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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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General comments

The paper reports on a mass balance inversion of methane emissions during the 1990s. The latitudinal distribution, seasonal cycle, and interannual variability are studied. A number of periods with anomalous growth rates are explored in detail and deduced fluxes on global as well as semi-hemispheric scales are related to the existing literature.

The authors have produced an interesting and clearly written paper. I particularly liked the thorough analysis of the anomalous growth rate events and the comparisons of retrieved flux anomalies with previous studies.

It is known that the results of a mass balance inversion depend critically on the surface

concentration boundary condition that is used. The paper shows that the introduction of new measurement stations can lead to spurious changes in this boundary condition, and hence in the retrieved fluxes. A fixed network is thus preferable to a network that changes over time. Interestingly, half of the 'selected' network used in this study already appears sufficient to deduce the fluxes accurately on the spatial scales investigated.

The main weak point, which is indeed acknowledged by the authors a number of times, is that all interannual variability (except that of the CH_4 concentrations and fluxes) is neglected. This includes:

- variability in the wind fields, affecting transport of CH_4. The authors refer to Dentener et al. (2003) to defend using annually repeating meteorology. Indeed, Dentener et al. found that methane fluxes were more sensitive to OH fields than to transport (and even much more to the concentration boundary condition), but this conclusion was only drawn for the global scale. On semi-hemispheric scales, the effects of transport might be more important. The authors rightly cite Warwick et al. (2002), who showed that variability in transport alone can explain large parts of the interannual variability of methane observed at surface stations.
- variability in OH, affecting the sink of CH_4. This variability can be caused by variations in water vapour, emissions of CO and other ozone precursor gases, stratospheric ozone, stratospheric aerosols, etc. Neglecting this variability is a serious limitation. In a number of cases (e.g., the post-Pinatubo period) changes in the sink have certainly played an important role. Note that variability in OH is also partly caused by variations in transport of CO. The authors might consider adding a reference to Dentener et al. (JGR, 108(D15), 4442, 2003), who investigated the role of many of the above factors in determining interannual variability (and trends) of OH.

While the authors' choice to neglect all this interannual variability should be respected,

ACPD

4, S1293–S1298, 2004

they should do a more careful job in making this clear to the reader. Therefore, it is recommended to add comments in the introduction section, and preferably also in the conclusions section, explaining the limitations of the present approach.

Specific comments

P3420, L5-7: A more logical place for this line would be at the end of the abstract.

P3422, L5-22: It appears that the authors want to argue that the effect of stratospheric ozone on OH and CH_4 growth rate is probably smaller than suggested by Bekki et al. (1994). However, their discussion contains a number of irrelevant statements. The role of tropospheric ozone (L98-14) is minor, multiple scattering (L17) is a second-order effect, and the enhanced stratospheric aerosol loading (L17-18) is only important in the first months after the Pinatubo eruption. It is recommended to remove these lines from the text. The authors should not try to reason away the OH effect, but make clear that they are not looking at it in the present paper.

P3425, L10-13: This is *not* the focus of the paper. The focus is to explain observed variations in methane concentrations in terms of changes in its sources. Morever, since the OH field is kept constant in this study (except for the feedback of CH_4 on OH, but this is shown to be completely unimportant for interannual variability), the possibility to take into account the non-linearity of chemistry is not an argument for using the mass balance method here. The inversion might as well have been done with the synthesis technique.

P3428, L12: Is is mentioned that the model does not represent deep convection very well. How bad is this, and what are the possible consequences for the inversion?

Section 4: What is the point of a detailed discussion of all these different OH fields? In the remainder, only the Match and Spivakovsky distributions are used, but these have very similar OH averages (table 3). Since the OH-1, OH-2 and OH-3 distributions are not applied in inversions, they might as well be removed from the paper.

ACPD

4, S1293–S1298, 2004

P3432, L14-17: Why have not all stations with a 12-month gap *somewhere* during the period been discarded?

P3435, L4: I am surprised that the deduced average yearly CH_4 flux is virtually the same (within 4 Tg) for all five runs. How can this be understood? Or is it a coincidence?

P3439, L16-19: The fact that the OH runs give the same results concerning interannual variability is indeed no surprise (as the authors already state on P3438, L8-14). This should be noted again, as it could easily lead readers to think that OH is not important for interannual variability. (Another option is to delete figure 13a altogether)

P3439, L24-25: This statement does not seem to be supported by figure 13a. The 1998 peak in growth rate appears to be closely matched by a peak in the flux. Deviations at other times are much larger. Do the authors agree with this observation?

P3440, L23-28: This finding seems to be much more general: *none* of the concentration anomalies south of 30°S and north of 60°N appears to lead to significant anomalies in the fluxes. If the authors agree with this observation, could they discuss its consequences in the paper?

P3441, L27 - P3442, L1: Since the sum of (linear) trends in a number of time series equals the trend in the sum of those time series, I do not understand why "the sum of the flux anomalies from each semi hemisphere is not always equal to the global flux anomaly".

P3446, L8-9: The authors refer to Law and Vohralik (2001) who found that "small fixed networks are not capable of resolving regional flux anomalies". However, this contrasts with the present results for the other growth events, where the half network gives very similar results as the selected network. This indicates that a small network is capable of resolving fluxes on semi-hemispheric scales. Perhaps the word *regional* is confusing here: I would associate it with smaller than semi-hemispheric scales. Indeed, one would expect a small network to become insufficient when going to such smaller

ACPD

4, S1293–S1298, 2004

Interactive Comment

scales. What did Law and Vohralik mean with *regional*?

P3449, L1-2: Again, the statement on OH is confusing. Although the chemical sink is computed interactively, the only driver for interannual variability in OH is the CH_4 concentration.

P3450, L9-13: It should be noted that not only CO emissions but also those of NO_x and non-methane hydrocarbons are important for OH. Interannual variations in these emissions should also be taken into account.

Table 4: There is a large discrepancy between the values presented here and in Lawrence et al. (2001). The most striking difference is that in table 4 about the same amount of CH_4 is oxidised in the 30°-60°N region compared to the 0°-30°N region, whereas in Lawrence et al.'s figure 4 the oxidation in the 30°-60°N region is only 1/2 to 1/3 of that in the 0°-30°N region. Could the authors explain this discrepancy?

Table 11: The results in table 11 (for 0°-30°N and 60°-90°N, selected and half networks) seem to be opposite to those in figure 16. In Fig. 16c, the half-network emission is lower than the selected, whereas in the table this is the other way round. A similar observation can be made concerning Fig. 16d.

Figure 1: The global mean CH_4 annual change shown here is similar to Dlugokencky et al. (2001) figure 2b. There is, however, a large difference in the relative magnitude of the peaks in growth rate. In particular, the present study has comparable peaks in 1994-1995 and 1998, whereas Dlugokencky et al. had a much larger peak in 1998. Can the authors explain this discrepancy?

Figure 12: It would be interesting to see the global-scale growth rate as inferred from the selected (or half) network. Probably, this shows a much more pronounced peak in 1998, thus being more consistent with Dlugokencky et al. (2001).

Figure 13: Add in the caption that the curves represent a 5-month running mean.

ACPD

4, S1293–S1298, 2004

Technical corrections

P3427, L5-6: Change to '... the interannual variability on top of the fluxes from ...'

P3432, L28: Replace 'measurements' by 'stations'

P3439, L10: Remove 'takehere we have'

P3441, L12: Change 'Fig. 13b' to 'compare Fig. 16 with Fig. 13b'

[Thanks to Michiel van Weele for his assistance in making this review.]

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