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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Technical Note: Methods for interval constrained atmospheric inversion of methane" *by* J. Tang and Q. Zhuang

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We sincerely thank the anonymous referee for the insightful comments to help us improve the technical note. Our specific responses following each comment are documented below.

Comments: *This technical note deals with sequential schemes for atmospheric inversion. Atmospheric inversion seeks a vector sources and sinks $\vec{s}$ (in the following, for convenience, I write just "sources" or "fluxes") that are consistent with a vector of concentrations (in the present case of methane) given an atmospheric chemical transport model* $\mathbf{H}$:

$$\vec{z} = \mathbf{H}(\vec{s}) \tag{1}$$

The authors present an algorithm (including its implementation) that restricts the components of the source vector to intervals specified by the user. The presentation of the study is very weak. A few examples are highlighted in the specific comments. It appears that the study and the manuscript were produced in a hurry. As a consequence of the weak presentation, much of what follows below had to be guessed as good as possible from the presented material and the references.

In their approach the authors make several simplifications. First, they treat $\mathbf{H}$ *as a linear function (their Eq. 1). This is a simplification, because the strength of the sink processes is proportional to the concentration. Second, they aggregate the sources to only a few components ("big regions"), for which they compute basis functions. This is a simplification, because the space-time structure within the "region" is prescribed although it is uncertain. Third, they divide the inverse problem into a sequence of sub problems, which they solve one after another. Each sub problem uses one month of data to constrain six months of sources. The sequential scheme is set up such that the final estimate for each month of sources is influenced by six months of concentrations (from the same month plus the following five months). This is a simplification because a source affects all future concentrations. Bruhwiler et al. (2005) (Fig. 7) demonstrate that the error of this simplification can be as large as 1GtC/yr even for a linear problem and when using a "big region" non sequential setup as reference. I wonder how large the error gets in the case of a non-linear model (as here), and when benchmarked against a reference inversion that solves for the sources on the model grid. In the linear case of Bruhwiler et al. (2005) one knows (see, e.g., Enting, 2002) at least that, without imposing any error, one can set up of a sequential inversion scheme, in which each inversion step uses a sub set of the concentrations, provided that each step estimates all sources and that each step uses as prior the posterior estimate of the previous step. This does not hold in the non-linear case treated here. The authors choose a particularly favorable reference setup, namely a big region setup that is linearized in the same way as in their experiments and produce their pseudo concentrations with this setup. Hence, we don't know how the suggested method deals with concentrations* *that are affected by the non-linear sink processes or with source components that are unresolved by the big region setup.*

Our reply: We agree with the reviewer's comments that a series of assumptions have been made in order to make the problem tractable. We below try to justify these assumptions point by point.

First, for the linear assumption, we argue that it appears reasonable for an aggregated inversion of atmospheric methane. In our setup for the forward transport of atmospheric methane, the only atmospheric sink to the methane concentrations is from the OH oxidation (and stratospheric loss is prescribed similarly as the surface fluxes at the tropopause). Following other studies, this reaction is represented by a second order Stoichiometry

$$CH_4 + \cdot OH \rightarrow CH_3\cdot + H_2O \tag{2}$$

We did not consider the consumption of OH by CH_4, considering the lifetime of OH is much shorter compared to that of CH_4. Therefore, the CH_4 destruction rate by OH is linear with respect to the OH field, which is derived from an O_3-NO_X-NMVOC simulation (Fiore et al., 2003). Other sources and sinks in the setup are treated as parameters needing optimization through inversion. It seems that the only source of nonlinearity is the transport process. There could be chances for a weak flux not to be detected at a site where air samples were collected. If this flux is strong enough, its signal can be detected at the site. Aggregating into big regions (the second point raised by the reviewer), as done in other studies, e.g. in Bruhwiler et al. (2005) for CO_2, Houwelling et al. (1999) and Wang et al. (2004) for CH_4, as well as in this study, could to some extent avoid such nonlinearities. Aggregating grids into big regions will result in some errors (e.g. the pattern error) that cannot be improved in the later inversions. This does not happen in this Note, because all the prior and true fluxes are specified for big regions. Our results are thus regularizing the poor inversion that is simply a result of ill-posedness because of the dimension mismatch between fluxes and observations.

