Atmospheric Chemistry and Physics Discussions

Interactive Comment

Interactive comment on "How quickly do cloud droplets form on atmospheric particles?" *by* C. R. Ruehl et al.

Anonymous Referee #3

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The authors report on measurements of the rate of cloud droplet formation on atmospheric particles sampled at four different sites in the US. Using a supersaturated column (SSC ; a similar instrument to the Droplet Measurement Technologies cloud condensation-nuclei counter "DMT-CCNC") with a modified particle detection system, droplet growth rates of ambient particles were measured and expressed as mass accommodation coefficient α, and compared to that of pure ammonium sulfate (AS) particles. A substantial fraction of particles grew at a rate smaller than that of AS. These particles were identified as "kinetically limited" to droplet formation.

Overall remarks:

The topic is certainly appropriate for publication in ACP. The measurement of the

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droplet growth rate and its expression as mass accommodation coefficient relative to that of AS particles has the potential to be adapted by other scientist in this field to deliver a more extensive dataset of accommodation coefficients interpreted and analyzed as done in this work. However, the capability of measuring the droplet growth rate is a unique feature of the modified detection system of the SSC which might be worth stated more exclusively in the text (i.e. briefly discuss if the used method can be adapted for the commercially available and widely used DMT-CCNs). The data is very well presented and the links between Köhler theory, mass accommodation coefficients and possible kinetic limitations to droplet formation is sufficiently discussed within the scope of this paper.

The only major concern with respect to the overall very good impression of the paper is the lacking support for some interpretations and conclusions given in the manuscript. The dataset is statistically not relevant enough to explain a) the observed daily variability in droplet growth rates at BON and GSM with descending air masses and cloud cycling, and b) the diurnal cycle observed in HOU by photochemical aging. The authors might think of rearrange the discussion and conclusion section more around the more robust statements, e.g. the last statement ("results suggest that aerosols containing CCN with α_{app} significantly lower than that observed for laboratory-generated AS are fairly common in the atmosphere ..., p 14246, 15), and expose the given interpretations for the above mentioned cases more as "ideas". Otherwise the manuscript is - in my opinion - ready for publication with only minor remarks the authors should take into consideration.

Specific remarks:

1) Section 2.2 Instrumentation: The explanation for the observed variation of the SSC flow rate ("possibly due to deviations from a parabolic velocity profile") should be discussed more extensively. Did other user of the CCN instrument encounter similar problems? What is a possible connection between a change in ΔT and the evolution of a steady-state velocity profile? Was this instrumental behavior only observed in the field

or also during the lab experiments? Based on the fact that a maximum of 20% of the data was affected a more detailed investigation/discussion might be of interest.

2) Section 2.2 Instrumentation: Why do you assume that meaningful values of α_{app} will be derived "as long as ambient particles with D (at 80% RH) from 100-250nm are sampled" (p 14243, 4-7)? How do you get the upper threshold? Is it given by the DMA?

3) The preconditioning of the particles to 80% RH upstream of the DMA and the size-selection using the DMA might bring a possible RH-history of the investigated droplets into play. The ambient particles are most likely internally mixed. Sampling the particles at ambient RH, expose them to 80% RH, and then using dry sheath air inside the DMA may alter the hygroscopic properties and the droplet growth rates of the particles entering the SSC compared to that upstream of the RH conditioner due to the loss of highly volatile compounds. If the DMA is running with sheath air of 80% RH to prevent this please state it in the text. Was the ambient sample in Houston heated to reach an RH of 80% (assuming an ambient of RH $>$ 80% at summertime conditions). Is there a chance that any ambient particles are not deliquescent at 80% RH? Please briefly discuss why 80% RH was finally chosen as working condition?

4) Section 3 Results: A table that summarizes a' uncertainties/boundaries (e.g. $\alpha'<10^{-1}$), the corresponding sigma-value, α'-descriptions (e.g. "very low-α'"), the used shortcut (e.g. f_{VL}), and the kinetic inhibition descriptions (e.g. "strong kinetic inhibition") might help the reader to keep track of the different cases.

5) Section 4 Discussion/conclusions: Not all back trajectories are shown for the entire set of days analyzed. In the case of an increasing kinetic inhibition when air came from aloft (BON and GSM) - was the opposite behavior observed in any of the analyzed cased (also including HOU and SGP), i.e. descending air masses but no increase in kinetic inhibition. This might disprove (or at least weaken) the authors' theory of cloud cycling being involved in altering the CCN properties of the investigated particles.

Technical remarks:

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1) Fig. 5-9, legends: the red dots refer to "$\alpha' > 10^{0.33}$" and not to "$<$"

Continuative remarks:

With respect to a further data analysis of the investigated cases: It might be helpful to employ the FLEXPART-Model (Stohl et al., 1998) for the interpretation of the air mass origin, too. The combination of the dispersion model and cluster analysis and especially the FLEXPART product 'footprint residence time' that accounts for the parcel's origin and it's source strength averaged over the lowest 150m above the surface would give a good indication of anthropogenic emissions/chemical compounds that might influence CCN properties. In the case of the Houston measurements the TexAQS II data server should provide this information.

Stohl et al., A replacement of simple back trajectory calculations in the interpretation of atmospheric trace substance measurements, Atmos. Environ., 36, 2002

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 14233, 2007.