We would like to thank reviewer 1, Dr. Luis Ladino, for the helpful comments and suggestions. In line with the comments and suggestions, we revised the manuscript. Below are the comments (in bold) followed by the replies. The parts that are in italic are corrections that are included in the revised version of the paper:

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?

Information on particle losses was added to the paper under the experimental setup section: *Particle losses were calculated by measuring the particle concentration at the entrance and at the bottom of the chamber (i.e., before PALMS). Particle losses were 14±10%.*

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?

Information about the neutralizer was added to the paper under the experimental section: *A neutralizer, containing two Polonium-210 strips (0.64 cm thickness and 15 cm long), is placed in the lower part of the DGN.*

Information on laminar flow was also added to the paper under the result and discussion section: *Calculations of Reynolds number were performed using the experimental conditions and chamber geometry. Reynolds numbers from 0.12 to 0.16 were calculated and, based on this, we assume the aerosol particles and droplets interact in flow condition close to laminar throughout the chamber.*

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 2.1s? I am wondering if the 20 µm droplets did completely evaporate during the low RH experiments (i.e. 15%).

Information on the droplet size was added to the paper under the experimental setup section: *Due to the position of the camera, droplet size could not be monitored during an experiment or within the chamber. Droplet size was, however, measured before and after the experiment, and the size was constant within the quoted uncertainty. Droplets size during experiments was also verified by the residual size after the droplets evaporated.*

The droplets completely evaporated in both RH conditions; the evaporation was verified by measuring the AS residual sizes with the OPS and by using an evaporation model calculation. Information on droplets residence time was added to the paper under the result and discussion section: *Total droplet evaporation time (i.e., residence in the generator section and experimental chamber) was calculated based on the average droplet size and the RH condition: 2.1 and 16.6 seconds for the Low and High RH cases, respectively. The droplets residence time in the chamber was 0.7 and 6.1 seconds, for the Low and High RH cases, respectively.*

Explanation on the calculation of CE was added to the paper under the result and discussion section: *CE value was calculated for each experiment, based on the average droplet size measured from each experiment and when similar RH, aerosol size and concentration conditions were used.*

Since all three reviewers asked about the droplet sizes due to evaporation and the effect it has on the CE, we decided to change our CE calculation in order to include the fact that droplets evaporate in the chamber. In addition, we include a new paragraph on the subject in the result and discussion section. The droplet size at the time that collection occurred is not measured in our system; therefore, we used different droplets sizes that corresponded to the range of evaporation times in the system in order to calculate theoretical CE values. The following was added to the paper in the result and discussion: *As noted earlier, the droplets evaporated completely while in the chamber at both RH conditions. Since droplet size could not be determined precisely at the moment when collection occurred in the chamber, calculations of theoretical CE were performed for three relevant droplets sizes: The first was the original droplet size as measured from the droplet generator (21.4 and 21.9 µm, for Low and High RH conditions, respectively) for the full droplet lifetime. The second, droplet size with half the volume of the original droplet (radius of 17 and 17.4 µm, for Low and High RH conditions, respectively) over the full lifetime. For the third an extreme case was considered, droplets with a radius of 5 µm for the full droplet lifetime. The results of these calculations are presented in Fig. 10. Overall, as droplet size decreases, CE values increases. In the extreme 5 µm case, CE values increases by more than an order of magnitude. For the Low RH case the best agreement is with the 5 µm case, which logically follows from the rapid evaporation of these droplets. In the High RH case the experimental CE values fall nearest the half volume case, which again logically follows since these droplets more slowly evaporate.*

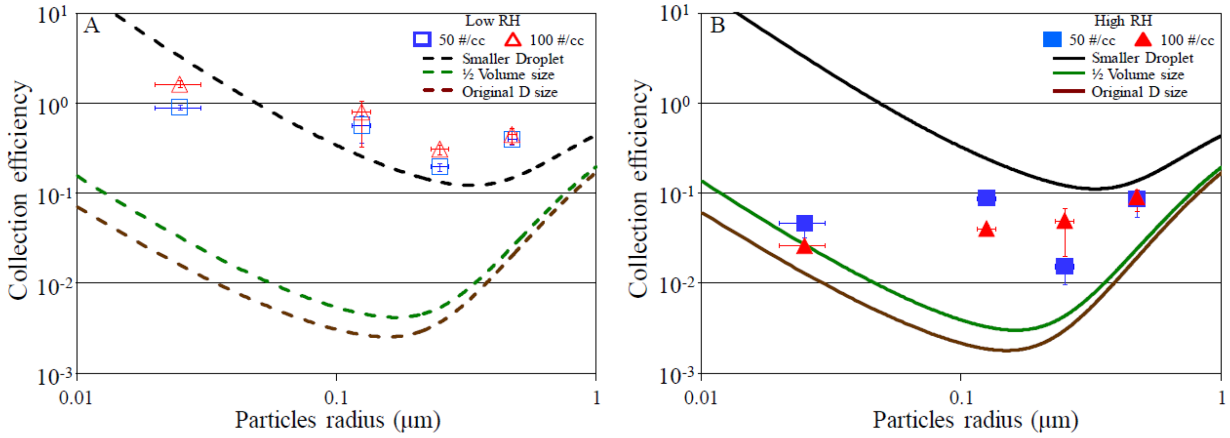

Figure 10: CE as a function of particle radius at Low and High RH (Panel A and B, respectively). CE experimentally determined in this study (points) with theoretical calculations

(lines). The lines represent calculation with different droplets sizes: the measured droplet size (brown), droplets with half the volume (green) and 5 μm droplets (black). See text for details.

