

Interactive comment on “Characterization of non-photochemically formed oligomers from methylglyoxal: a pathway to produce secondary organic aerosol through cloud processing during night-time” by F. Yasmeen et al.

F. Yasmeen et al.

magda.claeys@ua.ac.be

Received and published: 21 February 2010

We thank Dr. McNeill for constructive comments, which were highly appreciated and helped to improve the manuscript. The comments have been/will be addressed as outlined below.

My primary concern is that the non-photochemical oligomerization of methylglyoxal in aqueous systems is already well-established as a potential source of SOA and this

C11013

work should be placed in that context. It is well-known that methylglyoxal becomes hydrated and forms acetal and hemiacetal oligomers in aqueous solution (Nemet et al., 2004; Loeffler et al., 2006; Zhao et al., 2006; Krizner et al., 2009; Paulsen et al., 2005).

Response: We fully agree to compare our work with that of already published studies on the non-photochemical oligomerization of methylglyoxal. With regard to the suggested articles, Loeffler et al. (2006) has already been cited in our manuscript; the other articles will be properly cited, except Paulsen et al. (2005), which we found out of context.

The manuscript in its current form does not make it clear that this is not the first demonstration of methylglyoxal oligomerization in aqueous solutions containing ammonium sulfate; my group published two manuscripts on this subject in ACPD in July 2009 (Schwier et al., 2009; Sareen et al., 2009). As pointed out by Anonymous Referee #1, we used aerosol chemical ionization mass spectrometry (Aerosol-CIMS) to obtain mass spectral evidence of aldol condensation and hemiacetal formation by methylglyoxal in atmospherically relevant aqueous systems (Sareen et al., 2009). DeHaan et al. (2009) presented mass spectra of similar oligomers formed by methylglyoxal in evaporating droplets. Placing this work in the context of the publications I mention here would strengthen the authors' presentation of their data.

Response: We believe that our study provides the first demonstration of acid-catalyzed self-oligomerization of methylglyoxal via aldol condensation reactions under dark conditions; more explicitly, we have studied the nighttime chemistry of methylglyoxal in a simulated cloud medium. Up till now, only computational studies by Krizner et al. (2009) and Barsanti et al. (2005) have suggested that self-oligomerization of methylglyoxal is a thermodynamically favorable process but no experimental evidence under dark conditions was available. We have further demonstrated that oligomerization is not only catalyzed by $\text{NH}_4(\text{SO}_4)_2$ but rather depends on the ionic strength and acidity of the medium. We fully agree to compare our work with that of recent studies on the

C11014

oligomerization of methylglyoxal. We only became aware of the recent articles of your group after our manuscript was submitted; the two articles, now merged to one (Sareen et al., 2010), as well as the article by De Haan et al. (2009) will be properly cited in the revised manuscript.

Additional references (to be included in the revised manuscript)

Barsanti, K. C. and Pankow, J. F.: Thermodynamics of the formation of atmospheric organic particulate matter by accretion reactions. part II: Dialdehydes, methylglyoxal, and diketones. *Atmos. Environ.*, 39, 6597–6607, 2005.

De Haan, D. O., Corrigan, A. L., Tolbert, M. A., Jimenez, J. L., Wood, S. E., and Turley, J. J.: Secondary organic aerosol formation by self-reactions of methylglyoxal and glyoxal in evaporating droplets, *Environ. Sci. Technol.*, 43, 8184-8190, 2009.

Krizner, H. E., De Haan, D. O., and Kua, J.: Thermodynamics and kinetics of methylglyoxal dimer formation: A computational study. *J. Phys. Chem.A*, 113, 6994-7001, 2009.

Nemet, I., Vikić-Topić, D., and Varga-Defterdarović, L.: Spectroscopic studies of methylglyoxal in water and dimethylsulfoxide. *Bioorganic Chemistry*, 32, 560-570, 2004.

Sareen, N., Schwier, A. N., Shapiro, E. L., Mitroo, D., and McNeill, V. F.: Secondary organic material formed by methylglyoxal in aqueous aerosol mimics. *Atmos. Chem. Phys.*, 10, 997-1016, 2010.

Zhao, J., Levitt, N. P., Zhang, R. Y., and Chen, J. M.: Heterogeneous reactions of methylglyoxal in acidic media: Implications for secondary organic aerosol formation, *Environ. Sci. Technol.*, 40, 7682-7687, 2006.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 9, 22993, 2009.

C11015