We would like to thank reviewer 3, Dr. Alexei Kiselev, for the helpful comments and suggestions. In line with the comments and suggestions, we revised the manuscript. Below are all 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:

Specific comments:

Introduction:

1. The motivation of this research is not completely clear from the introduction. Scavenging of the aerosol particles by cloud droplets is a part of aerosol and cloud interaction process and as such undoubtedly contributes to cloud dynamics (including precipitation) and radiative properties. The questions remains, however, what is the contribution of the uncertainty associated with the scavenging efficiency into the overall uncertainty of radiative forcing due to the indirect aerosol effect? To justify the necessity of a sophisticated experiment aimed to quantify the collection efficiency (CE) this connection has to be discussed at least briefly.

We would like to thank Dr. Kislev for this suggestion. We made several changes in the introduction section: *The interplay between aerosol particles and water droplets in the atmosphere, especially in clouds, influences both aerosol and cloud properties. The major uncertainty in our understanding of climate arises from this interplay: the ability of an aerosol to affect cloud formation and, consequently, alter the global radiative balance (IPCC, 2007). When an aerosol particle comes in contact with a water droplet, the interaction can result in a collision followed by coalescence of the two. This process is known as "collection" or "coagulation". The collection process is considered an important mechanism that can "scavenge", and thereby remove, aerosol particles from the atmosphere (Starr and Mason, 1966; Owe Berg et al., 1970; Hampl and Kerker, 1972; Pranesha and Kamra, 1996). Collection can also affect cloud dynamisc, the precipitation process and cloud lifetime, and thereby change the global radiation budget (Rasch et al., 2000; Croft et al., 2009).*

In supercooled clouds, where droplets are present at temperatures below 0ºC, the collection process can have an effect on precipitation when the contacting aerosol initiates ice nucleation. The result is the creation of an ice crystal, a process known as "contact nucleation" (Vali, 1996). Contact can influence cloud lifetime and precipitation formation in mixed-phase clouds, which will also affect the *global radiation budget. In order to understand the contact freezing process, it is important to determine the efficiencies at which the aerosol particles collide with a liquid droplet within a cloud.*

2. Although numerous theoretical and experimental studies of the aerosol scavenging are listed in the introduction, it remains unclear what is the status quo in this research field? Is the theory insufficient to describe the CE in most cases? Were previously reported experimental results very far from the theoretically predicted values? Are there any specific cases where observed collection efficiency could not be explained by accurate consideration of all droplet-particle interaction mechanisms?

Based on the reviewer comment a new paragraph was added to the paper under the introduction section: *Theoretical calculations of CE in a cloud environment have been the subject of many studies, driven by the necessity to explain aspects of both warm and cold precipitations. An experimental validation of the theoretical knowledge related to CE, particularly for droplet–aerosol collisions, is difficult and far from complete (Ladino, 2011). According to Santachiara et al. (2012), significant discrepancies between theoretical and experimental studies have been found for sub-micrometer particles in the "Greenfield gap", and the measured values can be one to two orders of magnitude higher than predicted. According to Wang et al. (2010), this disagreement could be because some physical processes considered in theoretical models are neglected, difficult to represent or hard to control in experimental studies.*

Additional information about this comparison was also added to the result and discussion section: *Differences between theoretical and measured CE may be considered a result of conditions not modeled theoretically or difficult to constrain experimentally. Possibilities include rare multiply charged particles, aerosol droplet electric interaction that are not fully considered (such as the induced dipole force), the evaporation rate of the droplets, variable terminal settling velocity due to changes in droplet size, and the present of solute in the droplets.*

3. The authors put a special stress on the statement that the presented work is the first experimental study of CE "on a single droplets basis". I believe this is not exactly so: (Hoffmann et al., 2013) has reported the determination of the collection efficiency measured for

the individual droplets levitated in the electrodynamic balance. Though dominated by electrostatic forces, the experimental CE was in a good agreement with theoretical predictions and the multiple collection events have been explicitly taken into account.

A correction was added to the paper under in abstract and the Introduction sections:
To our knowledge, this is the first collection experiment performed on a single droplet basis with atmospherically relevant conditions such as droplets sizes, droplets charges and flow.

