Review of "Laboratory Studies of Collection Efficiency of Sub-micrometer Aerosol Particles by Cloud Droplets on a Single Droplet basis" by Ardon-Dryer, Huang and Cziczo.

General comment:

This manuscript introduces a new technique to measure the collection efficiencies between submicron aerosol particles and cloud droplets on a single droplet basis. This is a novel approach that can be used for other applications such as contact freezing. Aerosol particles and cloud droplets with sizes similar to in-cloud scavenging were used. Collection efficiency experiments were conducted for four aerosol particle sizes, one droplet size, two different aerosol particle concentrations and two different relative humidities at room temperature. The manuscript is sound and the use of the PALMS for these types of studies is encouraging. However, I have several questions that need to be clarified before the paper can be accepted for publication in ACP.

Major comments:

1. I am wondering if the authors accounted for particle losses. What was the transmission efficiency of the particles? Is it possible that the low collection efficiencies observed for particles smaller than 0.2 μm could be due to particle losses in the glass walls, the dryers at the end of the chamber, or in the transition from the chamber to the PALMS? In the absence of droplets is the aerosol particle concentration at the entrance of the chamber and before the PALMS comparable?
2. It is mentioned in the text that the flow within the chamber is laminar. Did the authors conduct computational fluid dynamic simulations to support this? If I interpret Figure 2 correctly, the neutralizer was placed inside the chamber. Does it have any effect on the laminar flow?
3. The authors indicate that the droplet size was 20 μm and that it was measured prior to the experiments. Did you measure the droplet size inside the chamber? Did you monitor the droplet size while running the experiments? What was the droplet size used for the collection efficiency calculations? Given that the RH inside the chamber is below water saturation, droplet evaporation occurred along the chamber. Was this accounted for in the CE calculations? What was the droplet size at the bottom of the chamber?
4. What is the residence time of the droplets within the chamber? The authors said: "*Average droplet evaporation time was calculated based on the average droplet size and the RH condition: 2.1 and 14.7 s for the Low and High RH cases, respectively*". This means that the residence time of the particles was below 2.1 s? I am wondering if the 20 μm droplets did not completely evaporate during the low RH experiments (i.e. 15%).
5. Ladino et al. (2011) showed the high variability in measuring collection efficiencies from the bulk collection of coagulated particle-droplets. Does this also apply to the single droplet basis analysis? How reproducible are your coagulation experiments? If I interpret Table 3 correctly, each coagulation experiment was conducted only once.
6. I think that the atmospheric relevance of your results needs to be clearly stated in the conclusions. This is currently missing.

7. What did we learn from the single particle basis analysis compared to the bulk analysis? Is it better to use the single particle basis approach? Why?
8. How were the CE uncertainties calculated? What is the meaning of the error-bars reported in Figures 5, 6 and 8.

Minor comments:

1. Brownian motion is very effective at promoting collisions of aerosol particles cloud droplets at aerosol particle sizes smaller than 0.25 μm as shown experimentally by Lai et al. (1978) and Ladino et al. (2011); however, this was not observed in this study. Can the authors discuss this? Why do you think you were unable to clearly see the theoretically predicted Greenfield Gap? Why this was experimentally observed by Lai et al. (1978) and Ladino et al. (2011) and not in the present study?
2. I am wondering why the aerosol particle concentration was measured with two different instruments. It is clear that the OPC has a lower operational limit higher than the smallest studied particles; however, the CPC has the capability to count/measure particles with sizes similar to those used in this study. Why you did not use the CPC for the whole set of experiments? Are the uncertainties from the CPC and OPC comparable? Why did you size select the small particles (0.025 µm, 0.125 µm) and not the large ones (0.25 µm and 0.475 µm)? PSL spheres are supposed to hold a specific size (that is the reason they are used as standards for calibration). What was the motivation to size select them?
3. Which was the motivation to choose these theoretical models to inter-compare the experimentally obtained collection efficiencies? The used theoretical models were developed for below-cloud scavenging where rain drops are included instead of the small droplets used in this study.
4. The authors showed that there was not any difference in the CE values when the aerosol concentration was increased from 50 to 100 cm^{-3}. Why would you expect to see a difference here? The CEs are normalized by the total particle concentration; therefore, there should not be any effect. "*Wang and Pruppacher (1977) used RH condition similar to that used in this work but with a higher aerosol concentrations*". Why would a higher concentration result in higher CEs?
5. On page 6219, lines 11-12 it is written: "*it is possible the size and charge conditions offset each other, lending to the comparison to our data*." In order to confirm this hypothesis, can the authors infer from your calculations how much the CE will increase/decrease when the droplet size is increased from 20 to 200 μm or when the charges are increased from 400 to 5x10^5?
6. Fig. 6 and its corresponding discussion: The Wang and Pruppacher (1977) data needs to be used with caution. They used different droplet sizes but this is neither mentioned in the figure nor in the text. This needs to be clearly stated because in its current form it seems like they run different experiments with one droplet size only and 0.25 μm aerosol particles. I suggest to only use one data point from the Wang and Pruppacher (1977) study (i.e., CE for 0.25 μm aerosol particles and 150 μm droplets)

Technical comments

1. In some cases the droplet size is said to be "~20 μm" but on page 6217 line 7 it is said that the size is 22 μm.
2. Page 6208, line 22: It should be IPCC
3. Coagulation and collection are used throughout the text. I suggest sticking to one of them.
4. The term "coagulated droplets" is used in several places. I am not sure if this will be clear for readers. Can the authors use a different term?
5. Brownian motion and Brownian diffusion are used throughout the text.
6. In many places either ~20 μm or ~80% are used when referring to the droplet size and the relative humidity. The authors measure the size of the droplets and the RH quantitatively. What is the need to use the approximation symbol (i.e., "~")?
7. Page 6209, line 2: I think Ladino et al. (2011) is not the best reference here. I suggest to replace it with a more appropriate reference (e.g., Rasch et al. (2000) and Croft et al. (2009))

References

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