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Interactive comment on “The optical, physical and chemical properties of the products of glyoxal uptake on ammonium sulfate seed aerosols” by M. Trainic et al.

Anonymous Referee #3

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The authors present results on glyoxal uptake by ammonium sulfate seed particles at varying RH. They find that glyoxal is taken up when $RH > 35\%$, leading to particle growth. These results are interesting and important in the context of the various previous measurements that have been made regarding glyoxal uptake. The emphasis on the change in ‘optical activity’ of the particles is however, in my opinion, misplaced. The greater increase in cross-section for small particles is exactly what is to be expected, given that the change in extinction efficiency is very steep for diameters less than 200 nm or so. The estimate of the refractive index for the glyoxal reaction products is potentially interesting, however the analysis procedure used to obtain this estimate has some significant short-comings. The authors should consider their data comprehen-

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sively, rather than picking only a subset of the data for analysis.

General Comments: Does drying the particles after reaction have any effect on the measurements? This has implications for the discussion provided on p. 19238, line 10.

Regarding the optical property measurements, were the cross-sections corrected for contributions of doubly-charged particles, the contribution and importance of which likely vary as a function of particle size?

The authors should provide a definition of what they specifically mean by “optically active.” What is the criterion used?

The authors should determine mean RI values for the combination of AS and their glyoxal reaction products for the deliquesced particles (i.e. for the reactions above 85% RH). If the particles are internal mixtures, as opposed to core-shell, the RI should be a combination of the pure AS and coating RI's, weighted appropriately (by volume?). This seems an important self-consistency check with the core-coat results. Unfortunately, I think the authors will find that the use of a weighted average RI, where the glyoxal reaction product RI is based on the lower RH core-coat particles, will not reproduce the observations, especially if all of the different size particles are included.

The authors use $1.552 + 0.002i$ for the RI of the ammonium sulfate seed particles. However, their dry particle measurements appear to be more consistent with a real part of the RI closer to 1.5; use of 1.55 with Mie theory over-predicts the measured Q_{ext} values for the dry particles for the 200 nm and 300 nm particles, in particular. How much does the choice of the core RI affect the deduced coating RI?

As the authors note, the uncertainties associated with fitting a single data point can be very large. The authors should instead rely on the simultaneous fit to all data points to determine their best fit RI value for the coating. Further, I do not understand why the fit was limited to the 300 nm AS particle data-set only. Why not use the 250 nm

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data set, where there are many more data points? Or better yet, all of the available data. It would be much better if the authors were to do a global fit to all of their data at all sizes (excluding, perhaps, the deliquesced particles). I am guessing the authors' argument against this might be that the RIs of the coatings may actually be different for the different RH's because the composition is different. However, I think that the use of single data points is insufficiently constrained (see next paragraph), and thus the merging of the data is the less problematic choice.

Related to the previous point, the ranges reported by the authors for the RI values of the coating (p. 19235) seem to be insufficiently broad and possibly incorrect. For example, I find that, without even considering the uncertainties, an RI of $1.81 + 0.02i$ gives perfect agreement with the 378 nm data point, as does $RI = 1.75 + 0.04i$ or $RI = 2.1 + 0.09i$. These are somewhat arbitrary choices: again, using a single data point does not really constrain the possible range of values, which the authors acknowledge, but do not seem to fully appreciate. If uncertainties are taken into account, an even larger range of values is possible.

The uncertainties in the Q_{ext} values given in Table 2 are inconsistent with the uncertainties in Table 1. If the relative error in the diameter is around $\pm 10\%$ (approximately what is shown in the table), then the uncertainties in the Q_{ext} values must be at least as large, given the nature of error propagation upon division. This does not seem to be the case. Presumably the 'uncertainties' in the diameter are not really uncertainties, but more of a specification of the width of the size-selected mode?

The authors were using a high resolution AMS. Presumably, this instrument is capable of determining the atomic composition of the aerosol. Why do the authors instead rely on the unit mass resolution analysis procedure of Galloway et al. (2009)? Why not try and utilize the full capabilities of the instrument? What sort of bias is possible by using the UMR data?

Figure 3 is un-necessary, in my opinion. It would be better to replace it with a figure

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that shows the measured $Q_{\text{extinction}}$ values as a function of particle size along with the best fit curves.

I do not believe that the authors can make any claims as to an increase in the imaginary RI with RH, as they do on p. 19237, line 14. This goes well beyond the uncertainties of the measurements, especially given that the authors have done their fitting for single data points (which inherently have large uncertainties). The authors state specifically that “Although the increase is within the model error, we cannot rule out that it indicates a trend of growth in the imaginary part of the RI with increasing RH.” This is misleading: just as they cannot rule it out, neither can they rule it in. Their data provide absolutely no support in either direction, given the uncertainties. This discussion should be removed from the manuscript. If they wish to even attempt to make such a conclusion, then they must, at the very least, include the data from all of the measurements (i.e. all AS particle sizes below deliquescence).

Page 19237, Line 20: The authors compare their results to the literature (specifically Liggio et al. 2005), but seem to restrict their comparison to the 300 nm AS particle results. It is not clear to me why they exclude their 100, 200 and 250 nm particles from the discussion, especially given that Liggio et al. used 125 nm particles. I also do not see where the authors have actually shown that they “found in this study that the reaction is enhanced with decreasing initial particle size.” They did show that the optical growth is larger for the smaller particles, but this says nothing about whether the reaction is enhanced or not. The authors also do not justify (or even discuss) their “linear reaction rate” assumption to extend their results to 4 hrs (Table 3). Overall, I find the discussion in this paragraph to be weak.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 19223, 2011.

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