To our knowledge, no previous study allowed for determination of collection on a single droplet basis with atmospherically relevant conditions of droplets size, droplets charge and flow, which are a key to many cloud processes, including contact nucleation.

Experimental methods

4. If I interpret the figure 2 correctly, both droplets AND *aerosol particles* are passing the neutralizer at the lower end of DGN section! In this case, the initially single charged aerosol particles should assume Boltzmann distribution centered on zero charge. The free path of alpha particles produced by Po-210 source (5 to 7 cm in air) is sufficient to produce homogeneous concentration of ions inside the DGN section for that. It seems that the authors are aware of this effect (see also comment 10). If so, the question arises if the true charge distribution for aerosol particles after the neutralizer has been taken into account for the calculation of theoretical CE?

Dr. Kislev is correct, the particles will experience a Boltzmann distribution, where most particles have zero charges and most charged particles have ±1 elementary charge. Per Dr. Kislev comment, an explanation and correction was added to the paper under the result and discussion section: *These calculations were made for charged particles that contained one elementary charge per particle. Most particles in a Boltzmann distribution contain no charges and will therefore not be affected by electro-scavenging forces. The most common charge state other than neutral is a single charge, about 10% of particles, and this forms the basis of our calculation (Hinds, 1999). This is further supported by a decreasing effect of multiple charges when considering the effect on CE, (Fig 8).*

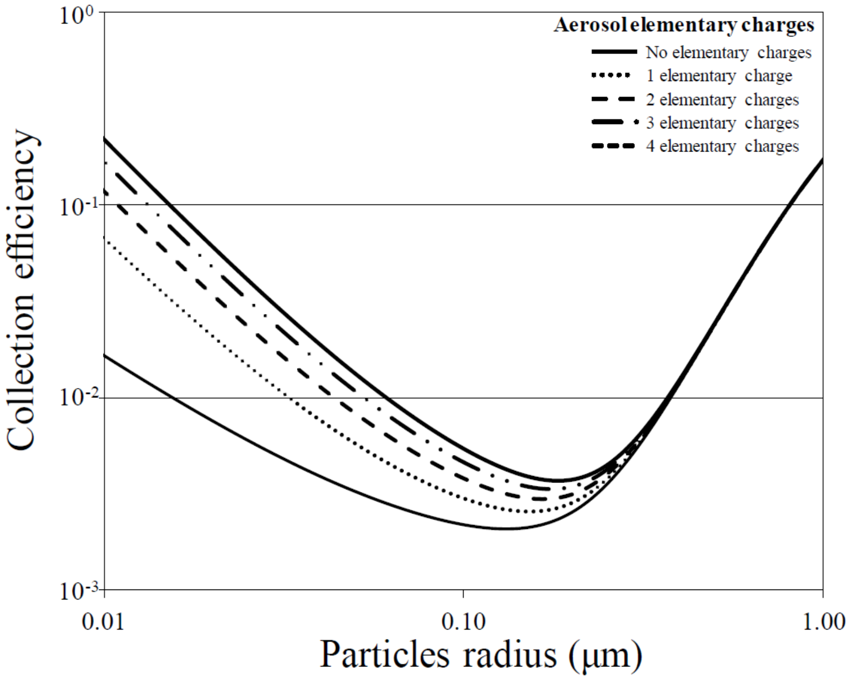


Figure 8: CE values for 50% RH and 400 elementary charges per droplets with different particles elementary charge for a droplet radius of 21.6 µm and room temperature.

Data Analysis

5. The calculation of the CE (equation 3) is done under assumption of constant droplet size during the droplet fall through the chamber. However, the evaporation time of the droplets is obviously shorter than the residence time (which I was unable to determine exactly because the flow rate in the chamber is not given). The experimental values of CE cannot be correct if the droplet size is reduced by factor 10 or more after travelling just a few centimeters! How this reduction of the droplet diameter is taken into account in the theoretical calculation?

Per Dr. Kislev's comment, Information on the flow rate and droplets residence time were added to the paper under the result and discussion section.

