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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Viscous organic aerosol particles in the upper troposphere: diffusivity-controlled water uptake and ice nucleation?" *by* D. M. Lienhard et al.

D. M. Lienhard et al.

ulrich.krieger@env.ethz.ch

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The authors would like to thank Referee #1 for the very positive recommendation, the comments and queries. We address the reviewer's comments in our response given below. We will incorporate corresponding changes and clarifications in a revised version of the manuscript.

Referee #1: In the abstract the authors should indicate the type of secondary organic aerosol (i.e. photooxidation of alpha-pinene) they investigated.

Authors' response: We will do this in the revised version. The first sentence of the abstract will be changed to "New measurements of water diffusion in aerosol particles produced from secondary organic aerosol (SOA) material produced by oxidation of alpha-pinene and ... "

Referee #1: Abstract, lines 20-25; Page 24485, lines 3-6; page 24486, lines 3-5: The authors state "that condensed-phase water diffusivity is unlikely to have significant consequences on the direct climatic effects of SOA particles under tropospheric conditions." I assume here the authors are referring to the "direct effect of aerosols on climate" whereby the aerosols can affect climate by scattering and absorbing radiation. The authors may want to define the "direct climatic effect" somewhere in the manuscript for clarity.

Authors' response: Yes, the reviewer is correct in that we are referring to scattering and absorption. We will leave the expression as is in the abstract, but will change the sentence on page 24485 line 5 to "The direct climatic effects of SOA particles, i.e. their scattering and absorptive properties, are thus unlikely to be significantly affected by condensed-phase water diffusivity."

Referee #1: Methods. It would be useful to add a few additional details on the conditions used when generating SOA in the PAM. For example what was the mass loading and collection time.

Authors' response: We will add the following sentence to the revised version (24478 line 28): "The α-pinene SOA samples investigated in this study were generated with a Potential Aerosol Mass (PAM) flow tube reactor as described in detail by Lambe et al. (2011) from the gas phase oxidation of α-pinene with OH radicals and collected **onto**

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15, C9510–C9513, 2015

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47 mm teflon filters. To collect sufficient sample mass (4-5 mg) for the offline analysis, steady-state SOA mass concentrations of approximately 300 - 400 μg m^{-3} were produced in the reactor and collected at 8.5 L/min for 24 hrs."

Referee #1: The SOA results shown here are made with one type of SOA (photooxidation of alpha-pinene) generated with one oxidation level (O:C = 0.53). The authors then use results from this SOA and other proxies of SOA to make general conclusions on the atmospheric importance of condensed-phase water diffusivity. How confident are the authors that these results will extrapolate well to other types of SOA? Somewhere in the manuscript the authors should add the caveat that the results and implications were based on one type of SOA, and further studies are needed to understand the importance of water diffusion in other SOA.

Authors' response: We agree with the reviewer that more work is clearly needed to understand in detail the influence of O:C on water diffusivity as well as the influence of the process and precursors generating the SOA. However, our model compounds clearly show that there is no trivial relationship between any physicochemical property and water diffusivity. Therefore, we argue (cp. Fig. 3a) that there is range of diffusivity at a particular temperature which seems to cover at least a few proxies for SOA as well as our sample from photooxidation of alpha pinene. Of course this does not prove that in another SOA, water diffusivity may fall out of this range. We will add the following sentences to the end of our discussion on page 24481 line 5 as: "Based on these results we take the water diffusivity of the alpha-pinene SOA extract (within $\pm$ 1 order of magnitude) as being representative for water diffusivity in SOA. Clearly, further work is needed to prove that this choice is justified."

Referee #1: Figure 5. In the simulations, how much does the homogeneous freezing temperature change as the updraft velocities are changed? It would be useful to state

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this information somewhere in the manuscript.

Authors' response: We will add this information to the caption of Fig. 5. You may read out of panel c and a that it is about 0.5 K for the 3 m/s updraft velocity. Also, we notice that the color coding in Fig. 5a was accidentally interchanged. We will correct it in the revised version.

Additional change in revised version: The values in the first column of Table A1 are not given in $cm^2\ s^{-1}$ as stated, but in $m^2\ s^{-1}$. The numerical values need to be changed by adding +9.21 in order to be consistent with the stated unit. We will change those accordingly in the revised version.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 24473, 2015.