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Interactive Comment

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

T. M. Butler et al.

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Opening Remarks

While both of the referees made positive overall comments about the paper, both also had many specific comments to make and suggested many changes. In most cases we agree with the comments and suggestions from the referees, and have taken them into account in the production of a revised manuscript. Below is a detailed list of responses to the individual comments from the referees. Page and line numbers refer to the numbers given in the print version as published on the ACPD website.

Response to First Referee

General Comments

The referee would like to see more acknowledgement of the limitations in the interannual variability of the chemistry in this study. This general theme has been addressed in the revised manuscript in response to a number of comments (for example the fourteenth specific comment by this referee). In addition, as suggested, the following text, which includes a reference to Dentener (2003), has been added to the introduction (page 3424 line 4): Other important effects, such as the radiative changes following the eruption of mount Pinatubo, variability in the emissions of other chemically reactive species, and other effects discussed more fully in Dentener (2003) are neglected in this study and left as topics for future work.

Specific Comments

1. The sentence beginning on page 3420 line 5 has been moved to the end of the abstract as suggested.

2. Since we do not examine the size of this effect in the present paper we agree that this text is not necessary in the paper, so we have removed the text from the beginning of the sentence on page 3422 line 8 to the end of the paragraph. We have retained the original reference to Bekki et al (1994) as a part of our literature review. This sentence has been incorporated into the previous paragraph, with a small change made at the beginning of the paragraph to accommodate the fact that the subject of the paragraph is now both 1991 and 1992.

3. See the response to the second general comment from the second referee.

4. The sentence beginning on page 3428 line 13, which was originally intended to answer the issues raised by this comment, has been changed to This is more of a problem for species with lifetimes comparable to the timescales of deep convection (such as NO_x) than it is for CH_4, although it could potentially influence the inverse result through effects on the chemical sink of CH_4 due to the OH radical. A reference to the next section, where the OH radical is discussed in more detail, has also been added to the paragraph.

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5. We agree that the discussion of all of the OH distributions is not necessary in this paper, so we have removed it, along with the appropriate parts of Table 3.

6. The methodology chosen for this work was chosen to allow better comparability with the study of Law and Vohralik (2001). At times when there are temporal gaps in the measurement record this is discussed in the text. The sentence beginning on page 3432 has been replaced with The selected network is derived following Law and Vohralik (2001). Stations with more than 12 months of missing data at the start or end of the period were not used.

7. The following sentence is added to page 3435 line 7 by way of explanation The diagnosed CH_4 emissions fall within such a small range due to the similar CH_4-reaction weighted averages of the OH distributions used (Sect. 4), and the fact that the source and sink of CH4 are approximately in balance.

8. The reviewer is correct that we have already noted the (lack of) difference in inter-annual variability between the OH runs. We don't need to make this point again, so we have removed Figure 13a and most of the text from the first paragraph in Section 7.1.

9. The figure and text to which this comment refer have been removed from the manuscript as recommended in the last comment from this referee.

10. We would not go as far as the referee in interpreting our Figures 14 and 15. For example, the selected network case in 1998 shows colocated concentration and flux anomalies north of 60 N (Figure 15). We also remark (page 3441 line 2) that the lack of significant flux anomalies south of 30S (which we know to be implausible because there are no significant sources of CH_4 so far south) despite the presence of concentration anomalies is "highly encouraging for our admittedly simple inverse technique".

11. We have added "for a given year" to page 3442 line 28 to make this sentence clearer.

12. The referee correctly points out that in most cases there is good agreement between our selected and half network cases, and that does indeed seem to contrast with the results of Law and Vohralik. We believe a better place to make this comparison is in the discussion of Figure 16, where all of the growth events are discussed together, rather than here. The differences between our study and that of Law and Vohralik are then discussed in terms of the number of stations in the network. Specifically:

    (a) The number of stations in each network is added to page 3432 line 25.

    (b) The sentence beginning on page 3446 line 7 is deleted.

    (c) The material in the paragraph beginning on page 3441 line 6 is used to begin a new paragraph.

    (d) The discussion of network size vs. regional flux anomalies as compared with Law and Vohralik (2001), including the meaning of the word "regional" is added to the end of this paragraph.

13. Due to the addition of extra material about the chemical sink resulting from other comments by this referee, the sentence beginning on page 3449 line 1 is deleted.

14. The importance of interannual variability in NO_x and NMHC emissions is added at the end of the conclusion section.

15. The following sentence is added to page 3431 line 12: The distributions used here also lead to proportionally more CH_4 oxidation between 30 and 60 degrees north compared with Lawrence (2001).

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16. There is no Table 11, and the reviewer can't mean Figure 11 as his comments don't apply to that. We need more information if we are to respond to this comment.

