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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.

G. Dufour et al.

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The authors thank the referees for their interest in the article and their suggestions for improvements. The comments made are addressed below.

One major point has been underlined by the both reviews: the lack of details in the discussion and interpretation with the model. The authors agree that major improvements have to be done in the description of the sources and sinks of methanol used in the model and in the discussion when ACE data are compared to model simulations. Nevertheless, the authors would like to precise that their main goal with this paper was first to date with a new interesting data set never measured before and provide some insights of the way of interpreting it with a model. The authors are then completely aware that a lot of questions stay opened with this paper and that some of these questions might be difficult to assess. However, the authors would like to thank the referees

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again for this important comment that will help them to greatly improve the value of the paper with the addition of a more detailed discussion and possible interpretation of the disagreement observed between model and measurements.

In the following, the authors will answer to each referee separately.

Reply to Referee #1:

Reply to the major comment #1: The authors have taken a stand to use the latest known biogenic emissions of methanol (Jacob et al. 2005) as departure point of the comparison between simulations and observations as well as one of the latest fire emission cadastre in order to use "state-of-the-art" entries for the model and then test our current knowledge. However, the authors agree that the results have to be discussed more carefully and the large uncertainties in the emissions in Jacob et al., 2005 have to be mentioned to help readers to understand the limitations and the difficulties of the assignment of emission magnitudes. The authors also note that their derived lifetime is in the high side of the range found by Jacob et al. but is in the middle range of lifetimes given more generally in the literature and summarized by Jacob et al. in their Table 1. A better description of the emissions and sinks considered in the paper in comparison with other studies will be implemented in the revised version of the paper.

Reply to the major comment #2: The authors have written p9188 line 2-3 that "For comparison with the ACE occultations, the simulated daily averaged methanol profiles are interpolated to the measurement locations". The revised version of the manuscript will precise that the interpolation is bilinear in latitude and longitude and that only the days with measurements have been considered. This statement will also be added to Fig. 4 for clarity.

Reply to the major comment #3: The authors chose to show typical individual occultations for 2 reasons: 1- In order to show that even without any averaging the errors are good accounting for the difficulty of detecting methanol in the spectra. 2- The authors

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haven't calculated the errors for all the occultations between March 2004 and August 2005 because the procedure to characterize the errors is a perturbative method and is then time consuming (more than 6 months would have been necessary to process this error study). By displaying the vertical profiles of errors at 5 very different locations (in term of methanol profiles as well as latitudes and seasons), the authors aim to cover the range of possible errors in the occultation discussed in the paper.

Reply to the major comment #4: The discussion is apparently not enough clear in the paper: the predominance of the biogenic emissions in elevated methanol is only true for the northern mid- and high latitudes during spring and summer. In the tropics during the SON season, the large values observed are attributed to biomass burning sources (see the beginning of the last paragraph of the discussion and Dufour et al., 2006). However, to prove that the large values observed in the northern high latitudes during summer are mainly due to the biogenic sources and not only to boreal fires taking place in these regions in summer, the authors will provide an additional figure showing the ratio of the methanol vmr coming from sources except biomass burning. The biomass burning part will be derived using the emission ratio with respect to CO given by Andreae and Merlet, 2001 for extra tropical fires. First estimates show that about 80% of the measured methanol at 8.5 km come from other sources than biomass burning (in agreement with the partition of the total sources of methanol: biogenic sources represent more than 60% of methanol against only less than 10% for biomass burning). Additional figures showing emissions by regions and their seasonality will also be added to show the predominance of the biogenic source for the northern latitudes.

Concerning the comment on the vertical transport, many comparisons with tracers such as Radon, CFC, Krypton or SF6 are presented in the paper from Hauglustaine et al. 2004. The model was shown to be of equivalent quality as other global model in representing both horizontal large scale and vertical transport. This means that no evidence of a permanent bias or strong disagreement was found when comparing with measurements but, obviously, the vertical transport remains subject to large

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uncertainties in such global models in particular regarding large scale convection or stratospheric intrusion. This study of Hauglustaine et al. 2004 also pointed out that the model is too diffusive which is visible in Fig. 4: the gradient across the tropopause is smaller than those observed and that may explain a part of some disagreements observed. The authors will insist on the other sources of uncertainties in the global LMDz-INCA in the next version of the manuscript.