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.*

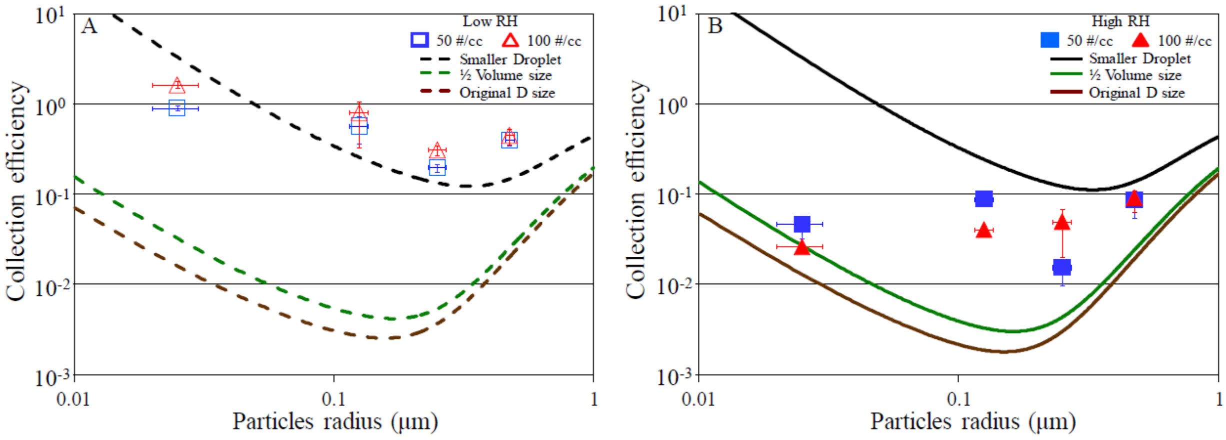


Figure 10: CE as a function of particle radius at Low and High RH (Panel A and B, respectively). CE experimentally determined in this study (points) with theoretical calculations (lines). The lines represent calculation with different droplets sizes: the measured droplet size (brown), droplets with half the volume (green) and 5 µm droplets (black). See text for details.

Result and discussion

6. The evaporation time of the droplet (page 6217 line 10) is given for pure water (at least I obtain the same values if calculating the evaporation time for pure water droplets using the

formula in (Hinds, 1999). However, droplets of aqueous ammonium sulfate solution have been used, meaning that the Raoult term has to be taken into account. Under "high-RH" conditions droplet would not evaporate completely (efflorescence RH is not reached) so that the solute droplet of approximately 1.3 µm diameter (corresponding to the equilibrium saturation ratio of 0.88) is left. Note that this droplet will have the same charge so that the electrostatic interaction with aerosol particles would be completely different (see discussion of the electrostatic interaction). The same considerations apply to the dry residual of the ammonium sulfate left by evaporating droplet under "low-RH" conditions.

We agree with Dr. Kislev that the residual presented at the end of the chamber (based on OPS distribution) was at about 1.3µm. A collection experiment of PSL with AS residual (evaporated droplet), with similar sizes as mentioned by the reviewer, were performed. Several thousand spectra were examined with PALMS with no coagulation event observed. This information was added to the paper under the experimental setup section: *It should be noted that an experiment of PSL with AS residual (from the evaporated droplet) was performed. Several thousand spectra were examined with PALMS but no collection event was observed.*

7. The number of elementary charges carried by the droplet in the study of (Lai et al., 1978) cannot be as high as 10^n (page 6219, line 5). I assume the authors used the charge of the droplet given in the table 2 of (Lai et al., 1978) where the minus sign in the exponent is erroneously omitted ("…Average charge x10^{10}C …").

We would like to thank the reviewer for finding this mistake, the values were corrected accordingly, and they are now $6.6x10^8$-$1.9x10^9$ elementary charges.

