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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Glyoxal's impact on dry ammonium salts: fast and reversible surface aerosol browning" *by* David O. et al.

Anonymous Referee #2

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Summary and recommendation:

In this study, De Haan et al. report on fast browning of ammonium sulfate (AS), AS/glycine and methylammonium sulfate (MeAS) aerosol particles under dry conditions when exposed to gas phase glyoxal. It is shown that this browning process is not accompanied by noticeable particle growth and that this is reversed when water vapor is added. Remarkably, dry methylammonium sulfate aerosol was found to brown 4 times more than dry AS aerosol, and deliquesced AS aerosol browns much less than dry AS aerosol. Lastly, the authors estimate the impact of these browning processes on global radiative forcing, concluding that its contribution might only be important on a regional scale under polluted conditions.

The authors acquired a nice dataset with state-of-the-art instruments during their

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Discussion paper

chamber experiments, which will contribute to our understanding of brown carbon formation in aerosol particles. However, I see several points in the manuscript, which need to be addressed before I can recommend its publication in ACP (as detailed below).

Major comments:

1) Figure 1 / L111f: I cannot follow the authors' explanation of the SMPS mass loss upon chamber humidification. There is no mass increase upon GX addition under low RH levels, indicating that GX uptake to particles should be rather low. This is also consistent with the authors' assumption that most GX goes to the walls during this time (L102). So, how can a mass loss of ∼15% upon humidification then be explained by destruction of light-absorbing products formed through GX uptake? If there was only minimal uptake of GX, the particles should still consist mainly of AS. One factor that might be important here is the wall loss correction, as particle wall losses can significantly change upon RH increase. Did the authors consider this in their analysis?

2) I think the manuscript would largely benefit from some reaction mechanisms. This would help non-expert readers a lot in grasping the chemistry behind the browning reactions / imidazole formation. In the current version, all details on known and hypothesized chemical pathways are rather "hidden" in the text.

3) The discussion section is extremely speculative. Especially, since RH conditions of <5% are commonly far from tropospheric conditions. The mere statement that "tropospheric aerosol is nevertheless typically semi-solid or solid" (Line 212) does not eliminate this constraint. Therefore, I find it difficult to agree on the authors conclusion that aerosol browning by AS + GX under dry conditions may be regionally important.

Specific comments:

1) L67 and L93f: How was the wall loss correction performed? It would be helpful to have some more details here.

2a) Figure 1 / L90: I would suggest to set t = 0 h for the GX addition to improve

readability.

2b) Figure 1: unit of aerosol density is missing in the caption

3) L124 / Figure S1: The strong difference between the experiments with AS and AS/glycine seeds needs a more detailed discussion. Only referring to differences in volatility without further discussion seems too vague.

4) Table 1 and corresponding figures: It is not reasonable to assume a density of 1 g/cm3 for the seed particles. Please use some more realistic numbers.

5) Figure S2: Axis labels are missing.

6) Line 129f: Are these increases in AMS ion signals significant? Would it be possible to give some more quantitative information here? Otherwise, it is not possible to judge whether this is an important contribution to the composition of the particles. Furthermore, is the resolution of a Q-AMS sufficient to assign all these signals unambiguously to certain compounds and corresponding fragments?

7) Why are experiments 8 and 9 discussed before experiment 5–7? I would suggest to rename the experiments or to reorganize the manuscript.

8) Figures 3, S3, S4, and S6: I would suggest using a relative time on the x-axis instead of the time of the day. This would improve readability. Moreover, it would be consistent with Figure 1.

9) It would be nice to see at least for one of the experiments on wet AS an experimental overview figure (e.g., in the supplement) in the style of Fig. 1. Currently, the reader has to imagine how the experimental procedure and corresponding data looked like.

10) With a logarithmic y axis it should be possible to show both curves in one figure.

11) Table 1, experiment 1b: How did the authors infer a decrease of 3 μg/m3? From Fig. 1 the decrease seems to be in the range of 8–10 μg/m3. Moreover, this is inconsistent with the authors' statement (L111), that a mass decrease of $\sim$15% was observed.

Technical comments:

1) L95: I guess this should read “lack of uptake”.

2) Caption of Table 1: The $\pm$ sign should be formatted in black.

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