

We sincerely thank all three referees for their helpful, insightful and thorough reviews that have helped us to significantly improve our manuscript. Below, each reviewer comment is listed followed by our responses in blue and changes to the manuscript in *blue italics*.

### **Response to Referee #1, S. A. Nizkorodov:**

P28576, L13: parentheses can be removed around this sentence.  
We have revised as recommended.

P28577, L6: I do not see a reason to single out the visual observations of the solutions as "a preliminary study". I would just describe what happens when the solutions are sitting for a while.

We revised the text as shown in blue below:

*After ~~s~~Surface films were first detected on solutions of propanal, and its mixtures with glyoxal, and/or methylglyoxal in sulfuric acid that were allowed to react for several weeks ~~during a preliminary study~~ (see Fig. S1 in the Supplement for photos of typical surface films).; Subsequently, controlled survey studies were performed to examine the conditions required for formation of surface films”.*

P28579, L19: Can you provide examples of this variability?

Please see our response to referee #2 who requested similar information in reference to the same ACPD line number (P28579, L19).

P28585, L13: I think the fact they are ionizable may be more important than polarity

We agree and have replaced “polar nature” with “*ionizability*”.

P28589, around L20: one possible aspect of aldehydes in the LS to consider is their relatively short lifetime with respect to gas-phase photolysis – they may not have enough time to get to the particles. Perhaps this is worth mentioning.

We agree that this is worth mentioning and have added the clause in blue to the manuscript text quoted below beginning at P28589, L15:

*Neither the solubility nor the reactive uptake coefficient of propanal in sulfuric acid has been measured, but, based on the low concentration of propanal vapor in the UT/LS (~15 ppt at 11 km and presumably much lower in the stratosphere (Singh et al., 2004)) and on the short lifetimes of gas phase aldehydes with respect to photolysis, uptake and reaction of propanal alone to form polyacetals is not expected to be a significant source of organic material in UT/LS aerosols.*

Reference to Vinnik et al.: Should the "+" sign be part of the reference?

It appears unusual to us, but ,yes, the “+” sign is part of the reference abbreviation which was corrected for us by the editorial staff according to the ISI Journal Title Abbreviations Index.

Figure 2: I like the scaling to indicate the maximum contribution of different components but it makes it hard to look at the lower-intensity traces. Perhaps this figure should be split into two panels containing these data, one with absorbance scale up to 0.8, and another up to 0.1?

We have added the recommended figure with a smaller absorbance scale to the Supplement (Figure S2) along with a reference to Figure S2 in the caption for Figure 2 for readers who may be interested in details of the spectra of the standards. We prefer to place this figure in the Supplement since the smaller peaks in the lower-intensity traces of the standards cannot contribute significantly to the ATR-FTIR spectrum of the film.

Figure S1: even though you state the focus is on the ripples it is hard to understand what is going on in this photo. Is there a better quality photo available by any chance? We agree and have replaced the photo with two better ones.

Figure S2: I would recommend splitting this into 4 panels, one for each type of sample. Right now the figure is too busy to look at.

We have added the requested figure as Figure S4. The original figure is also maintained (but renumbered to Figure S3) in order to preserve the ability to compare peak positions and intensities between different organic mixtures.