Theoretical CE models

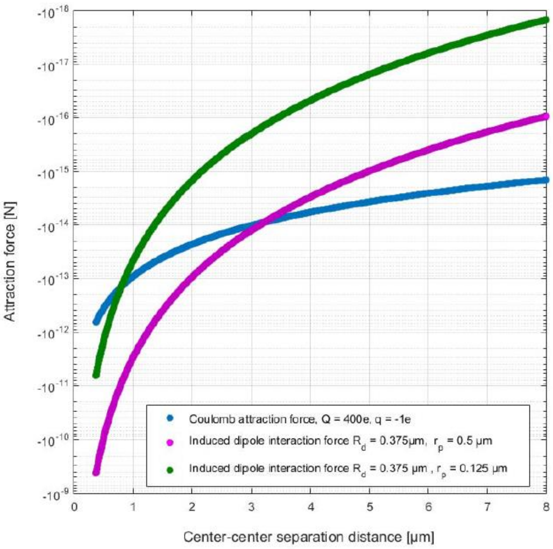
8. More details should be provided on the theoretical calculations. Is the terminal settling velocity kept constant together with the size? Is the presence of solute (ammonium sulfate) is taken into account in the calculations of phoretic forces? Is the true charge distribution of aerosol particles taken into account for calculations of electrostatic interaction? I believe that

taking into account all these effects together with careful consideration of droplet evaporation would allow for much better agreement between measured and calculated values of CE.

This comment is related to earlier point 5. Please see response to point 5 above and the figure addition to the paper.

In addition, information on parameters that could not taken into account for the theoretical calculation were added to the paper: *Differences between theoretical and measured CE may be considered a result of conditions not modeled theoretically or difficult to constrain experimentally. Possibilities include rare multiply charged particles, aerosol droplet electric interaction that are not fully considered (such as the induced dipole force), the evaporation rate of the droplets, variable terminal settling velocity due to changes in droplet size, and the present of solute in the droplets.*

9. I wonder if the effect of electric charge should be considered more thoroughly for the theoretical calculation of CE. The long-range electrostatic interaction between the charged droplet and the aerosol particles is correctly identified as Coulomb attraction (equation 9 of the manuscript). However, at short distances comparable to the size of evaporated droplet, the induced dipole interaction has to be considered (equation 13 in Hoffmann et al., 2013; Tinsley, 2010; Tinsley et al., 2000). For small droplets carrying strong charge (on the order of 500 elementary charges), this interaction force dominates all others and can significantly increase collection efficiency. In the figure below, I illustrate my point comparing Coulomb and induced dipole interaction forces between the droplet residual particle (0.75 µm, 400e) and single charged PSL particle of 1 µm (magenta curve) and 0.25µm (green curve) as a function of distance between the centers of the particles. The negative sign of the force denotes attraction between the particles. Note that for larger PSL particle at short separation distances (below 3 µm from center to center) the induced dipole interaction force is much stronger than the Coulomb attraction force (blue curve), potentially increasing the coagulation probability between PSL and residual particle.



Dr Kislev raised a very interesting point here. It is clear that induced dipole interaction is a stronger force than Coulomb attraction force that could increase the electro-scavenging force and the CE values. Unfortunately, our experimental work and the theoretical calculation could not examine such small-scale separation between the droplets and PSL, therefore such information could not be included in the paper. Indeed, to our knowledge, these types of theoretical models are not used for this length scale of effect in the models used in this paper. The problem is that inclusion of a new force at this scale is beyond the scope of this paper and would also not allow for direct comparison with previous calculations which is fundamental to this work. We agree with the importance and, based on Dr. Kislev's comment, we included the following: *Differences between theoretical and measured CE may be considered a result of conditions not modeled theoretically or difficult to constrain experimentally. Possibilities include rare multiply charged particles, aerosol droplet electric interaction that are not fully considered (such as the induced dipole force), the evaporation rate of the droplets, variable terminal settling velocity due to changes in droplet size, and the present of solute in the droplets.*

10. Page 6220 line 20: "*One elementary charge was used for the particles, consistent with a Boltzmann distribution imparted by the neutralizer*". Again, a Boltzmann distribution is centered around the zero charge, not single charge of any sign. For particles of 0.01 µm (radius) 90% of all particles would have no charge, whereas for particles of 5µm 70% of all particles will carry more than 3 elementary charges (of any sign) (see Chapter 15.7 in Hinds, 1999, sec. ed.).

Dr. Kislev is correct, per Dr. Kislev comment, an explanation and corrections were added to the paper, see our reply to comment 4. For this paper it should be noted the aerosol would not extend to the super-micrometer size so 10% is the likely value to consider here.