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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Attribution of recent increases in atmospheric methane through 3-D inverse modelling" *by* Joe McNorton et al.

Anonymous Referee #2

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General comments :

This paper by McNorton et al. addresses the important question of attributing the unexplained recent changes in atmospheric methane concentrations (since 2007) to methane sources and sinks. The validation part is good and useful.

The main originality of the paper is to include 13CH4 observations and OH changes in an inversion based on a 3D transport model, as compared to previous crude box modelling approaches (including Nature papers !)

The results of this paper are very important for the methane & climate communities, although, in the present form of the paper, they are not valorized because of : - a rather confusing organization of section 3 and conclusions. - a general lack of details

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and precisions in all sections - a lack of precise comparison with previous analyses

My main demands are (see specific comments) - to rewrite sections 3.2 and 3.3 clearly presenting and separating results & analysis of o mean emission and sinks versus their changes, o results & analysis global scale versus regional o results & analysis of emissions versus the sinks - to comment on all emissions (anthropogenic microbial emission poorly commented) - to report emission changes in Tg/yr between 2 time periods and not in trends (Tg/yr2), - to add a table with emissions changes, - to add a discussion section where comparison with other studies can be grouped

In short, this is an important paper to be published in ACP, material is mostly there but a profound rewriting/organization of the results & conclusions sections is needed.

Specific comments :

P2-L15-20: please note that these mean isotopic signatures are associated with rather large range. It may be worth writing also that total source signature is -51/-53‰

P2 L25 : “although they emphasised that the problem is not very well constrained by existing data Âż I suggest to be more precise : although these two studies cannot discard the hypothesis that OH is not changing.

P3 L5 : please define shortly here “synthesis inversion” (3D modelling, reduction of the size of flux and observation spaces to solve the inverse problem, . . .) focusing on the imorvement compared to box models.

P3 L18 : what is a one-year inversion spin-up ? please detail a bit.

P3 L21-22 : what is the influence of this choice ? what do you take for geological emissions ? It might be worth making a sensitivity test by taking the values from Saunois et al (2016) (update of the Kirschke paper, please quote) instead of the Schwietzke paper.

P3 L26 : I would be worth mentioning which of your sources is prescribed in the prior with internannual variability. Maybe in table 1.

P3 L30 : I understand that you compute monthly response functions using the forward model? Please specify this here.

P4 L 5 : why increasing OH and Cl ? please justify this choice ?

P4 L9-10 and P5 L9-14: not clear. How do you deal with the long-term equilibration of 13CH4 (e.g. Tans 97 paper) with 1-yr inversions ?

P4 L11 : Âń For the inversion including OH concentrations" : this suggests that there are inversions without OH in the state vector. Please clarify.

P4 : I understand that isotopic signature are not optimized in this procedure. Please precise this point.

P5 L4-7 : Bousquet et al 2011 addressed this point and tested a second iteration with only small impact on the inversion results, so consistent with your hypothesis. It might be worth quoting.

P5 L9 : "The model OH is constrained by CH4 and δ13CH4" : this is a weak constraint as many combination of total source and mean OH can fit the atmospheric changes. Please notice it here ? With such a configuration you largely depend on the prior for the mean emissions and sinks so I would not insist a lot in the paper on the posterior versus prior comparison but more on budget changes with time and between your different inversions.

P6 L10-11 : putting only one value of uncertainty for all stations is a bit crude as model error will not be the same for remote sites of the southern hemisphere and continental sites of the northern hemisphere. More refinement is needed here or at least a sensitivity test varyng observational errors

P6 : It might worth doing a sensitivity test with more atmospheric observations, when appearing in the network. The apparition of stations is an issue but can help analyzing

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regional gradients more safely. As you perform yearly inversions, why not adding each year the stations appearing in your inversions ?

P7 L22 : “slow inter-hemispheric transport within the model” : please provide the Inter Hemispheric Time and/or a reference for this possibly from transcom experiments ?

P7 L27 : For Garmisch did you try to extract the station at different level in your model ?

P7 L29 : 21.4 ppb is still a quite large value. Can you at least make hypotheses to explain them ?

P8 – sect3.2 : “OH concentrations in INV-FULL and INV-CH4 are relatively constant throughout the period 2007-2015 (Figure 5) but these values are smaller by 1.8±0.4% and 0.3±0.5%” : unclear : do OH is constant or diminishing. Please clarify. Also, I find a bit strange to start by the section by the sink and not by source changes.

P8 L9-10 : it may be good to refer to the sensitivity test on OH (S9) here.

P8 l11-15 : mixing the mean changes compared to the prior and the time changes from 2003-2006 to post-2007 period is confusing. What about change in agriculture flux ? I suggest to group discussions on the mean sources and sinks global and regional (table 4) and then address the changes (table 5)..

P8 L24 : how did you estimate the 30% for OH and 60%/10% values ? Did you use S9? Please justify.

P8 : The choice to report changes in trends (Tg yr-2) is a bit technical. A suggestion would be to report emission change between two periods (e.g. 2003-2005 and 2012-2015) in Tg/yr and quantify the % of contributions from this.

Table 5 : there are some values worth to comment in your analysis : increase emissions from NA ? dipole +0.59 / -0.58 for energy between NA and EA ? wetland increase in Eurasia ? Why ? visible in other studies ?

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Section 3.2 : You do not comment waste sector (+0.46 in your table 5). Indeed, anthropogenic microbial emission contribute almost as much as wetlands (0.46 and 0.2 trends globally versus 0.8 for wetlands). Please add comments on anthropogenic microbial emission changes.

P9 L5-9 : you have very few stations constraining NA inland emissions. It should be notices here as the increased inferred emissions over NA is a hot topic. Again including NA inland stations in a sensitivity inversion seems necessary to confirm such a result. In any case this has to be commented here.

P10 : It is a bit difficult to follow all the trends provided and to compare them to the standard inversion. I suggest to make a table with results of sensitivity test for global scale compared to INV-FULL(in Tg/yr difference between 2 periods and not trends in Tg/yr2). Then you can more clearly comment on the differences in the main text.

P10 L7 : "the magnitude in post-2006 changes is typically increased in S9", please add something like : which is normal considering that constant OH as compared to decreasing OH in INV-FULL requires more emission change to match atmospheric observations

P10 L21-22 : Again, pleas acknowledge here that global total OH versus total emissions are not very well constrained without an external proxy as many commbiantion can match the growth rate. You largely rely to the prior in this case so I would not insist a lot on the posterior versus prior comparison but more on budget changes with time and between your different inversions.

P11 L15-30 : This comparison with EDGAR should be in in a discussion section between 3.3 and conclusions where you could compare your results with other studies. More references to previous results would be good e.g. Pouter et al. 2017 for wetlands, Saunois et al., 2017 ACP for all sources, more precise comparison about OH with Rigby and turner papers...

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P11 L21-22 : “As a result emissions from these regions are influenced by posterior emission changes and assumed to be underestimated in both magnitude and growth rate in the prior” : unclear to me, please rephrase.

P11 L27 I do not see this -2.2 Tg/yr2 in table 5 ? please clarify.

P12 L11 : you do not believe your results ? this sentence introduce confusion. Please rephrase it or remove it.

P12 L13 : Saunois paper is not an inventory by a synthesis of inventories and inversions. Please rephrase.

P12 L27 : Limitation of synthesis inversions (monthly means, coarse regions. . .) should also be mentioned here.

P12 L31 : what about NO2 decrease in Asia in the late 2000s ? Please mention this hypothesis as well.

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