

Interactive comment on “Secondary organic material formed by methylglyoxal in aqueous aerosol mimics – Part 2: Product identification using Aerosol-CIMS” by N. Sareen et al.

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Received and published: 10 November 2009

We thank Referee 1 for his or her helpful comments and address the Referee’s concerns below.

1. Purity of the methylglyoxal solution: The manuscript notes that the solutions had a pH of 2 (p. 15570) and the companion paper states that the methylglyoxal solution itself also had a pH of 2. This indicates that other compounds, e.g. pyruvic acid-the oxidation product of methylglyoxal, were also present.

The reviewer is correct, aqueous methylglyoxal solutions typically contain a small amount of pyruvic acid. Pyruvic acid is a relatively strong organic acid, with pKa =

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2.49. Therefore, the fact that our stock solution is pH =2 corresponds to a very small (0.07%) impurity of pyruvic acid in the methylglyoxal stock solution. We have included a passage to this effect in the revised manuscript.

2. The data should be presented in the context of control experiments and a progression of additives. It would be useful to include each of the reagents individually as controls, and include experiments that are performed using permutations of ions (i.e. ammonium chloride and sodium sulphate), with attention to ionic strength of these solutions, to demonstrate the effect of each ion.

Note that, upon the suggestion of the reviewers and the Editor, we have combined the two manuscripts into one. Experiments demonstrating the effect of individual salts and salt concentration were performed using UV-Vis spectrophotometry (this information was previously part of the companion manuscript). We narrowed the focus of the Aerosol-CIMS studies based on the UV-Vis experiments.

As per the reviewer's suggestion, we have performed Aerosol-CIMS control runs with methylglyoxal in the absence of salt and these results will be included in the revised version of the paper.

3. With the exception of nitrogen containing compounds there is insufficient evidence for an effect of AS or NaCl on methylglyoxal: m/z 217, 271, 273, 275 and 289 in figure 3 comprise most of the m/z discussed in the manuscript. These m/z appear to be related to m/z 109, 163, 165, 167 and 181 in figure 5. The patterns are very similar, except for slight intensity differences. The assignment, expressed as C₃H₆O₃ (I-/H₃O⁺), C₆H₈O₄ (I-/H₃O⁺), C₆H₁₀O₄ (I-/H₃O⁺), C₆H₁₂O₄ (I-/H₃O⁺), C₆H₁₀O₅ (I-/H₃O⁺), which is one of the possibilities given in the paper, can explain this pattern.

Compared to the differences observed in figure 3, the intensity differences observed between the AS and NaCl solutions for the above m/z in figure 5 are very small. Thus from figure 5 one cannot conclude that there is a specific effect of AS versus NaCl, except for the nitrogen containing compounds (m/z 125.9 and 143.8 in fig. 5). This

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raises the concern that the intensity differences observed between the two solutions (and thus an inferred difference in reactivity between AS and NaCl in figure 3) are caused by a difference in ionization efficiency (e.g., the compounds in question do not get ionized efficiently by I⁻ in presence of Cl⁻). The above m/z exist in both salt solutions and without a methylglyoxal control experiment it is difficult to conclude that the salts have influenced their formation.

The results of the control runs with methylglyoxal in the absence of salt demonstrate more clearly the influence of salts on product formation. The results suggest that NaCl may catalyze the formation of non-light-absorbing hemiacetal species.

The reviewer's concern that the compounds might not get ionized efficiently by I⁻ in the presence of Cl⁻ is not justified since volatilization happens at relatively low temperature upstream of the chemical ionization region. That is to say, Cl⁻ ions will not be present in the gas phase when the organic compounds are ionized. The use of Aerosol-CIMS to study organics in mixed inorganic/organic aerosols has been previously established by McNeill et al. (2007, 2008).

4. The companion paper clearly demonstrates an effect of AS on the optical properties. However, the carriers of this signal could be present at low concentration and/or unrelated to many of the m/z discussed in this manuscript.

The assignment of the UV absorption bands using information from our ab initio calculations is discussed in the revised (combined) manuscript (as well as in the Part I discussion paper).

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9, C6980–C6983, 2009

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