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Atmospheric
Chemistry
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Discussions

Interactive comment on “Humidity-dependent phase state of SOA particles from biogenic and anthropogenic precursors” *by* E. Saukko et al.

Anonymous Referee #2

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The manuscript “Humidity-dependent phase state of SOA particles from biogenic and anthropogenic precursors” reports measurements of laboratory-generated organic aerosol particles with a focus on the phase (solid, semi-solid, liquid) of these particles. Overall the manuscript is well written and scientifically sound. The length is appropriate for the results and the topic merits publication in Atmospheric Chemistry and Physics.

However, prior to final publication the authors should clarify a few things. Overall, the methods used to generate the particles raises the question if and to what degree these results can be applied to the real atmosphere or how the implications from these experiments would hold true for “real-world” aerosols. The generation of oxidized compounds using the PAM technique leads to a high degree of oxidation of organic compounds but does barely resemble the life cycle of an aerosol particle in the real atmosphere. Aging

of particles or air masses in the atmosphere involves several processes like the dial cycle, temperature changes, air mass transport, cloud processing and oxidation. Unlike in the PAM, *OH is not the only oxidant. Furthermore, different oxidants lead to different products and path ways of atmospheric oxidation. Also, in the atmosphere organic aerosols are produced from thousands of precursors and not from only a limited number.

Another small point of criticism is the representation of the mass spectrometry results. Although these are not the main focus of the manuscript a large part of the findings is based on the O/C ratios obtained by the AMS; however a clear description on how these results were obtained as well as quality assurance for the instrument are simply omitted. Where there any changes applied to the fragmentation table of the AMS analysis? While under ambient conditions the fragmentation table requires only small adjustments, for laboratory measurements the situation is quite different, especially if organic compounds are involved. Often it can lead to artifacts and misinterpretation of results. The same applies to the sulfate measurements. Sulfate will dissociate and form sulfuric acid during the thermal desorption process; therefore, water is one of the most important fragments. Any source of water, but especially changes in relative humidity, particulate water or water from organics might lead to a bias. Were the results corrected for such artifacts? Also, what are the uncertainties of the AMS measurements and O/C ratios? In general I believe the mentioning of errors or error bars (not only for AMS data) would be largely appreciated by many readers.

Specific comments:

P4449 L24-26 “Organic matter (OM) forms up to 90% of observed aerosol particulate mass, and secondary organic aerosol (SOA) represents up to half of the organic fraction (Jimenez et al., 2009; Hallquist et al., 2009) on the global scale.”

In this reviewers opinion this statement is wrong or at least misleading. While there might be circumstances where organic matter can contribute up to 90% of the particulate aerosol mass, these occasions are rather limited to very few places. The composition of the atmospheric aerosol is by far dominated by inorganic compounds, although organics sometimes dominate the non-refractory sub-micron size range.

P4450 L1-3 “SOA particles are generally associated with a direct cooling effect as they scatter the incoming solar light and they participate in cloud formation by acting as cloud condensation nuclei, CCN (IPCC, 2007, ch.2).“

I would change this statement. Secondary organic aerosol particles are definitely not the most important sources for CCN. In the current understanding, organic aerosols are believed to influence cloud formation by rather suppressing the CCN activity of a particle.

P4452 L2-5 “The reactor is capable of simulating atmospheric oxidation timescales of days to weeks with actual residence times of minutes.”

Why is it important or atmospherically relevant to simulate oxidation timescales of weeks? The average lifetime of an aerosol is believed to be around eight days. Is there any measurement/source that suggest an aerosol particle could actually undergo oxidation for such a long timescale without being removed?

P4453 L12-13 “Gas-phase SOA precursors used in these experiments were a-pinene, longifolene, isoprene, naphthalene and n-heptadecane.“

I suggest removing this sentence. The information is redundant within this paragraph.

P4457 L22-23 “AMS particle time-of-flight measurements confirmed that the SOA and sulphuric acid were internally mixed in all cases.”

Please clarify. Did you investigate single particle spectra, or simply assume internal mixture from the particle-time-of-flight distributions? What if the particles had a sulfate core and an organic coating? Wouldn’t the PToF distributions still suggest internal mixture?

P4457 L 27-28 “The ammonia is presumably from trace concentrations from the PAM system and makes up 11–14% of the total mass.” The ammonia can originate from many origins including the tubing, or it can be an artifact of the AMS analysis and the fragmentation table. Without the mentioning of detection limits or quality assurance from the AMS analysis this number could well be within the noise of the instrument. What was the total mass measured by the AMS? This information could give the reader more confidence in the AMS results.

Table 1

O/C and H/C values are presented with two digits precision; however no uncertainty or errors are presented. Are the differences significant (for example, for isoprene)? Please add this information.

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