17. The next two comments are related. The reviewer rightly points out that the selected and half network cases produce a methane flux in 1998 which is more consistent with Dlugokencky (2001). We hesitate to add yet another figure to what is already a very large paper, especially since the dependence of the inverse results on the concentration boundary condition has been mentioned a number of times in this paper. In particular, section 6.3 makes this connection, and has a forward reference to section 7.1 where the differences between the global flux diagnosed from all networks for 1998 is discussed. Text is added to the end of section 7.1 to make the connection with Dlugokencky (2001) stronger.

18. The caption for Figure 13 is changed as requested.

Technical Corrections

All technical corrections requested by the referee have been made.

Response to Second Referee

General Comments

1. The referee rightly points out that the introductory sections are unnecessarily long. Section 1 has already been shortened as part of the response to the second specific comment of the first referee. To shorten and make more synthetic Section 2, the two paragraphs beginning on page 3425 lines 14 and 23, which describe what Saeki (1998) and Law and Vohralik (2001) have done are deleted. The reference to both studies is preserved by adding a new introductory sentence to the following paragraph, which is where the important results are summarised.

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2. The referee makes several valid criticisms of the mass balance inverse methodology employed in this study, and suggests that we focus more on these, while also mentioning other methodologies. The referee specifically mentions Kalman filtering as an alternative. We believe our original manuscript failed to make the point strongly enough that our study is intended to help lay the foundation for future analyses using models containing the full suite of chemical interactions, for which we believe the mass balance method is best suited, rather than a traditional inverse modelling study which relies on linearisation of the chemistry. This issue is also related to the general comments and the third specific comment from the first referee. We believe we have addressed this comment with our substantial reworking of the first two paragraphs of Section 2.

3. We did not look in detail at the effects of individual stations on the inverse result, although we agree that this would be very interesting. We have added the following text to page 3449 line 13: Comparison between this study and Law and Vohralik (2001) seems to suggest that a network of about 20 stations may be appropriate for resolving flux anomalies at semi hemispheric scales. An interesting topic for future work would be to explore the degrees to which different stations influence the inverse result. This also refers to new text added as part of the response to comment 12 from the first referee, which is relevant to this issue.

4. The referee also comments on our figure scales, suggesting that they should be the same in some cases to allow better comparison. We believe that our choice of scales best reflects the variability existing in the data, and that the reader is capable of comparing the relative magnitudes in each case. We also note that the first referee makes no such suggestion. We have, however drawn attention to the different scales used in Figure 9, Figures 14a and 15a, Figures 14b and 15b and figure 16 by adding notes to page 3436 line 7 and the caption of figure 9, page 3440 line 14, page 3440 line 17, and the caption to Figure 16 respectively.

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Specific Comments

1. As part of the response to the general comments from this referee, most of the text discussing Saeki (1998) has been removed from the paper.
2. Rodenbeck (2003) uses the synthesis method, not mass balance as used here. The details of their methodology are not relevant to this study.
3. Low resolution is used due to the computational expense of running the model.
4. The comment about the vertical mixing has been addressed in response to comment 4 from the first referee. The comment about the large scale advection has been addressed by adding a reference to the transom experiment to page 3428 line 4.
5. The sentence beginning on page 3429 line 17 is modified to read: The use of annually repeating dynamics from a climate model rather than analysed winds means that the effect of interannual variability in the transport can not be explored.
6. Information about the size of the measurement networks used has been added as part of the response to comment 12 from the first referee.
7. This point has already been addressed in the response to comment 7 from the first referee.
8. The following text is added to page 3435 line 15: The shift in the northern midlatitude emissions maximum seen in Figure 8b is due to the effect of extra measurement stations coming online in the full network during the 1990s, as seen in Figure 4a.

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9. The text “(selected and half network)” is added after “fixed network” on page 3436 line 20.
10. “about 1%” is changed to “6 Tg, or about 1% of the annual CH_4 flux” on page 3438 line 4.
11. Our choice of the 5 month running mean was somewhat arbitrary. The unsmoothed monthly data is very hard to interpret visually, and the 5 month running mean seemed to be the smallest averaging period which allowed interpretation. It was not our intention to remove all seasonal influence.
12. Changes made to the manuscript during the review process have hopefully made it clearer that the influence of OH on the inverse result is a future perspective of the present study.
13. We believe the introduction to Section 8 lays a good foundation for the analysis which follows in Subsections 8.1 to 8.5.
14. We have already mentioned atmospheric transport in the conclusions (page 3449 lines 27 to 29), and clarified our discussion of inverse methodology as a response to the second general comment from this referee.

Other Changes

As a result of private communication, two further changes are made to the revised manuscript.

1. To help the reader find the document, the URL for Law and Vohralik (2001) is added to the references.
2. A missing acknowledgement is added.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 3419, 2004.