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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 #1

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The manuscript "Attribution of recent increases in atmospheric methane through 3-D inverse modelling" by J. McNorton and co-workers adds to the considerable number of studies focussing on the question why atmospheric methane concentrations started rising again post 2007 after a decade of virtually stable concentrations. Although many suggestions based on different methodologies have been brought forward in recent years, there is still no final consensus on which source or atmospheric process or which combination of these has caused the renewed rise. By applying a 3D transport model to methane concentration and to 13CH4/12CH4 ratios the authors add an interesting and valuable study to the discussion that is in general worth publishing in ACP. However, I have several comments concerning the applied inversion method and model validation that need to be addressed before publication. I would also like to suggest that

the authors check the manuscript carefully again for a more precise language. Some examples are given below, but there are many more where a more precise wording would improve the readability of the text.

Major comments

1) Regional sub-division

What is the rational for the regional division applied in the transport model and the inversion? Especially the large EA and AO emission regions combining countries and regions with very different socio-economic developments in the last decades are very questionable choices. As the inversion is set up right now it can, for example, not distinguish between Western Europe with well established and generally decreasing CH4 emissions from most sectors from emissions in Russia or North-East Asia. These are areas with potentially growing emissions from different sectors in the last decades. Although, these trends may be presented in the a priori it seems more likely that there are uncorrelated uncertainties in the emission estimates for these areas. Similar arguments can be found for a required sub-division between south-east Asia and Australia. In the end, the current sub-division alters the derived regional trends in the a posteriori emission very questionable. For example opposing regional errors in the a priori trends in these large regions may alter it impossible for the inversion to correctly correct these trends. Instead the missing/excessive emissions may be lain down in/removed from regions for which little direct constraint is available from the utilised set of observations, but only a more global sensitivity exists in the model (such as the AM region). Maybe not surprisingly these are the regions for which the authors find the strongest changes in a posteriori emissions, a result that somewhat differs from conclusions in previous work. The regional sub-division certainly needs some further justification. This could be done by a more in-depth validation of the model performance at surface sites in contrasting areas like Europe vs. East Asia. For this the use of additional surface observations should be considered (see major comment 3).

2) General applicability of inversion method

As stated correctly on page 2 line 23, Rigby et al. (2017) and Turner et al. (2017) both conclude that the problem of the post 2007 methane rise may be under-constrained using the observed CH4 concentrations, 13CH4/12CH4 ratios and other tracers. Their conclusion is based on simpler box-model simulations without detailed regional division of CH4 emissions. In the present study an even larger number of unknowns is optimised through the inversion. Wouldn't this mean that the individual elements of the state vector are even less well constraint? The authors should spend some time justifying why their more detailed results should be better constrained than those from box-model analyses. In this context it may be worth looking at the covariances in the a posteriori emission and OH factor as well. Large negative covariances may indicate that the inversion cannot clearly distinguish between regions and sectors.

3) Surface observations

The authors base their inverse flux estimates on a limited set of surface observations (22 flask sampling sites). This may be justified in order to keep the influence of CH4 concentrations to 13CH4/12CH4 ratios similar, with the latter only being available at this limited number of locations. However, for validation purposes there would be many more CH4 observations available worldwide (flask and continuous). These should be evaluated as independent observations as well and may better than GOSAT and TCON observations demonstrate the success of the inverse flux estimate.

Minor comments

P2L26f: Although the global a priori emissions by source category are available in Table 1 and regionally divided a posteriori emissions are given in Table 4, I am missing the same kind of information for the a priori. An additional table in the style of Table 4 but for the a priori emissions should be added.

P4L7ff: If I correctly understand the inversion setup, the inversion step is performed on

batches of 12 months. Does this mean that the emissions from the previous year are not influenced by the observations of the next year at all? Meaning that January observations will not influence December emissions from the previous year? This would result in December, but probably also November and October, emissions always being constraint by less observations than emissions in other months and, therefore, probably are less corrected from their a priori values and/or show systematically larger a posteriori uncertainties than emissions in other months. Was this observed in the a posteriori factors?

