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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Identification and characterization of aging products in the glyoxal/ammonium sulfate system – implications for light-absorbing material in atmospheric aerosols" *by* C. J. Kampf et al.

Anonymous Referee #3

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This manuscript presents results of the aqueous phase reaction between ammonium sulfate (AS) and glyoxal, a topic of great interest in the field of SOA formation and their possible climatic importance. Specific interest lies in the formation of absorbing compounds due to condensed phase reactions. The authors present a new biimidazole product as well as new low polarity products. While these products form in minute yields they may significantly contribute to the observed absorption peaks.

The manuscript is well written, and the findings contribute to a better understanding of reactions leading to SOA formation. Therefore, the manuscript is appropriate for ACP

after addressing the comments below.

My overall comment is that the discussion about the expected absorption in atmospheric aerosols due to the reaction is inconsistent and at times inaccurate (see below).

The authors state in the introduction p. 6237 line 22 that 'Imidazoles were identified as major products from the reaction of dicarbonyl compounds and ammonium ions..' but this is not accurate for all cases. Galloway et al., (2009), and Trainic et al., (2012) show that the contribution of C-N compounds to the total organic mass is only approximately 0.5% and 0.1%, respectively.

In section 3.5 p. 6246 line 19, the authors state about similar conditions to those mentioned in the introduction that 'However, the formation of imidazoles under these conditions was reported to be of minor importance..'

This is indeed in agreement with the cited papers, and therefore the authors should make this point clearer both in the introduction and in the conclusions sections, and in general throughout the paper.

Inferring about the optical properties of atmospheric aerosols from reactions in solution is problematic. The conditions present in heterogeneous reactions between aerosols and gas phase glyoxal are very different than in the bulk solution. As the authors state, the bulk is a better representative of reactions occurring in cloud droplets. Additionally, there are questions regarding the humidity dependence of the reactions as well as questions about the stability of the products measured upon drying.

Therefore, in p. 6246 line 3: 'In ambient aerosols the number of potential reaction partners is much higher and an identification of appropriate tracer compounds is necessary to estimate the organic carbon production or the impact on aerosol optical properties from distinct precursors.'

The authors should either remove the part in the sentence which refers to aerosols or

emphasize that they are in high ambient RH conditions, such as in clouds or the vicinity of clouds.

p. 6238 line12: 'The purpose of this paper is to further complete the understanding of the product spectrum of the reaction of glyoxal and ammonia in aqueous aerosol mimics' should be 'the reaction of glyoxal and ammonium sulfate' since some products are specific to the reaction of glyoxal with ammonium in the presence of sulfate and are not observed in other ammonium solutions (Noziere et al., 2009).

p. 6242 line 10: '(see Supplement for more details)' – state specifically where in the supplements (page, paragraph, figure, etc).

p. 6244 line 6: 'Our values for IC tend to be a bit lower' – could the authors give a quantitative evaluation, perhaps state the percent difference between the values for IC, and their cause?

p. 6245 lines 1-16: this paragraph discusses unidentified structures of low polarity products. I suggest the authors either do some further analysis which will enable characterization of the products. If no such analysis is available, the authors should consider whether this paragraph is necessary. It is my suggestion to delete this paragraph and state only that a functional group containing C-N bonds was identified.

p. 6247 lines 5-7: 'As a consequence, certain organic multiphase processes, e.g. the uptake and further reactions of organics and ammonia leading to secondary light-absorbing products (formation of brown carbon), can be expected to be strongly enhanced.' – Can the authors give a quantitative estimate of the enhancement – by how much? Have the authors conducted experiments under such conditions or they may be able to extrapolate from their findings?

p. 6248 lines 23-27: 'Especially in regions with conditions potentially favoring the formation of imidazoles, i.e. a higher pH value and high ammonium ion concentrations in aerosols (e.g. reported by Kulshrestha et al. (1998) and Parashar et al. (2001) for

Northern India), this influence could be very strong.'

Same comment as the one above.

Technical comments:

p. 6239 line 16: '2-(1H-imdazol-2-yl-)' should be '2-(1H-imdazol-2-yl-)'

'-1H-imidazole' should be '1H-imidazole'

'(2,2'-bisimidazole, 2,2'-biimidazole, BI)' should be '2,2'-bisimidazole, 2,2'-biimidazole, (BI)'

p. 6243 line 18: '(11$\pm$1) %' should be '11% ($\pm$1)'

p. 6249 line 1: 'as reported e.g. by See et al., 2006)' should be 'as reported by See et al., (2006)'

Figures 4, 5, and S11: Authors should use different markers and line-styles in order to dedifferentiate between the different parameters shown in the figures.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 6235, 2012.