We thank the reviewer for their thoughtful and positive comments. Their comments below are in plain text, with our responses in bold italics. Citations below correspond to the reference list in the manuscript.

Response to Anonymous Referee #2.

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.

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.
This appears to be a misunderstanding. NOx and ozone were kept at a constant concentration only for the box model runs described in Section 2.2, not for the full 3D model runs discussed in the remainder of the manuscript (including Table 2 and Figure 1 that the reviewer mentions). We apologize for any lack of clarity on this point. We now specify (Section 2.2, second paragraph) that the constant NOx and ozone applies only to the box-model runs. The reviewer is correct that the box model output gives us an "estimated range" of acetaldehyde production from the various precursors. We have modified the text at various points through Section 2.2 to emphasize this point more clearly.

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.
See response to previous comment on the same issue. Also, as we point out in Section 4.2 (last paragraph), Hudman et al. (2007) showed that vertical concentration profiles for both NO_x and PAN are well-simulated by GEOS-Chem over North America. This gives us confidence in the modeled NOx levels over this region (where most of the comparisons are focused).

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.
Done.

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?
Please see our discussion of low-NOx isoprene oxidation in response to the related question in the Short Comment by D. Tarraborrelli. For propene, the reviewer is correct that the chemistry is treated differently in GEOS-Chem than MCM, because GEOS-Chem uses a lumped species PRPE to represent alkenes with greater than or equal to 3 carbon atoms (excluding isoprene); this is discussed in Section 2.2.1. However, this was not the reason for the difference in acetaldehyde production from isoprene at low-NOx (see response to Short Comment).

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?
We carried out a sensitivity run using increased alkane emissions from the US as indicated by Xiao et al. (2008) and Warneke et al. (2007), and as the reviewer implied it did not remove the model bias in polluted air. However, since this change better represents current understanding of US alkane emissions, we now use it as our baseline simulation as described in Section 2.3.2.

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?
While any artifact may also be present in polluted air, previous work has indicated that the problem is most significant in clean air (Northway et al., 2004; Apel et al., 2008). Under these conditions, acetaldehyde concentrations are low and any artificial production (e.g. on the sampling inlet) would be relatively more important. In the continental boundary layer when acetaldehyde concentrations are high, acetaldehyde measurements correlate well with other tracers and appear broadly consistent with understanding of its sources and lifetime (for example, see de Gouw et al., 2005). We therefore treat these data as more reliable. The reviewer is correct, however, that this point needs to be borne in mind. We include a discussion on this point at the end of Section 1, and have added another mention of it in Section 4.1.

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).
The high measured concentrations in clean air, particularly in the free troposphere, imply either a missing acetaldehyde source in the model (as the reviewer points out) or a measurement problem. There are three independent lines of evidence that point towards the latter. First, in the clean free troposphere, we are far from VOC emission sources, and

research to date has failed to identify an in-situ acetaldehyde source that could come close to explaining the high observed values there (for instance see Kwan et al., 2006). Second, as we show, the PAN:NOx ratio is well-simulated by the model. This implies either that the measured acetaldehyde levels are too high, or else a major flaw in current understanding of PAN-NOx chemistry that by coincidence exactly compensates for the too-low acetaldehyde in the model. Third, laboratory investigations have demonstrated measurement problems with acetaldehyde that are most pronounced at low levels and are correlated with high ozone levels (Northway et al., 2004; Apel et al., 2008). We feel that together these lines of evidence, and as laid out in the paper, make a strong case against the high observed free tropospheric acetaldehyde levels.

Specific Comments & Technical Corrections

page 24231, line 15: and photolysis?
No, photolysis is not a significant ethanol sink.

page 24233, line 6: "during"?
Changed wording.

page 24236, line 10 and 15: is it the same "gamma(t)"? if so, use capital or lower "t" for both.
Yes, it is the same. This seems to have been a typesetting error. Thank you for catching it.

page 24237, line 30: does it mean the emission rate is assumed to be the same as CH3OH?
No, the emission rate is based on acetaldehyde measurements. The corresponding emission rate for methanol in GEOS-Chem is four times higher (Millet et al., ACP, 8, 6887–6905, 2008).

page 24238, line 8: is this assumption made in the POET inventory?
No, the POET inventory provides unspeciated emission estimates for >C1 alcohols and >C1 aldehydes. We clarified this point in the text.

page 24239: please clarify the difference between CDM and CDOM.
The difference is that CDM includes detrital and dissolved materials, whereas CDOM includes only dissolved materials. We clarified this point in Section 2.4.

page 24241, lines 28-29: specify that the flux downwind of continents and over cool water is in opposite directions.
They are in the same direction (into the ocean), and we modified the text to clarify this point.

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.
Done.

page 24243, line 23: how valid is the assumption that annual variability is small? Can this information be extracted from the model?
We ran the model for 3 separate years to examine this issue. The maximum difference between

any two years for any given acetaldehyde source (primary or secondary) was <10%. We have added a statement to this effect to Section 4 (first paragraph).

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