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.

<u>Response to Anonymous Referee #1.</u>

The topic of this paper is important and within the scope of ACP. The methodology is novel and sound. The manuscript is well written. I recommend publication after minor revision to address the following specific issues:

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1. Page 24247, lines 20-25; and Figure 9 and 10: The authors stated that the current model produces a good PAN:NOx ratio over the ocean surface, and use that as the main argument against a large sea-to-air acetaldehyde flux. It would be useful to see what the acetaldehyde profiles and PAN:NOx profiles look like when the 125 Tg a-1 ocean source is used.
We carried out a sensitivity run with 125 Tg/y net ocean emission, and find that the model consistency with various observations degrades:
- Ocean mixed layer acetaldehyde concentrations increase to 2.6-42 nM (0.1-0.9 quantiles), higher than seems tenable based on the range of observations [Zhou and Mopper, 1997; Mopper and Kieber, 1991].
- Using 125 Tg/y, the model strongly overestimates atmospheric acetaldehyde concentrations in the marine boundary layer with respect to most of the airborne datasets (PEM-TB, INTEX-A, C130-INTEXB), and the shape of the CH_3CHO vertical profile over is steeper than observed.
- The shape of the PAN:NOx vertical gradient over the ocean is well-captured using our baseline simulation with a 57 Tg/y source, but is not well-captured with a 125Tg/y source (the model ratio increases with altitude less steeply than the observations). We conclude that the balance of available evidence is against a source as large as 125 Tg/y. We added a discussion of this to the manuscript (Section 4.3).

2. Table 2: Units are in Tg y-1. Everywhere else in the main text the budget is in units of Tg a-1. Modify to be consistent.
Done.

3. Figure S2 is confusing. Why is the emission factor for acetaldehyde and ethanol the same? If this is due to a lack of speciated emission factor for OVOCs in the MEGAN inventory, which resulted in the same biogenic emission estimates for acetaldehyde and ethanol, then this should be pointed out in the main text. The current text gives the impression that there is speciated information in MEGAN.
There is speciated OVOC information in MEGAN. As is described in Section 1 of the Supplemental Information (final paragraph), the few available observations of ethanol emissions from vegetation indicate fluxes similar to those of acetaldehyde, so that "As a starting point for introducing ethanol emissions in MEGANv2.1, we have used the parameterization developed for acetaldehyde to also represent ethanol emissions." So the use of the same emission factors for the two compounds reflects the available observational constraints, rather than a lack of speciation in MEGAN. We feel this point is adequately

explained in main text of the Supplemental Information, but agree that Figure S2 (now Figure S1) was confusing. We have modified the caption to make this point more clear.

Also, what exactly is plotted in Figure S2? Is it one of the emission factors ε_i, or is it the weighted $\Sigma\varepsilon_i\chi_i$, i=1,5?

It is the latter. The figure caption (now Figure S1) has been modified to clarify this point.