Reply to the minor comment #1 The term "medium" will be removed.

Reply to Referee #2:

The authors agree with the referee that the comparison between the observations and the simulations has to be discussed in more details. The interpretation of the differences with regard to the uncertainties in the retrieved observations (that remain large for small methanol vmrs) as well as in the model (emissions, transport), will permit to raise the real discrepancies between the model and the observations and to provide more convincing conclusion concerning the underestimate of the biogenic source of methanol.

Reply to the general comment #1: The meaning of relative and absolute errors in the context of the paper is the common one: the absolute error is the absolute value of the error (in pptv) and the relative error is the ratio between the absolute value of the error and the corresponding vmr value. This will be clarified in the text. Concerning the description of the characterization of the error, the authors refer to a previous paper in which the method and all the parameters leading to errors in the retrieval have been detailed (Dufour et al. 2006, ACP). However, the authors agree that a summary of the procedure and the different sources of errors is necessary in the paper to help the reader. A small paragraph will thus be added.

Reply to the general comment #2: A detailed description of the sources and the sinks of methanol considered in the model will be added to avoid the reader to read all the references cited concerning the model. The authors will also add a detailed description

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of the budget for the main continental regions as well as the seasonality of the emissions. Concerning the ocean source/sink, the contribution of oceans to the budget is rather small according to Jacob et al. 2005 (less than 5%) and the more recent paper by Sinha et al. 2007. The fact that ocean is a source or a sink is still unclear, even if the paper of Sinha tends to demonstrate that ocean is a sink for methanol. As at the time of the study, this point was still under discussion, this sink is not included in the model. Only the dry deposition on ocean can occur in the model.

Reply to the general comment #3: As already mentioned at the beginning of this reply, the authors fully agree with the referees that it would be gainful for the paper to discuss in more details the comparison model/observations and thank the referee for his useful comments. The paper and the Table 1 will be reorganized to fill the recommendations of the referee and to have a more constructive discussion part. In particular, attention will be given to interpret the data and the simulations with regard to their respective uncertainties (retrieval errors and large emissions uncertainties). For Table 1 (and Fig. 7), the regions considered will be adjusted to avoid overlap and also the stratospheric part will be suppressed in Table 1 for clarity in the discussion and also to be able to add more information about the statistics given for the upper troposphere.

Reply to the general comment #4 about the Fig. 4: The coverage of the ACE instruments is quite poor in the tropics leading certainly to a lack of representativity in the zonal means in Fig. 4 for some latitude bands. That is why the number of occultations considered in each average is represented with black squares (scale on the right part of the plots). For example, the strong gradient observed at the Equator in SON2004 in the model data is based only on one profile. Due to the resolution of the model, it can miss some variations observed on a finer scale. Comparisons with simulated zonal means without restricting the simulated profiles at the time and the location of the measurements should help to characterize the representativity of both the measurements and the simulations (sampled at the measurements points). The authors thank the referee to have raised this important point that has needed to be discussed

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in the paper. A note on the diffusivity of the model and more generally on the uncertainties related to vertical transport in the model will also be added to discuss the differences in the vertical structure of the profiles. Concerning the point on the large values of methanol in the highest latitudes during SON2004, the persistence of these large values are not completely understood in the model; The cycle of the emissions used seems in agreement with the ACE observations but the large values in SON (and maybe the underestimation during JJA) might result of a misrepresentation of the OH chemistry or deposition in the model. This point needs certainly to be raised in the discussion.