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.

By definition in a single particle experiment, each droplet residual is a sample. We believe this is a noteworthy advantage of this type of experiment and it should not be compared with the different off-line analysis results. Instead, each 'experiment' in this work contains more than 1000 droplets evaluated on a single droplet basis. For clarity, Table 3 summarizes all measurements per experimental condition.

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?

Based on Dr. Ladino's suggestion the following was added to the conclusions section: *This technique overcomes some of the limitations inherent in previous studies which required a bulk collection of material. The analytical methods employed were limited by issues such as signal to noise and an inability to observe multiple collection events on single droplets. Moreover, very few experimental works have been performed with atmospherically relevant particles sizes (Radke et al., 1980; Andronache et al., 2006), another advantage of this technique. The droplet size and charge state used here are also consistent with atmospheric conditions.*

8. How were the CE uncertainties calculated? What is the meaning of the error-bars reported in Figures 5, 6 and 8.

CE uncertainties calculations are based on droplet size, aerosol size and aerosol number concentrations, which were measured in each experiment. This information was added to Fig. 5 caption: *CE calculated as a function of particle radius. Shapes represent different aerosol concentrations. CE error bars based on droplets size, aerosol size and aerosol number concentration measured from each experiment as describe in Eq. 3.*

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?

An explanation about this difference was added to the paper under the result and discussion section: *Moreover, as described by Tinsley et al. (2001), the electrical effect is more important for smaller particle sizes (< 0.1 µm) than Brownian diffusion. This could explain why the Greenfield Gap is highly pronounced in the data in Fig. 6, while it is more pronounced in the data of Lai et al. (1978) and Ladino et al. (2011).*

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?

The SMPS is optimal for sizes smaller than 1µm radius while the OPS is optimal for particles larger than ~0.35 micrometers. The OPS is also able to identify the aerosol size distribution more rapidly and is sensitive in the range of the droplet residuals, the latter of which cannot be detected with the CPC.

Explanation was added to the text: *Large particle (diameter >0.35 μm) and residual concentrations were measured by an Optical Particle Sizer (OPS; TSI, Inc., Shoreview, MN Model 3330). Particles, below diameter of 0.35 μm were measured using a Scanning Mobility Particle Sizer (SMPS) consisting of a Differential Mobility Analyzer (DMA; BMI, Inc. Model 2002) and a condensation particle counter (CPC; BMI, Inc. MCPC Model 1710). Similar concentrations were observed in the overlapping sensitivity region of both instruments.*

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.

Information was added to the paper under the Theoretical CE Models section: *These theoretical models include the known forces that affect CE values and which were measured or constrained by data in the experimental measurements presented here. It should be noted that although these theoretical models were developed for large droplets they have been used to calculate CE for sizes relevant to this work (Ladino, 2011).*

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?

We thank Dr. Ladino for this comment and we agree with the reviewer that CE is normalized. Based on the comment we clarify the sentence: *Wang and Pruppacher (1977) used a RH condition similar to that used in this study but with a higher aerosol concentrations. It is* *expected that a higher aerosol concentration will increase the chance of collision between particles and droplets, which will increase the value of ECR, but will not affect CE, which is normalized.*

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?

As requested by Dr. Ladino, calculations of CE values were made for droplets sizes of 20 μm and 200 μm with droplets charges of 400 and $5x10^5$ elementary charges. These calculations were added to the result and discussion part of the paper: *It is known that droplets carrying higher electric charge have higher CE (Barlow and Latham, 1983; Byrne and Jennings, 1993; Pranesha and Kamra, 1997a,b; Tinsley and Leddon, 2013; Tinsley et al., 2000; Tinsley, 2010), and this is consistent with our data in Fig 9. Droplets size also affects CE, where smaller droplets have higher CE values (Lai et al., 1978; Pranesha and Kamra, 1996). Fig. 11 shows a calculation of CE based on different droplet charges and sizes. Two droplets sizes were used: 20 μm, which is similar to the size used in this study and by Ladino et al. (2011) and 200 μm, which is the size used by Wang and Pruppacher (1977). Three different droplet charges were considered: 400 elementary charges, as used in this study, $5x10^4$ elementary charges, used by Ladino et al. (2011) and $5x10^5$ elementary charges, the lower limit of charges used by Wang and Pruppacher (1977). Shown in Fig. 11, CE values increase as droplet charge increases. Droplets size and charge conditions can counteract each other in the case of larger droplets (lower CE) with higher charge (higher CE). We suggest this may explain the agreement found between the CE values measured in this study and those of Wang and Pruppacher (1977) and the disagreement between our values and those of Ladino et al. (2011). It should be noted that the experimental CE values fall within the region of the 20 μm case. The CE values of the small particles (<0.1 μm) match the theoretical CE, while for larger particles (>0.1 μm) they are slightly higher. These differences could be a result of some conditions not modeled theoretically or conditions difficult to constrain experimentally, as discussed above.*

