Atmos. Chem. Phys. Discuss., 7, S4286–S4289, 2007
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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The influence of biogenic emissions on upper-tropospheric methanol as revealed from space" *by* G. Dufour et al.

Anonymous Referee #2

Received and published: 22 August 2007

General Comments

The paper by Dufour presents a step forward in utilising ACE data on organic compounds to look at the methanol content of the middle to upper troposphere. Previous model studies comparing models to (infrequent) aircraft data have reported large discrepancies and therefore, logically, it would be expected to be instructive to use satellite data to examine behaviour over wider regions. This should tell us whether problems are endemic or not. Therefore I would support the authors study as one of value. Unfortunately the present paper does need to go further in characterising and diagnosing the differences between model and data. There are some strong differences between the data and the model both in terms of mean values and variability, and the authors should not be afraid to say so.

There are 3 steps which I think are very necessary for the results of the paper to be interpreted: 1) the error analysis needs to be defined and explained better. What is the meaning of relative and absolute error in this context and what are the sources of error which fall into each category. For example, what is meant by the relative measurement error? 2) the emission sources and their distribution in the model need to be explained better. How are the source strengths distributed through the year, and how and where are they injected into the model? Ocean source/sink relationships for methanol (e.g. Sinha, ACP, 2007 are not discussed at all). 3) the differences between model and data (e.g. lack of correlation) need to be explained in much more detail. Are these likely to be due to differences between model source distributions and "real" source distributions, specific dynamical transport problems/limitations in the model, or are they expected given variability in the data or the models. The authors statements of "fair agreement" and "agreement within 50% for 50% of measurements" imply that really the model and data do not agree well.

Amplifying point 3) further, the authors should use their knowledge of the error sources in the data and the distribution of source emissions to further investigate and confirm the differences between the model and the data. For example, the authors should comment fully on Table 1 and the lack of correlation between data and model. Are the two really different? Perhaps this is real and if so the authors should say this! Alternatively it could be just due to model dynamical transport or the model distribution of emissions in the vertical. Why are the correlations between data and models better in MAM2004 (North Pacific) and SON2004 in Europe-Asia-Africa?

In Figure 4, there are considerable differences between latitudinal and vertical structure which need comment (SON2004 has a peculiarly strong gradient at the Equator in the model data - is this just a gridding problem?) and these are also visible in the map comparisons in Figures 5 and 6. For example, model SON2004 has a large polar increase compared to model JJA2004 and this is apparently larger than that in the ACE data where the high values occur sporadically at latitudes which are further south rather than at the highest latitudes. Furthermore, much of the vertical and latitudinal structure is different between data and models, according to Figure 4, and I think the authors should comment on this in much more detail.

The question of distinguishing biomass burning emissions from biogenic emissions is an important one since it may result in higher mean values for ACE data and a different longitudinal distribution at a given latitude, as appears to be the case in Figures 5 and 6. The differential sampling of biomass emissions caused by dynamical transport differences between the model and the real atmosphere may bias the mean comparisons. Also biomass burning has a seasonal cycle or at least may be strongly enhanced in high Northern latitude summer, depending on the year. The authors need to provide stronger evidence of the biogenic as opposed to biomass explanation.

Looking at Figures 5,6 and 7, I note the the model appears to have more methanol in MAM than do the data, which is surprising given the apparent lifetime of methanol in the model. Please provide an explanation of this.

Finally, I notice that there is a peak in the JJA methanol data in the upper troposphere over Europe-Asia but no sign of a model peak (Figure 7). Do the authors have an explanation for this? Could this be a biomass peak or long-range transport influence and does HCN or CO data help here? Please provide an explanation of this.

Specific comments

Specific areas where corrections should be made:

1) Abstract: "Fair agreement" and "50% of the observations reproduced... within 50%". A better statement would be that the model agrees well in particular regions and does not in others.

2) Section 2. Please provide some more details on the spectral windows. Which are the chief contaminants? Also please define relative, absolute and measurements errors, and list sources/magnitudes of errors which contribute to each set (see comment

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above).

3) Section 3. Please provide more information on the distributions (spatially and in time) of emission sources and their injection into the model. A concise summary of key points from Lathiere 2005 woould suffice for plant emissions. More detail is required for the biomass and urban sources. Why is the biomass source so low compared to the statement in section 1 and to some literature estimates, e.g. Holzinger et al, ACP, 2005? What is the definition of wildfires? The urban source seems quite small. Is this related to biofuels which play a large role in Asian emissions? How are the biomass and urban sources distributed in space and time. Finally, as noted above no mention is made of an oceanic sink for methanol, e.g. Sinha, ACP, 2007.

4) Section 4. "Overall, the model overestimates the satellite observations by 20% in the March 2004 to August 2005 period in the upper troposphere". I am concerned about this statement since the low number seems to arise simply from cancellation of errors of different sign. What is the mean of the absolute mean differences? Please justify this statement if you think it is useful.

5) In Figure 4, I am concerned about the low numbers of occultations in some of the latitude boxes. I think the number cut-off for an empty box should not be zero but perhaps a number like 20 to have reasonable longitudinal sampling on more than one day. Please consider using a number greater than zero and provide some rationale for it. This may lead to more empty boxes on the plots but might be more realistic.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 9183, 2007.