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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Global atmospheric budget of acetaldehyde: 3-D model analysis and constraints from in-situ and satellite observations" *by* D. B. Millet et al.

Anonymous Referee #2

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This manuscript presents a study of atmospheric sources and sinks of acetaldehyde, using the results from the GEOS-Chem global model and a number of datasets from different field campaigns. The approach used by the authors is sound and the results are a clear improvement of the current understanding of the chemical and physical processes involving this important molecule. The subject is appropriate for publication in ACP and I recommend the paper is published. I only have a couple of minor comments.

General Comments

In the manuscript, it is said that NOx and O3 are kept at a costant concentration throughout the model run, but VOCs are allowed to vary. This might cause a numerical

artifact in the calculations and the authors should specify that they have checked the model output carefully and that the results are not affected. More importantly, if only two levels of NOx were used, some results are presented in a slightly misleading way. For example, at the end of section 2.2.1 the authors say that the global production of acetaldehyde is 75-93 Tg/y depending on the NOx scenario. However this is not calculated based on actual NOx emissions but using two "representative" NOx levels (0.1 and 1 ppb) to which the model is constrained. Therefore, presenting these numbers as actual production of acetaldehyde, instead of as an estimated range, is not correct. The same applies to section 2.2.2, Table 2 and to Figures 1 and 3, and the corresponding parts of the discussion. The "representativeness" of the NOx levels should be justified, as well. Part or all of the model bias in polluted areas might be due to unrealistic NOx levels in certain regions of the model: considering that part of the argument is based on the agreement between model and measured PAN/NOx ratios, the authors should address this question.

There is some discussion about the different yields of acetaldehyde in the GEOS-Chem mechanism compared to the MCM: the results of the simulations with the MCM should also be in Table 1 for easy comparison. In addition, Figure 2 clearly shows much lower yield of acetaldehyde from isoprene at low NOx, between 10 and 20 hours of the simulation, and similar yields afterwards. This is not properly discussed in section 2.2.2. Besides, if acetaldehyde formation from isoprene occurs via formation of propene, Figure 2 shows that the yield of acetaldehyde from propene also varies with time in the GEOS-Chem mechanism, but not in the MCM. Are these related and is it because propene oxidation is treated differently in GEOS-Chem and in the MCM?

The authors explain the model bias in polluted regions suggesting that it may be due to an underestimate of the emissions of the precursor VOC. They also point to previous studies indicating that alkane emissions are underestimated. However, if alkenes provide a major source of acetaldehyde in polluted air, as this and the cited studies suggest, an underestimate of alkanes would not entirely explain the model bias. Have

the authors tried to apply a correction factor based on those studies to see if that would resolve the problem? Besides, there is a consistent problem with measurements of acetaldehyde in clean air. How can the authors rule out that acetaldehyde measurements are not overestimated in polluted air, as well? And vice-versa, how can they rule out that emissions of precursor VOC are not underestimated in clean air? The PAN/NOx ratio give some indication, but the argument assumes that the modeled ratio has no bias (see comment above).

Specific Comments & Technical Corrections

page 24231, line 15: and photolysis?

page 24233, line 6: "during"?

page 24236, line 10 and 15: is it the same "gamma(t)"? if so, use capital or lower "t" for both.

page 24237, line 30: does it mean the emission rate is assumed to be the same as CH3OH?

page 24238, line 8: is this assumption made in the POET inventory?

page 24239: please clarify the difference between CDM and CDOM.

page 24241, lines 28-29: specify that the flux downwind of continents and over cool water is in opposite directions.

page 24242, lines 5-10: it should be mentioned here that that the sensitivity of results to the assumptions in the flux calculations is addressed later in the paper.

page 24243, line 23: how valid is the assumption that annual variability is small? can this information be extracted from the model?

Fig S1 seems relevant to the discussion in section 5. The authors might consider moving it from the supplment to the main paper.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 24225, 2009.