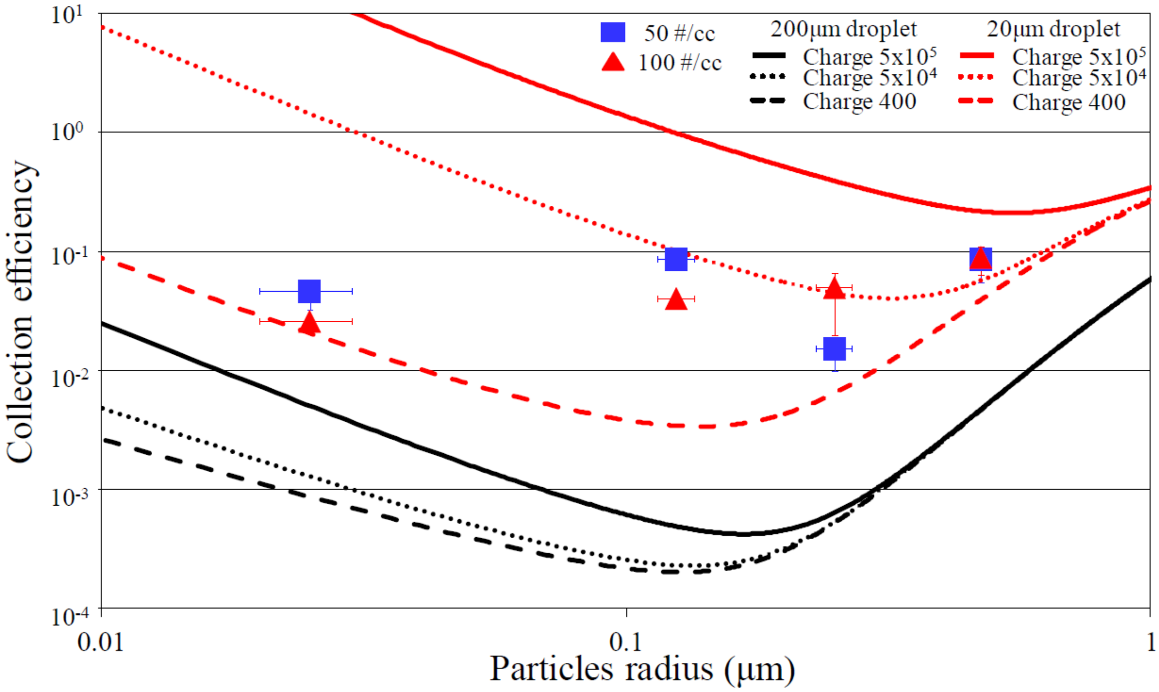

Figure 11: CE as a function of particle radius at High RH condition. CE experimentally determined in this study (points) with theoretical calculations (lines), where the charge number is in elementary charge units per droplet. Black lines are for CE of 200 µm droplet size and red for 20 µm droplet size.

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)

Note the information about Wang and Pruppacher (1977) droplets sizes is mentioned in the paper under the result and discussion section: *For example, Wang and Pruppacher (1977) and Lai et al. (1978) used somewhat larger droplets (of 170-340 µm and 620 µm, respectively).*

In order to clarify this point we added the droplets sizes to Fig. 6 caption: *Black diamonds are from Ladino et al. (2011), RH 88±2% with aerosol concentration 2000* cm^{-3} *and droplets size of 12.8-20.0 µm. Brown diamonds represent are from Wang and Pruppacher (1977), RH of 23±2% with aerosol concentration of about* 10^{17} cm^{-3} *and droplets size of 170-340 µm. Pink diamonds are from Lai et at. (1978), when 620 µm droplets were used; there was no information provided regarding the RH or aerosol concentration.*

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.

Based on the reviewer technical comments 1 and 6 changes have been made, droplet size was written as 21.6 µm.

2. Page 6208, line 22: It should be IPCC

We thank the reviewer for this correction ICCP was changed to IPCC.

3. Coagulation and collection are used throughout the text. I suggest sticking to one of them.

Changes have been made; the term collection was used throughout the paper.

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?

We believe that the definition "coagulated droplets" describes the process clearly, based on this comment we add an explanation to this term: *"Coagulated droplets" (droplets that collected aerosols) had mass spectra that contain signatures from both an aerosol particle and a droplet residual.*

5. Brownian motion and Brownian diffusion are used throughout the text.

Changes were made, the term Brownian diffusion was used throughout the paper.

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., "~")?

Changes were made, the symbol ~ was removed from the paper and quantitative values (such as 21.6 µm and 88%) were used throughout the paper.

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))

We thank the Dr. Ladino for this suggestion changes were made accordingly.