

Interactive comment on “Glyoxal uptake on ammonium sulphate seed aerosol: reaction products and reversibility of uptake under dark and irradiated conditions” by M. M. Galloway et al.

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We would like to thank Dr. Nozière for taking the time to read this paper and offer her comments and suggestions. We have answered the questions asked below, with Dr. Nozière's comments listed below in italics, followed by the authors' responses.

In addition to the formal reviews, we would like to make a few comments on the Galloway et al. paper. This is an interesting work, directly relating to our recently published study of the reaction of glyoxal in ammonium solutions (Nozière, Dziedzic, Cordova, J. Phys. Chem. A, 113, 231, 2009). Both studies complement each other well as the approaches are very different (SOA growth in this work vs bulk liquid-phase kinetics in our case). Our results reinforce those in this manuscript by demonstrating that glyoxal

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reacts in ammonium solutions independently from any acid catalysis pathway. We thus strongly encourage the authors to quote our work, as we will quote this work in our future papers.

We will certainly quote the work mentioned above in the final version of the paper.

Although our work first reported the reaction of glyoxal in ammonium solutions, this manuscript reports the determination of imidazole derivatives. For information, some works (Prasanthi, V., et al., Asian Journal of Chemistry, 19, 5778, 2007 and reference therein) have previously studied the formation of imidazole derivatives from glyoxal in ammonium sulfate, and report a yield of 4 to 9 percent for glyoxal concentrations lower than those of ammonium.

Thank you for referring us to the Prasanthi et al. paper, which studies imidazole formation in ammonia solutions, rather than ammonium sulphate solutions. The formation of imidazoles from glyoxal and ammonia is a well established synthetic organic reaction, and was first published in 1858 by Heinrich Debus (Debus, 1858). The reaction of glyoxal in aqueous ammonium solutions, including ammonium sulphate, has also been previously reported; for example, two patents, in 1973 (Schulze, 1973) and 2003 (Cho et al., 2003), have been granted for this. Schulze reported being startled at the high yields (69% under their conditions) of products when ammonium salts, including ammonium sulphate, were present. This could be a result of the ammonium catalysis proposed by Nozière et al. but it is also known that high anion concentrations can influence the behavior of glyoxal in aqueous salt solutions (Ip et al., 2009). To our knowledge, our work is the first to report the formation of carbon-nitrogen containing compounds (imidazoles) through reaction of a carbonyl with ammonium sulphate seed aerosol.

We also have two questions:

The abstract (li 8) states that the “analysis provides evidence for the irreversible forma-
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tion of C-N products". What is the experimental evidence supporting the fact that this formation is irreversible?

During dilution of the chamber, there is no loss of m/z 68 from the aerosol phase, indicating that this is an irreversible reaction, as loss of the starting materials (glyoxal loss from re-volatilization to gas phase) would shift equilibrium in a reversible reaction.

Similarly, the formation of organosulfates in some of the experiments is presented as a main result of this work. Yet, there does not seem to be any direct evidence for organosulfates in the analysis. If anything, the high-resolution MS analysis demonstrates that organosulfates were not present and that masses previously attributed to such compounds were, in fact, non sulfate-containing compounds. What are the evidences supporting this major claim (or can they be made clearer)?

Filter samples taken during several studies and analyzed with UPLC-HR-TOF-MS show organosulphate formation during irradiated studies, and a lack of organosulphate during non-irradiated studies. See Figure 4 for support.

References:

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