Since the problem we are dealing with is approximately linear, assimilating the observations either sequentially or non-sequentially should produce identical results, if all the fluxes are inverted simultaneously. We did check this in our code for linear batch inversion. However, even the linear batch inversion resulted in some unphysical values as we showed in Tang and Zhuang (2011) and also in the revision. There in Tang and Zhuang (2011) we also found that using a fixed lag inversion, such as the linear Kalman smoother could even result in better results compared to linear batch inversion. Therefore, we argue that the unphysical values are not because of our assimilating the observations sequentially or because only a fixed length of state variables are optimized in assimilating one month of observations, rather it is the nature of the problem.

Ideally, a grid-based inversion should be attempted. We did try this using the 4D-Var approach, similar to the study by Merink et al. (2008). It was found, for a high resolution inversion, strong correlations should be imposed between the different grid cells in order to obtain reasonable results. Often the parameters to specify the spatial correlations are obtained by trial and error or by reference to a well- setup linear batch inversion, such as done in Merink et al. (2008). Even if a high resolution inversion is available, aggregating the grid based fluxes and their uncertainties to big regions still show comparable uncertainties compared with linear batch inversion with big regions. Therefore, the big-region inversion discussed elsewhere and in this note is still useful for quick and meaningful assessment of the global methane budget.

Comments: *Another weak point is the diagnostics. The authors don't show a single posterior error. Hence, we don't know, how much adding the interval constraint reduces the posterior error. The references to Tang and Zhang (2010) are often not very helpful, because parts of the text are relatively similar, sometimes even identical. For example, understanding the details of the experimental setup is difficult if not impossible. Inspecting the corresponding subsection in Tang and Zhang (2010), one learns that "seasonal fluxes" denotes monthly resolution of the fluxes. But one has no idea how the grid cells are grouped to form the "11 seasonal fluxes" and "7 yearly constant* *fluxes", and how the sink processes are treated. Except that some lines above "stratospheric fluxes" are mentioned. For the general setup of the inversion scheme, one can just guess that it is based on the scheme of Bruhwiler et al. (2005). The results section (3) then mentions a "lag length of 6" without any unit. For the calculation of the matrix dimensions in the experimental setup section of Tang and Zhang (2010) one can just guess that the lag might be 6 months. The manuscript certainly contains some innovative material, but, as it stands, it has only a limited scientific significance.*

Our reply: We revised the graphical demonstration of our results and improved the language to make the presentation clear and succinct. Also we removed duplicate materials that have been presented in Tang and Zhuang (2011), so that more spaces are left to present the discovery in this note.

Comments: *Intro, p19982: The way the Kalman smoother refs are contrasted with the previous refs to Enting and Gurney et al. suggests that only the Kalman refs are based on Bayes' theorem/the theory described by Tarantola. This is not true.*

Our Reply: We corrected this misleading citation by referring to Tarantola as an example.

Comments: *Intro, p19983: The authors stress that the variable transform complicates the use of their Kalman smoother approach, because it increases the non-linearity of* $\mathbf{H}$*. The authors claim that the variable transform method poses problems regarding the interpretation of the posterior uncertainties. In fact, at least for monotonic transformations, this interpretation is straight forward: For example, the* $\pm$ *1* σ *range in the* ξ *space is transformed into an interval in the s-space which corresponds to the same probability.*

Our Reply: We agree with the point of the reviewer. However, in order to make a meaningful comparison with inversions that solves the problem in the space of state variables (e.g. the linear batch method), the variable transform should be avoided if other approaches are available. The variable transform technique would transform the

easily interpretable $\pm\sigma$ uncertainty into some bounds. Such bounds vary depending on the specific variable transform method used, which makes a comparison with other approaches cumbersome.

