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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Mass balance inverse modelling of methane in the 1990s using a Chemistry Transport Model" *by* T. M. Butler et al.

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Review on Butler et al ACP paper

General comments

The paper by Butler et al presents results of a mass balance inversion of methane in the 1990s using a CTM. I think the paper presents enough original material to be published in ACP. However some changes are needed in the paper to clarify the methodology used (and its limitations) and the main results. I have 4 general comments about this paper.

First, the paper is a bit fastidious to read because of the long descriptions of other studies and redundancies in the citation of their results (introduction / section 2 / section 8). Authors have made a nice effort to quote most of the recent studies in details, but it

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is sometimes too much and thus makes the paper hard to read. Authors should track the redundancies in order to limit the citation of a result to one time only (also for their own results). Thus, main results will appear more clearly. I would suggest to shorten a bit the introduction (be more synthetic) and the long description of other mass balance inversions (section 2). As a model of synthetic material, I find the conclusion very goodĚ

Second, I think the author do not focus enough on the methodology they have used, and especially on the limitation of mass balance inversions. I do not like much this methodology (because of limitations see below) but mass balance inversion is one of the two techniques currently used for chemically active gases optimisations. It corresponds to the use of a full CTM with a simple sequential approach. The other approach is the linearization of the direct problem with a global variational inverse approach. At present, either you simplify the direct model or the inverse model. Authors have chosen the latter possibility. They quote some limitations in section 2 (no uncertainty, no distinction of source process) but there are some others : - the zonal correction of fluxes is a huge limitation for a (mostly) continental tracer such as methane, - the impossibility to correct past fluxes as in a pure sequential approach Some more sophisticated sequential optimisation exists (Kalman filtering, ..) and should be mentioned as perspectives. I think the limitations & perspectives of improvements should be discussed in section 2.

Third, about the data used to perform the inversion. Your results are sometimes influenced significantly by the type of network. Investigating further what stations (or group of stations) are responsible for this would be of great interest ? Did you look at that ?

At last, I think that all figures showing the different cases should have the same scale. No IAV is an information as a large IAVĚ This is a case for : fig9abc, fig14a & fig15a, fig14b & fig15b, fig16abcd.

Specific comments

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P5 / Section 2 : § ŞSaeki etal ĚŤ : IAV of Şthe diagnosed source is smallŤ : how small is small ? Be more precise

P6 : Şusing a network of stations that remain constantŤ : About that a nice solution was found in Rodenbeck et al (ACP-2003) for CO2 inversion. He starts a new inversion each time a station (or a group of stations) appears in the network. This is a nice solution to use all available information without introducing possible IAV bias. You should mention this study.

P7 : 11.25° resolution in longitude is crude. Is it because you apply a zonal correction ?

P7 : the vertical mixing in your model seems quite crude. How does the model behaves for large scale advection (Transcom experiment ?) and vertical profiles representation ? Do some comparisons with other models or data have been made ?

P8 : It seems you use GCM winds. How do they compare with analysed winds ? You should mention such a comparison as the use of analysed winds is more the up-to-date option now.

P9 : Selection of data : the graphical density of network is nice, but could you give the typical size (mean ?) of each network in the text.

P11 / Section 6 : Ş569-573 TgCH4/yrŤ This is a pretty tiny range compared to previous studies. Is it because you need to perform extra sensitivity tests ?

P12 / section 6.1 : comment on the phase lag in fig. 8b

From section 6.2 : Şfixed networkŤ represent the two cases with selected & half ? Please clarify.

P13 : How many Tg is 1% ??

P14 : section 7.1 : Why applying a 5)month running mean ? Is it enough to remove all seasonal influence ?

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P14 : OH influence. You test different OH distributions but OH does not seem to vary interannually (it is not so clear to me). So it means that OH (spatial & seasonal) distributions do not influence much your results (probably because a surface network is used with OH being maximum in the mid/low free troposphere, what do you think ?).

Section 8 : be more synthetic with introduction (see general comments).

Conclusion : Şatmospheric transport may be a factorŤ : what about inverse methodology used ??

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