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Atmospheric Chemistry and Physics Discussions

Interactive comment on "C_3-C_5 alkanes in the atmosphere: concentration, seasonal cycle and contribution to the atmospheric budgets of acetone and acetaldehyde" *by* A. Pozzer et al.

Anonymous Referee #2

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This manuscript reports on the inclusion of explicit chemistry for C3-C5 alkanes in a global atmospheric model. Discussion focuses on the ability of the model to reproduce observations of the relevant species and their calculated impact on the production of acetone and acetaldehyde. While the overall model comparison with observations does look good, the authors share only a small portion of the available data for comparison. I would like to see more sites represented in the comparison figures. My biggest concern is the lack of any particular finding or guidance for the modeling community. What is the significance of this result? How should one look to alter their model with respect to emissions or chemistry or both? In the end, the model still does a poor job of reproducing observations of acetone in the upper troposphere where its impact on at-

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mospheric oxidation is most important. Specific details on these concerns are outlined below.

Comparison with observations: In section 2.2, the authors note that VOC data is available for 40 stations, but only choose to highlight 8 of them. Even in the zonal distribution plots, I only see 24 sites represented. There may indeed be good reason why other sites are not shown, but no clear rationale for the selected sites is offered. Figure 2 comes closest to giving a complete picture of the latitudes and times for which data is available to compare. I am particularly intrigued by the data around 40N in this figure. While I would agree that the model does a nice job of capturing the winter maximum, there is a dramatic difference between the summers of 2005 and 2006 that is not captured by the model. Since the preponderance of emissions falls within this zone, the difference deserves comment. It is also not clear whether the VOC data is publicly available. If it is, then instructions on obtaining it should be added to the discussion.

Comparison with previous studies: At the end of section 2.3, the authors state that “…emissions are the highest source of uncertainties between the processes influencing C3-C5 chemistry.” This statement is challenged by the comparison with results from Jacob et al. Given the emissions reported in Table 1, I conclude that the differences in acetone production (section 2.9.1) are influenced more by chemistry assumptions than by emissions. This is evident for all three iso-alkanes, however, i-pentane clearly exhibits the largest difference. This is complicated by the fact that it is clearly over-predicted by the model by “factors of 3-5.” Accounting for this over-prediction, the source of acetone from i-pentane should be closer to 2 Tg/yr, which is only about 2% of the total global source and thus not terribly critical to accurately modeling global acetone distributions.

Impact on the acetone distribution: Figure 13 shows that the inclusion of C3-C5 alkanes does improve model acetone versus observations, but this does very little to improve estimates in the upper troposphere where the impact of acetone is greatest. The authors acknowledge the work of Blitz et al. on the temperature and pressure depen-

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dence of the acetone quantum yield (which many have already incorporated into their models), but even the expected increase of 60-80% would still fall well short of the observations. The Jacob et al. (2002) study was conducted prior to the work of Blitz and reports much higher acetone mixing ratios near the tropopause than shown in figure 13, suggesting that something other than C3-C5 chemistry is behind the difference.

Role of Cl and NO3: While I am not surprised that Cl and NO3 play a negligible role in the global average, I wonder if you have examined their potential for local impact. For instance, it is not unreasonable to expect that VOC observations at Alert might be significantly impacted by chlorine chemistry. Evidence for this might be diagnosed by examining seasonal changes in VOC ratios (e.g., n-butane/i-butane). Does the data show this and if so, does the model reproduce it?

Overall the paper is well written and hopefully my comments show that my concerns are primarily related to the significance of the work rather than its quality.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 1939, 2009.

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