Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-63-RC1, 2018


Atmospheric Chemistry and Physics Discussions

# *Interactive comment on* "Evidence for pyrazine-based chromophores in cloudwater mimics containing methylglyoxal and ammonium sulfate" *by* Lelia Nahid Hawkins et al.

**Anonymous Referee #1**

Received and published: 17 April 2018

The authors present the results of an experimental study of brown carbon formation in solutions intended to mimic cloud water which contained methylglyoxal and ammonium sulfate. BrC formation was studied at a range of pH, and for some solutions which were allowed to evaporate into a hood over the course of 7 days.

The authors use chemical ionization mass spectrometry to analyze products formed, and while the mass/charge ratios of many of the identified compounds had been identified earlier in other laboratory studies of this system in the literature, by performing experiments with isotopically labeled ammonium sulfate, the authors were able to distinguish the number of nitrogens in each species. This analysis led them to propose



a new type of light-absorbing organic compound which has not been previously identified in atmospheric aerosols or their laboratory proxies, pyrazines. This element of the study is a valuable new contribution.

I have a number of serious concerns about the evaporation experiments which I am not sure can be addressed. It may be necessary to cut this material from the paper, or heavily revise with additional control experiments. It seems that the experimental method involved leaving samples uncovered in a hood for a week.

- Cloud processing takes place on a timescale of minutes to hours, so this process does not resemble the experimental conditions. How does the timescale for drying affect the results? How would one derive quantitative kinetic information from this complex combined reaction/dehydration process, and how could it be justified as being similar to what actually happens in the atmosphere?

- Leaving samples uncovered in the lab is known to lead to BrC formation in SOA samples due to contamination (e.g. the early preliminary data of Bones et al. JGR 2010). How can the authors eliminate the possibility that contamination contributed to the enhanced absorption in the dried and reconstituted samples?

- It seems unnecessary to specifically compare the effects of pH and evaporation (line 34 page 3) when mechanistically these processes are distinct and not in competition with each other in ambient cloud droplets. It's relevant to quantify both processes, but I doubt a meaningful direct comparison can be made based on the data here.

Specific comments

Abstract, line 3 - why is Maillard type in parentheses

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-63, 2018.