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Atmospheric Chemistry and Physics Discussions

Interactive comment on "First space-borne measurements of methanol inside aged tropical biomass burning plumes using the ACE-FTS instrument" *by* G. Dufour et al.

G. Dufour et al.

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Reply to Referee #2:

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

Comment 1: "Abstract and page 3951, l 18: I'm not quite sure about the use of the term "reliable profiles". From the text, ... Also, application of this criterion to all measurements would leave very few points in particular for background situations but also at the lower end of Figure 5. Please clarify what has been done."

The sensitivity study on methanol retrieval shows that the relative error exceeds 100%

above 16.5 km (on average). Therefore, in the current study, we consider retrieval results for methanol (and CO in Fig. 5) up to 16.5 km, even for those occultations where the error exceeds 100% at the highest altitudes. Note that no background results have been presented in the figures (except Fig. 4) as this study focuses on occultations polluted by fires. The lowest vmr values in Fig. 5 correspond to the highest altitudes of the range 8.5-16.5km.

Comment 2: " p3948, l10: A vertical resolution of 3-4 km is given in the text. It is not clear to me how this relates to the steps in tangent altitude ..."

For the occultations considered in the current study, the tangent height spacing ranges between 0.5 and 2.5 km, but the altitude resolution of the retrieved profiles is ultimately limited by the field of view of the ACE-FTS instrument, which gives an altitude resolution of 3-4 km.

Comment 5: 'p 3952, l14: How can a sampling effect change the relative altitudes of the CO and CH3OH profiles in the same measurement? Is this the effect of differences in vertical resolution or just uncertainty from the individual errors?"

The retrieval parameters are vmr values on the tangent altitude grid (i.e., a vmr value for each measured spectrum), but the retrieval process involves interpolating onto a standard 1-km altitude grid (see Boone et al., 2005). Interpolation can lead to an apparent altitude shift between the vmr peaks for the two different molecules (if one does not directly sample the peaks). The retrieval errors in CH3OH could also be playing a role. The fitting errors are large enough that the shift could simply be a consequence of the difficulty in fitting such a weak signal. (i.e., one could draw a vmr curve with a peak at a different altitude that still went through all of the error bars).

Comment 6: "p3952, l 22: How were biomass burning plume data selected? Is the selection based on CO? I assume that the entire profiles were used even if only part of the profile was affected by biomass burning?"

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Actually, the selection of biomass burning plume data is based on CO enhancement as well as other species typically emitted by fires (HCN especially), see Rinsland et al., 2005a. The entire profile (between 8.5 and 16.5 km, see comment 1) is used for an occultation identified as being polluted by biomass burning.

Comment 7: "p 3952, l25: Shouldn't the correlation coefficients for the background cases always be smaller as some error contributions are absolute errors which will increase the scatter of the data points at lower mixing ratios?"

The error will increase in the background case but the profiles for the background occultations are consistent (without any oscillations): the scatter of the data points is similar to the scatter of the data points of the polluted cases when one considers the lower mixing ratios (Fig 5). These data points have not been added in Fig. 5 for clarity (and also because we focus here on the polluted measurements).

Comment 8: "p 3953, l2 and following: if I understand the discussion correctly, two implicit assumption are made in the analysis: 1) the CO and methanol background as well as the enhancement ratio are independent of altitude and 2) all plumes and data sampled have the same enhancement ratio. While these assumptions might be reasonable, they need to be mentioned and briefly discussed."

We agree that it is interesting to discuss this point. It is worth noting that the first assumption is correct because we sample air masses in southern tropical and middle latitudes where the biogenic emissions of methanol by plants (not a major source of CO) are negligible compared to fire emission in this case.

Comment 9: "p 3952, l 9: Here it is stated that secondary production of methanol is unlikely. As this is one of the main conclusions of the paper, some more details should be given to the reader, e.g. on which primary emission ratio is expected under which conditions, how much the ratio should change per day according to model calculations if secondary production takes place and how this relates to the age of the plume(s) sampled and the uncertainty in the enhancement ratio determined."

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The main scope of the paper was to show the ability of ACE-FTS of measuring organic compounds (methanol in our case) in the upper troposphere, lower stratosphere in polluted cases with a good degree of confidence. Our initial goal was then not to focus on a possible secondary production of methanol. The altitudes accessible from ACE-FTS measurements concerning methanol range from 8.5 km to 16.5 km (for this current study) whereas aircraft measurements sound altitudes below 12 km. Air masses sounded by ACE-FTS and polluted by biomass burning emissions have experienced a longer transport (~7 days against 2-3 days for aging plumes measured by Holzinger et al., 2005) and can then experience more chemical transformation. It was then interesting to compare the enhancement ratio derived from space measurements to those derived from aircraft measurements in order to see if these ratios are substantially different when transport is longer. The comparison presented in the paper show that our results are similar to many other studies conducted by aircraft measurements. Thanks to your comment, we realize that we have forgotten to cite an important study that can help for the discussion (Yokelson et al., JGR 108, D13, 8478, 2003). In particular, this study provides an estimation of fire-average initial emission ratio for methanol, that is the average of initial emission ratio of different kind of Savanna fires (grassland + some brush and small trees, open forest with a grass/brush understory, pure grassland). The initial emission ratio of methanol is 14.4 +/- 3.0 ppt/ppb, comparable with the enhancement ratio derived from our study. This leads us to conclude that there is no evidence from our measurements that a secondary production of methanol takes place in the plumes sounded by ACE-FTS. The conclusion of the paper has to be reworded in this sense. Because our measured emission ratio agrees so well with the initial emission ratio, there is no incentive to include details concerning the expected evolution of a plume undergoing secondary production.

Comment 10: "p 3952, l 12: No indication is given where the tropopause height is in Fig. 2, 4 or 5. If the ability of ACE-FTS to take methanol measurements in the lower stratosphere is relevant here, tropopause height should be mentioned as well as the relative errors of the measurements for the biomass burning and background case."

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The tropopause height is varying in the range ˜ 9-17 km during the time period considered in the current study (see Fig. 1 of Rinsland et al., 2005a). The tropopause height for the case of the enhanced profile presented Fig.4 is 14.76 km whereas the tropopause height for the background case is smaller (10.35 km). As shown in Table 1, the relative errors of the biomass burning case is lower than 100% below 16 km and is less than 40% up to 14.5 km. For the background case, the errors on the retrieval reach more than 100% above 15 km and stay lower than 45 % below 11 km (typically between 30 and 35 % between 8 and 10 km).

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 3945, 2006.