

## ***Interactive comment on “Nitrogen-containing Secondary Organic Aerosols Formation by Acrolein Reaction with Ammonia/Ammonium” by Zhijian Li et al.***

### **Anonymous Referee #2**

Received and published: 25 October 2018

In this work, the authors performed a series of experiments to explore the reaction of acrolein, the smallest  $\alpha, \beta$ -unsaturated carbonyl, with ammonium and ammonia in both the gas and aqueous phases. Through the use of small chamber and bulk-phase experiments, they showed that acrolein will react with both ammonia in the gas phase and ammonium in the aqueous phase to produce new products, and that the aqueous phase reactions lead to brown carbon formation. This work also identifies several products of these reactions and provides a framework for understanding unsaturated carbonyl reactions in atmospheric water. This work is important for continuing our discussions of the chemistry occurring in these solutions and this manuscript will provide important information to the field after the following comments are addressed.

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On page 5, the wording of the statement “The aerosolized  $(\text{NH}_4)_2\text{SO}_4$  or  $\text{NH}_4\text{Cl}$  particles must have quickly deposited to the bag walls forming a coating on the surface” needs to be changed. The statement itself does not seem to fit into this paragraph, but if it stays in the manuscript, the words “must have” needs to be explained or revised.

Due to the fact that the RH in these systems was 90-100%, how can the acrolein/ammonia system be considered to be only a gas phase system? The authors note that a film was formed on the walls, and ammonia and acrolein could easily partition into this film. Therefore any compounds observed in this bag could have reacted in the aqueous film and then repartitioned into the gas phase.

In Section 3.1, the authors note that they did not observe pyran aldehyde in the gas phase from the acrolein/ammonia bag. If this dimer was present in the solution before injection, as the authors speculate, then wouldn't it show up in all experiments? Further, if it was formed in the wetted surface of the bag, as also speculated, why doesn't it appear in the presence of ammonium? While the ammonium reaction may be much faster, I would still expect that a small amount of pyran aldehyde might be observed in the gas phase.

Page 8, line 8: Punctuation is needed after the reference at the start of this line.

In the last paragraph of page 8, continuing into page 9, the first sentence is confusing and should be reworded. This paragraph also contains many words such as “should” and “ought” that make it difficult to determine what the authors expected to occur and what actually was observed or deduced.

Page 10, line 22: Do the authors mean “similar to Reaction 2” instead of “similar as Reaction 2?”

Page 11, line 22: There is an odd symbol in the middle of this line.

On page 12, the UPLC results for the acrolein/ammonium bag are compared to the bulk pH=6 solution, but the authors note that the actual pH was somewhere between 4.6

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and 6 in the liquid film and that there are acrolein trimers detected in the bag solutions that are not detected in the bulk solutions. They explain that this is because “the actual pH in the liquid film in the bag was lower than 6,” but was any comparison done with the bulk solutions with pH=4 or 5? They should work as another point of comparison and give an indication of whether the acidity of the solution was important or if another factor is important here.

Page 14, line 16: “that will come part of SOA” is awkward.

Page 14, line 18: Should “in the atmospheric environment where human lives” be restated as “in the atmospheric environment where humans live” or reworded in some other way?

On page 15, line 12, the authors discuss the pH value being important to the liquid products of this reaction. Are these products really liquid or are the authors referring to products in the aqueous phase?

Throughout the manuscript, there are several grammatical errors and fixing them would improve the readability of the study.

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Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2018-942>, 2018.