

Interactive comment on “Global upper-tropospheric formaldehyde: seasonal cycles observed by the ACE-FTS satellite instrument” by G. Dufour et al.

Anonymous Referee #1

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This is a scientifically significant manuscript with high scientific quality. This reviewer recommends publication after the following points are addressed. Most of these points concern the scientific justifications for the present work discussed in the Introduction.

1. The last sentence in the Abstract and on page 4 (discussed below) needs rewording. The Abstract statement that the HCHO observations from the ACE-FTS instrument providing a unique data set for investigating and improving our current understanding of the formaldehyde budget and upper tropospheric chemistry; is way too strong given the stated limitations on the retrieved values. The stated error bars on these measurements are 30 %; 40% up to 9 km and exceed 100%

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above 13km, and these values become even larger for mixing ratios below 100 pptv. In addition, the stated vertical resolution is 3–4 km. As written in the Abstract and on page 4 in the paragraph before Section 2 entitled, ACE-FTS Measurements, which reads “provides a new opportunity to improve our knowledge of the HCHO upper tropospheric budget and to better quantify its role in HO_x chemistry” implies that the current measurements can successfully address these issues. This reviewer believes that although the current measurements are very valuable in providing extensive seasonal and geographic HCHO coverage throughout the upper atmosphere, the accuracy, precision and vertical limitations make it very challenging to address adequately the HCHO and HO_x budgets in the upper troposphere. To do so, requires high precision and high spatial resolution. Hence, these justifications should be toned down somewhat and instead the authors should emphasize the unique aspects of large temporal and geographic coverage.

2. In the last sentence in the Introduction, reword to read “Under low NO conditions, intermediate compounds like methyl hydrogenperoxide (MHP) may possibly be removed by deposition before reaction with OH takes place”;

3. The next paragraph in the Introduction regarding HCHO production from biogenic sources like isoprene being the dominant source has not been widely accepted. Even though satellite studies suggest this, aircraft and ground-based studies indicate that methane is still the dominant source of HCHO. This should be reworded.

4. At the end of this same paragraph on page 3 in the Introduction, reword to read “The sinks of HCHO are mainly photolysis and reaction with OH, and ultimately lead to the formation of carbon monoxide and HO₂”;

5. The next paragraph on page 3 discussing that considering only methane in the upper troposphere usually does not reproduce the observed HCHO does not square with the latest measurement-model comparisons. For example, the two papers by Fried et al. in 2008 clearly show good general agreement in the upper troposphere, even during

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some convection events. It was only during very fresh convection, where direct HCHO sources could be important, and in the presence of enhanced NO from lightning that the HCHO observations persistently exceeded the models. Also, the suggestion by Jaegle et al. regarding heterogeneous production of HCHO from methanol is only a suggestion and one that has never really been substantiated by direct evidence other than in clouds affected by biomass burning plumes. Fried et al. [2008] examined this for more pristine clouds and found no evidence for this. Thus, I would suggest rewording this section.

6. At the top of page 4, I have no problem with the statements regarding the role of convection in transporting direct HCHO and/or its precursors to the upper troposphere and that there are still some unexplained discrepancies, I do have a problem with the statement that models usually underestimate upper tropospheric HCHO. Again this is not correct (see item 5 above).

7. On page 4 right before Section 3, the authors claim that the statistical component of their error reduces by the square root of the number of observations. Is the atmosphere really this stable to achieve such an improvement? The authors should provide some justification for this assumption.

8. Page 8, 8th /9th lines from bottom change wording to read "Figure 5 shows that ACE-FTS measurements are more representative of background values than the TDLAS in situ measurements whose flight tracks were often driven by the search for plumes and convection";.

9. Page 9, 3rd line down, either change the Perrin et al. reference year in the text (2008) to match the 2006 date in the reference list or add the proper 2008 reference in the reference list.

10. The discussion in the 3rd paragraph on page 11 regarding the reason for the maximum observed HCHO during summer in the 6-9 km range needs to be modified. The authors mention that enhanced biogenic emissions from species such as isoprene

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are likely partially responsible for this in combination with the increase in convection during summer months. Since isoprene has a relatively short lifetime it is unlikely that it can reach such altitudes on a sustained basis and the second cause is more likely. As discussed by Fried et al. [2008a,b], longer lived precursors of HCHO like methanol and methyl hydrogenperoxide can indeed have an influence on upper tropospheric HCHO levels during convection. The authors should consider rewording this section.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 1051, 2009.

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