P5L16: The wording is not very precise here. J is a cost function and the inversion will find its minimum. J is not a minimisation function. Instead equation 4 represents the analytical minimum of equation 3.

P5L24: R is not the covariance matrix of the observations alone. R contains the observation/model mismatch covariance. Later this fact is taken care of by adding a model uncertainty to R, but it should be correctly introduced here.

P6L6: Was any month-to-month variability of the emissions included in the a priori? If yes where was it taken from?

P6L10f: This is a bit simplistic since the model uncertainty most likely varies with the location of the observation and the question how representative the model grid cell can be for a given site. There have been many different approaches in the past on how to assign site-dependent model uncertainties and, hence, this point should be justified a bit more.

P7L6 and elsewhere: A lot of this RMSE is due to a bias in the a priori simulation. It would be better to calculate a bias-corrected RMSE instead. The bias could be mentioned separately. In general it would be nice to include all these comparison statistics in a table as well (in the main text for all discussed inversions and observational data sets and in the supplementary material for all sensitivity inversions).

P7L23f: I don't think it is the model that is growing here. What about 'simulated atmospheric methane growth rates' instead?

P7L27f: This behaviour is very strange. For all other sites an increase in concentrations from a priori to a posteriori simulations was observed. Why not for Garmisch, a central European site not too far away from the Bremen site, where differences in the a priori and a posteriori simulations are as expected? One potential source of mismatch may be the location of Garmisch at the northern edge of the Alps, potentially introducing large mismatches due to smoothed model topography. Still this would not explain the lack of an increase from a priori to a posteriori. Although a detail, this needs to be checked again.

P8L5 and Figure 5: The estimated a posteriori OH time series should also be compared with work by other authors (e.g. Rigby et al. 2017). If OH is really the main driver of the post 2007 CH4 rise it would be good to know how TOMCAT OH compares to previous work.

P8L15: A reference to Table 6 should be added here.

P9L11: A reference to Figure 5 should be added here.

P9L28f: How similar? These numbers are not given anywhere. One can only guess them from the figures. A table (like Table 4) with the a posteriori emissions for the INV-CL case should be provided and the same for all sensitivity inversion (supplement).

P9L28f: How is the a posteriori performance for this experiment (S4)? Just because one sensitivity run gives different a posteriori emissions it doesn't have to be wrong. But if it also fails to reproduce the observations, then the given conclusion may be correct.

P10L31f: This sounds a bit like the authors of Rigby et al. worked on the current study as well. Which is not the case. This work may extend the previous work by using a more complex transport model, but other than that the approaches are fairly different

and unrelated (inversion system, used observations, etc.). So I would not write that it extends the work of Rigby or others, but rather it adds to the results gained by others.

P11L7: 'larger errors'. What kind of errors? Needs to be repeated here.

P11L7f: The sentence 'The constraint improves when the δ13CH4 observations are introduced' should be re-written to be more precise. What about: 'The agreement of the simulations with observations improved when additional δ13CH4 observations are used to constrain CH4 fluxes.'.

P11L12: This conclusion is just based on the different trend compared with GOSAT, whereas the trend in surface observations was captured well in the a posteriori simulation. Does that mean that there is a potential trend in the bias between GOSAT and surface observations? Would there be any GOSAT validation studies that may provide some clarification?

P11L15f: Once again: There are more surface observations available than used in this study. They should be used for validation during this critical period.

P11L29: It is unclear which period is referred to here? Table 5 suggests a growth rate in the energy sector of the AO region of 1.5 Tg yr-2 the text states -2.2 Tg yr-2. What is correct?

P12, 1st paragraph: This section should also repeat what was stated in the introduction concerning previous inverse modelling studies (P2L21ff), especially since the presented results contradict/correct these earlier findings.

Figure1: Is is impossible to see the red dotted lines in many of the sub-panels (also the ones for δ13CH4). Either the figure needs to be enlarged/split or an additional color and solid line should be used for INV-CH4.

Table1, Table4, Table6: These should also contain the uncertainty estimates.

Table1: Maybe I missed this before, but does the missing number for the soil sink mean

that it was neglected completely? If it was only not-optimised its value should still be part of this table.

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