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***Interactive comment on “Complex chemical composition of colored surface films formed from reactions of propanal in sulfuric acid at upper troposphere/lower stratosphere aerosol acidities” by A. L. Van Wyngarden et al.***

**Anonymous Referee #2**

Received and published: 31 December 2014

This is a well written manuscript which reports measurements on the composition of surface films formed on mixtures of propanal, glyoxal and/or methylglyoxal, at different wt% of sulfuric acid. The experimental system was chosen to investigate reactions which may occur in upper tropospheric/lower stratospheric aerosol containing significant fractions of sulfuric acid along with organic species. The authors use ATR-FTIR and proton NMR to identify some of the major species in the surface films, although the chromophore is not able to be explicitly identified. The rate of surface film formation under different conditions (temperature/illumination/pH) was also studied over long time

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periods, with the lack of film formation for solutions without propanal present noted by the authors. All aldehyde/sulfuric acid mixtures studied were observed to generate colored solutions. The study is atmospherically relevant as the production of surface films and light absorbing species will affect the physical, chemical and optical properties of aerosol particles.

I have some scientific questions on the manuscript, which are given below. I feel the manuscript would benefit from the inclusion of further details prior to publication in ACP.

P28577, L8-11: My reading of this sentence is that each organic is present in a mixture at 0.030 M ie. a mixture of propanal in sulfuric acid will contain 0.030 M of propanal, and a mixture of propanal and glyoxal in sulfuric acid will contain 0.030 M propanal and 0.030 M glyoxal. If this is the case, then the total concentration of organic in a solution will depend on whether 1, 2 or all 3 aldehyde species are in the mixture. I am not confident that the impact of different chemical species on the rate of film formation can be directly compared if the total organic loading is being altered as well as the composition of the aldehyde fraction.

P28578, L1-4: Were any steps taken by the authors to ascertain if the composition of the film generated from a mixture of 0.30 M propanal and sulfuric acid was equivalent to that generated from a mixture of 0.030 M propanal and sulfuric acid? The latter concentration is remarked in P28577, L11-14 as being more atmospherically relevant. Also, were any measurements taken to compare the composition of the films formed under the different environmental conditions studied (temperature/illumination)? The measurements performed at  $-19\text{ }^{\circ}\text{C}$  will be the most atmospherically relevant.

P28579, L19 to P28580, L2: Please elaborate further on the statement that there was variability in film formation rates between replicates of the survey experiments. How consistent were the data? It would also be helpful to have further details on the actual rate of film growth for the different aldehyde mixtures under the various conditions of pH/temperature/illumination. This could be reported in terms of whatever metric the

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authors used to quantify it, however it would give the reader an appreciation as to whether stated differences in rate were on the order of minutes, days, months etc. Such data could be included in the Supplementary Information.

Figure S1. The left hand image is extremely difficult to make out, even with the explanation given in the caption. I think this image either needs to be replaced with a clearer picture or removed.

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Interactive comment on Atmos. Chem. Phys. Discuss., 14, 28571, 2014.

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