

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

**Anonymous Referee #3**

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Summary and recommendation:

This paper details experiments the production of brown-carbon formation and light-scattering ability as a function of relative humidity and minor chemical differences in aerosol type. The authors find that dry particles produce larger albedo changes upon introduction of glyoxal into their chamber than for wet particles. They also connect relative humidity changes to albedo and particle mass measurements and explain these results in the context of global radiative forcing impact, which may hold local significance even if not globally significant. In general, the results are clearly presented, the conclusions well thought out, and offer meaningful contributions to the field at large. I believe there are, however, several points that the authors need to clarify before acceptance for publication to better articulate the impact of the work.

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Major comments:

1. In Table 1 it appears that reactions 1a and 3 are nearly identical with the exception of chamber. The results, as given by aerosol concentration, mass increase, and albedo change, however, are markedly different. Is the only difference here the chamber used? It is not clear from the text how the reader should understand these differing results. The authors would do to clarify how the two chambers affect their results and offer some guidance on how their results should be read in light of those differences.
2. In lines 95-99 the authors make the case that the lack of particle growth is consistent with a lack of uptake of glyoxal at low RH, but in lines 195-196, the possible (though small) uptake of glyoxal is highlighted as a reason for water depletion. If the results suggest that glyoxal is able to access surface water (and thus uptake to the particle even under dry conditions), shouldn't this be taken into account in the particle growth discussion?
3. Following on to point #2, in lines 102-104 the discussion is hard to follow. The loss of glyoxal is largely to the steel walls, and yet the large albedo decline is also due to glyoxal-driven surface reactions? Does this suggest that even minor amounts of reactivity lead to very high albedo changes? I think the confusion readers will have with this section that the authors need to clear up is related to the sizing language. How should we read “largely”, “large”, and “at least some” in order to understand the weight of the argument the authors are making here?
4. In regards to Figure 1, how much of the recovery of albedo upon introduction of water is due to exclusively to the introduction of the water and how much is due to there still being glyoxal in the chamber as evidenced by the PTR-MS signals at 59 and 31? Unlike the earlier addition of a small glyoxal concentration, this larger addition was not allowed to return to baseline, and while this may not matter in the resultant albedo recovery upon water addition discussion, this overlap isn't addressed adequately in the text. Essentially, is the albedo increase due to the addition of the water, or the resultant

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dilution/loss of glyoxal signal?

5. In lines 143-144, and Figure 2 the case is made for albedo change as a function of glyoxal concentration being a second order polynomial with respect to glyoxal concentration. Is there a physical explanation that would help defend this choice, or is it simply what fit the data? Or, is there a reason that using a two-slope approach (say a linear fit to the data at  $[\text{glyoxal}] < 0.35 \text{ ppm}$  and another linear fit to the data at  $[\text{glyoxal}] > 0.35 \text{ ppm}$ ) wouldn't also successfully capture the data? The authors should offer with what significance the readers should approach this polynomial fit and  $[\text{gly}]^2$  dependence.

Minor and technical comments:

1. At least when I downloaded a copy of the paper, Table 1 had some jumbled values in the last column (it would appear that those values are line numbers). The authors should check this Table to ensure that everything is in the place they expect it to be. This could very well be an artifact of the download and not the paper, and so this might disappear in final publication.

2. In regards to the discussion on lines 195-196 again, a more curiosity-driven question the authors may consider, if they wish, commenting on is: can this ability of glyoxal to access surface water lead to a localized area of highly concentrated glyoxal (and that would thus accelerate chemical reactivity)?

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