Comments: *Methods (2.1, l 11): "Combine the term ... into the measurement". Which mathematical operation does "combine" denote? Addition? Multiplication?*

Methods (2.2, Eq 4): What are $\mathbf{Q}$ *and* $\mathbf{R}$*?*

Methods (2.2): The presentation of the iterative procedure is seriously confusing. What is the "active set method", "Zigzag", "anti cycling"?

Our Reply: We clarified these points in the revision.

Comments: *Methods (2.3, text after Eq 20): What is the effect of the truncation in the eigenvalue spectrum on the posterior sources and posterior error. The truncation removes the leading eigenvectors of the inverse of* $\mathbf{Q}$*, which enters Eq 18. It is that part of the source space which is only weakly constrained and large adjustments can occur. The procedure appears to suppress these adjustments.*

Our Reply: The idea comes up from the reduced rank Kalman filter. The truncation is employed to make the numerical solution stable. Because with a sub-optimal sampling, the covariance matrix is usually rank deficit, and the least eigen values of $\mathbf{Q}$ appears more likely to be numerical noise rather than meaningful signal. Ideally, this should not happen if a sufficient large ensemble is used for a well-posed problem.

Comments: *Methods (2.3, top paragraph of p 19988): ICMLES has not be introduced. What is a "truncated multi-dimensional Gaussian distribution"? Our Reply: We introduced ICMLES before presenting the acronym. We also explained what a "truncated multi-dimensional Gaussian distribution" is. Methods (2.5): This section is particularly difficult to follow. Why is the shape of* $\mathbf{Q}^{-1/2}$ *rectangular, with dimensions* $m \times n$*? What is the instrumental distribution? Use capital letters for "QR" in "QR factorization".*

Our Reply: We set $\mathbf{Q}^{-1/2}$ as rectangular because there are cases that we can only afford sub-optimal sampling of the state space, which will result in the number of samples is less than the dimension of the problem. The instrument distribution refers to the distribution used to generate random number to test the rejection and accept ion criterion in the rejection sampling step. We used capital letters for “QR” in “QR factorization” in the revision.

Comments: *Results (Second paragraph): I presume the authors want to say that the problem is “illposed”. The regularization through the prior should actually render the problem “wellposed”. Without the regularization the ill-posedness might also be a consequence of the sequential treatment, because 1 month of concentrations (at 211 locations) is used to estimate six months of fluxes over 18 big regions, i.e. 6 x 18 flux components. By contrast, in a non-sequential inversion the ratio of concentrations to unknowns would be 211/18, which looks less “ill-posed”.*

Our reply: Since the problem is assumed linear, by theory the solution should be same if the same number of observations are assimilated, either sequentially or non-sequentially. We showed in the revision, and also in a previous study (Tang and Zhuang, 2011), that the problem we dealt with is by nature ill-posed. Even with the linear batch inversion we still obtained unrealistic values.

Comments: *The authors are using the term “correlation of fluxes”, when they actually mean “correlation of the error (or uncertainty) in fluxes”, e.g. in “unrealistic negative values of fluxes are inferred due to some spurious correlations among the different fluxes”. They also switch between the use of “posterior fluxes” and “inverted fluxes”. I would recommend to use the former term throughout.*

Our Reply: We made the terminology consistent in the revision.

Comments: *p 19982 l 2: replace “including” by “i.e.”, since here you are listing all methods that you are testing.*

p 19987 l 6: “in terms” instead of “in term”

p 19988 l 4: “for the next update” instead of “for next update”

p 19988 l 9: “after initialized” instead of “after being initialized”

p 19991 l 19: “show” instead of “showed”

Our Reply: We incorporated the reviewer’s suggestions and corrected typos and improved grammars and languages in the revision.

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