

Interactive comment on “Glyoxal’s impact on dry ammonium salts: fast and reversible surface aerosol browning” by David O. et al.

Anonymous Referee #1

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In this very well written manuscript the authors discuss results of thoroughly planned and described chamber experiments on the browning of aerosol particles upon exposure to gaseous glyoxal. Seed aerosols consisted of ammonium sulfate, methylammonium sulfate, mixed ammonium/glycine sulfate, and sodium sulfate, and were exposed to glyoxal under different relative humidity conditions. The authors observe a ‘reversible’ browning of the aerosol particles, when two conditions are met: 1) dry chamber conditions (<5% RH), and 2) amine functionalities are present in seed particles. Furthermore, the authors try to quantify the contribution of the observed browning to global radiative forcing induced by secondary brown carbon and conclude that its contribution is negligible with <1%. The findings are discussed very well and contribute significantly to the field. I have a few comments beside technical corrections that need

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to be addressed in a minor revision prior publication:

General comments:

- 1) The observation that the browning process appears to be reversible is discussed at several points throughout the manuscript. Indeed, the single-scattering albedo in Figure 1 starts to recover with declining glyoxal gas-phase concentration. However, the addition of water vapor results in a sudden recovery of the albedo back to the baseline. This effect could also be due to a dilution effect, as the absorption properties of glyoxal derived brown carbon is known to be highly concentration dependent. Could the authors comment on a potential dilution effect, especially of surface active brown carbon constituents?
- 2) Non-reversibly formed brown carbon from glyoxal might anyways not be present in appreciable amounts, given the timescale available for their formation. Maybe it would benefit the paper to expand a corresponding discussion a little bit, e.g., in line 201, where the formation of light-absorbing double bonds is mentioned.
- 3) The control experiment utilizing sodium sulfate seed aerosol is very helpful. It not only confirms the involvement of amine species in the observed fast and reversible browning effect, but also that the CAPS and PAS measurements were not biased by gas-phase glyoxal. Which other measures have been taken to ensure artifact-free analysis?

Technical comments:

- 4) Figure 3: Could the x-axis be labeled with “Elapsed Time (h)” as in Figure 1?
- 5) Line 91: Brackets could be removed.

Comments on the Supporting Information:

- 6) Figure S1: Start of chamber illumination (red line) – Did I miss it or is not there?
- 7) Figure S2: Please include axis labels; the molecular masses for m/z 119 in brack-

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ets should be 96+23; Is it common for AMS analysis that formic acid appears as a protonated species at m/z 47?

8) Figure S3: Exponents of the wall loss rate should be superscript

9) Figures S3, S4, S6: Could the x-axis label be changed to "Elapsed Time (h)"?

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