**Review of the manuscript acp-2015-5 "Laboratory Studies of Collection Efficiency of Sub-micrometer Aerosol Particles by Cloud Droplets on a Single Droplet basis" by K. Ardon-Dryer, Y.W. Huang, and D.J. Cziczo**

The manuscript is describing a novel approach in measuring the scavenging efficiency of water droplets freely falling in the aerosol-containing airflow. The novelty of approach is ensured by using PALMS instrument to detect the collision events on a single droplet basis. This approach could be very promising for studying aerosol-cloud interactions under well-defined laboratory conditions. Studies of turbulent scavenging of aerosol by droplets or ice crystals and contact freezing of supercooled droplets would be the applications that could greatly benefit from reliable quantification of collection efficiency (CE). The simplest case of aerosol scavenging by droplet freely falling through the laminar airflow containing aerosol particles is realized in the presented experiment. Unfortunately, the evaluation of the experimental results is suffering from several non-valid assumptions, which are strongly reducing the quality of the data analysis. My main concern is the assumption that the droplet size (and associated magnitudes like terminal settling velocity, volume swept by the droplet etc.) is not changing during the residence time in the chamber, which, as authors admit themselves, is not the case. It is astonishing that the CE values derived under this assumption from the experimental data are in such a good agreement with theoretical predictions (figure 8). At the same time, the quality of the experimental work is sufficiently high to allow the re-evaluation of the primary data and therefore the authors should spent some effort to revise the manuscript based on the improved data analysis. I hope my remarks would help to achieve this goal.

*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.

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?

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.

## 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?

## 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?

## 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.

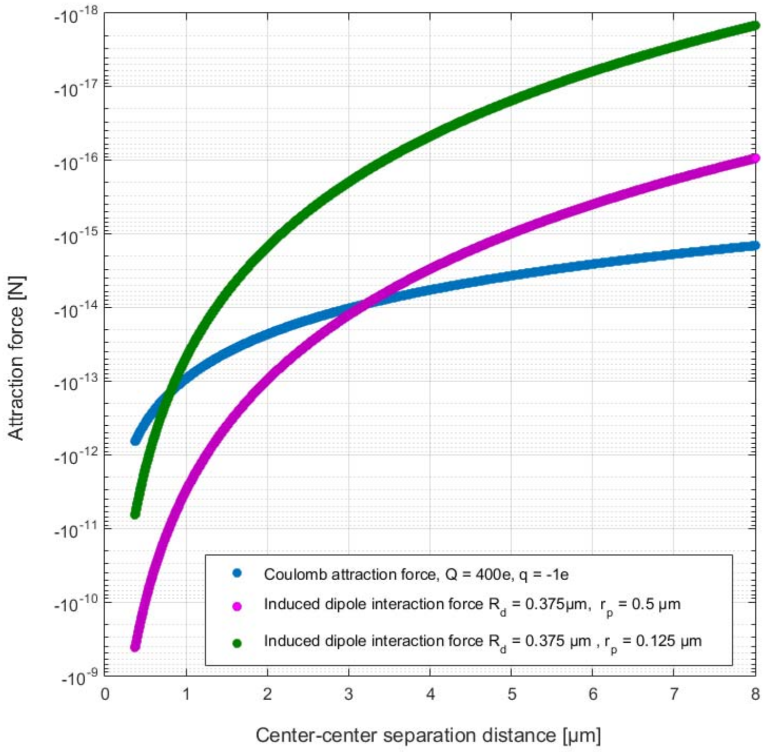
7. The number of elementary charges carried by the droplet in the study of (Lai et al., 1978) cannot be as high as $10^{29}$ (page 6219, line 5). I assume the authors used the charge of the droplet given in the table 2 of (Lai et al., 1978) where the minus sign in the exponent is erroneously omitted (“…Average charge x$10^{10}$C …”).

## 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.

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.



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.)

**References:**

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