate from nearby stations?

S3.5: Perhaps choose and different subheading that "conclusions" here. Make it more specific since it is not an overall conclusion.

References

McNorton, J., Chipperfield, M. P., Gloor, M., Wilson, C., Feng, W., Hayman, G. D., Rigby, M., Krummel, P. B., O'Doherty, S., Prinn, R. G., Weiss, R. F., Young, D., Dlugokencky, E. and Montzka, S. A.: Role of OH variability in the stalling of the global atmospheric CH4 growth rate from 1999 to 2006, Atmospheric Chemistry and Physics, 16(12), 7943–7956, doi:10.5194/acp-16-7943-2016, 2016.

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