

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 #1

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The authors describe quantitative measurements of the changes in physical and optical properties of ammonium sulfate (AS) seed aerosol undergoing heterogeneous chemistry with glyoxal over a range in relative humidity (RH). These are clearly very challenging measurements and the work deserves publication in ACP. However, I do feel there are a number of points of varying significance that require greater clarification and thought.

Abstract, page :19224: I do not like the sentence ‘The observed increase in the ratio of the investigated absorbing substances is slightly indicated in the RI values found by the model, as the imaginary part of the product RI increases from 0.01 to 0.02 with

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increasing RH.’, particularly the use of the word ‘slightly’. An increase from 0.01 to 0.02 is a factor of 2, which is not ‘slight’. However, I suspect that this is close to the lower limit that can be measured by this technique (it may then be considered to be slight) and the reader is left questioning what the level of reported error should be. The authors should include an error with this value and this should also be discussed in greater depth later in the manuscript.

Page 19232: The phrase ‘leads to more optically active aerosols’ could be misleading, based purely on the extinction cross-section. I would prefer the change in size to be normalized out to give the extinction efficiency change, as the authors do later in the paper. Although the geometric cross-section increases by only a factor of 1.35, the extinction cross-section increases by 2.64. Generally, I don’t like the use of the phrase ‘optically active aerosols’. All aerosols are ‘active’ at some level, by scattering or absorbing light. It is just that their extinction cross-section may be too low to be significant.

Page 19233: The reasons why reaction is seen at an RH below the deliquescence point of AS seem to be quite tenuous. Given that the particles are dried and the deliquescence RH is not reached until 80 % where the particles will become solution droplets, they attribute the growth in size entirely to a monolayer of water, which is thought to occur above 20 % RH. If this reason is correct, why is no growth seen above 20 % RH, but only at 35 % and above? The authors talk about the monolayer as being like a bulk solution phase saying: ‘The aqueous conditions allow for the dissolution of the AS and the glyoxal gas into the outer layer of the particle, leading to a highly concentrated aqueous component around the particle.’ This seems inconsistent with the description of a monolayer of adsorbed water and more consistent with a particle that is undergoing dissolution/deliquescence. Following this sentence the authors state: ‘This may also explain why the change in the optical and physical cross sections of the product aerosol is enhanced with decreasing RH values in the reaction.’ It is not clear why this sentence follows logically from the preceding one and more explanation is required.

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To what extent must gas phase reaction be considered with the deposition of water soluble secondary organics on the particle surface which then behave then as a true solution phase? In general, I find the consideration of the phase of the aerosol and the phase in which the different components are residing to be rather ill-defined. This may be with good purpose as it may not be known, but there does appear to be confusion in the way the manuscript is written.

Page 19233: For the smallest size (100 nm) can the authors give some estimate of the uncertainty in the extinction cross-section for the pre-reacted particles? How sensitive is the CRD technique to such smaller particles and what concentrations are required to measure such a small cross-section? I presume that the dramatic increase in the extinction cross-section for these very small particles when compared with the larger particles is just due to the region of the extinction efficiency curve that they fall on. I do not feel that this is ever adequately explained.

Page 19234, core-shell model: If some of the ammonium has been used up by reaction, given the considerable increase in the particle size due to reaction, is there not a potential significant error in assuming that the core size has not changed or even changed in refractive index? The real parts of the refractive indices retrieved for the two shells are considerably different (1.67 and 1.71) and much larger than the value of 1.61 for HULIS compounds reported previously – I do not understand the phrase 'qualitatively consistent', do they just mean the refractive index has increased on aging?

Page 19238: The final sentence states: 'This suggests that under the same RH conditions (in the 40–80% range), the reaction of an evaporating AS droplet will be slower than for the reaction under hydration conditions, due to the higher water content of the dehydrating particles compared to the hydrating ones.' This clearly depends on other factors such as particle size. Are the authors comparing rates for particles of the same dry AS size, the same total size etc.? I feel this is a poor sentence on which to finish section 3 without suitable clarification.

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For size parameter, it is common to use x rather than greek χ . This should be changed.

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