Reply to the general comment #5 on biomass burning: In the Northern Hemisphere, about 80% of the methanol measured during JJA comes from other sources than biomass burning if we consider the emission ratio of methanol with respect to CO given by Andreae and Merlet, 2001 (see also the reply to reviewer #1). This is in agreement with the partitioning of the sources: biomass burning represents less than 10% of the total sources. As a consequence, it is unlikely that the biomass burning could be the driver of the seasonal cycle of CH3OH at these locations.

Reply to the general comment #6 on the overestimation of CH3OH by the model in MAM According to the atmospheric sources (oxidation of hydrocarbons and CH4), a North-South gradient is expected. This gradient is not apparent in the data whereas it appears clearly in the simulations. This apparent disagreement has to be considered carefully because of the large errors that affect the smallest methanol values retrieved. Fig. 2 shows that for vmrs smaller than 250-300pptv, the associated errors can reach 80% in the upper troposphere. If we consider these errors, the discrepancy between model and observation during MAM may not be so effective.

Reply to the general comment #7 on the peak observed in JJA in Europe-Asia. If we consider the similar means but for the observed CO profiles over Europe-Asia during JJA, a peak is also observed around 8 km, likely due to biomass burning emissions. Using the emission ratio of methanol with respect to CO given by Andreae

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and Merlet (2001) for extra tropical fires, we derive that the contribution of biomass burning in methanol values is between 20 and 30% at the peak. If the contribution of biomass burning is subtracted, the difference between the altitude potentially affected by biomass burning emissions (typically 8 km) and those not affected (6km) is largely reduced. This would confirm that a significant part of the variations observed with the altitude in the upper troposphere is due to biomass burning during JJA. However, two points are important to point out: (1) the vmr values of methanol without biomass burning contribution remain large (about 1500 pptv, evidence of other sources); (2) the errors in the measurements are of the same order than the biomass burning contribution. Concerning the model, two reasons can be given to explain that this peak is not obviously reproduced. Firstly, the model seems to transport too much methanol in the lower stratosphere leading to a lack of methanol in the upper troposphere. Secondly, injection of fire products is only considered at the ground level in the model. This point will be clarified in the paper.

Reply to the specific comment #1: This will be rewritten in the light of the in-depth comparisons and discussion that will be implemented in the revised version of the paper.

Reply to the specific comment #2: As already mentioned above, a summary of the procedure and the different sources of errors in the retrieval will be added.

Reply to the specific comment #3: As already mentioned above, a full description of the sources and the sinks considered in the model plus a detailed budget by region and the seasonality will be provided to readers. The authors realize that the sentence "Biomass burning, urban activities and atmospheric oxidation of hydrocarbons are also identified as methanol sources and together contribute 27-55 Tg/year" can be confusing and let think that biomass burning is the dominant of these 3 sources. Actually, atmospheric oxidation produces 38 Tg/year of methanol against 13 Tg/year for biomass burning and 4 for urban activities (Jacob et al., 2005). With a better description of the budget this should be clearer in the revised version of the paper. Moreover, the authors are

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not convinced by the estimate of Holzinger et al. 2005 that derive the global methanol emitted by biomass burning per year from very few fire plumes measured only in one particular region (Mediterranean) and then not representative of the emissions in other fire regions.

Reply to the specific comment #4: To calculate the biases given in Table 1, the arithmetic biases for each individual occultation and altitude have been considered. The authors are aware that can lead to cancellations of errors but doing such permits to determine if the model systematically overestimates or underestimates the observations accounting for the different variability encountered in the simulations and the observations. Actually, the variability of methanol during a season for a given region will be represented differently in the simulations and in the observations (the simulations show a lower variability due to the coarse resolution of the model, see Fig. 7). However, the authors agree that this point should be mentioned to avoid misleading of the readers. This also has to be discussion with respect to the uncertainties of the retrieved methanol and of the emissions used in the model. The Table 1 will be modified in order to add the average of the absolute values of the biases and also the bias between the mean observed and simulated values for each region and period.

Reply to the specific comment #5: The problem of representativity of the zonal mean has already been discussed above. The authors will consider the suggestion of the referee and look forward a better way to present these results.

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