

## **Reviewer #2 comments**

Dépée et al., “*Laboratory study of the collection efficiency of submicron aerosol particles by cloud droplets. Part II - Influence of electric charges*”.

- For information, this reviewer is also a reviewer #2 of the companion paper by Dépée et al.: “*Laboratory study of the collection efficiency of submicron aerosol particles by cloud droplets. Part I - Influence of relative humidity*”.

- The present paper presents a measurement of the collection efficiency of aerosol particles by sedimenting raindrops under various electrical charge states of the raindrops and aerosol particles. The electric force between a charged aerosol particle and a charged raindrop has two components: 1) the long-range Coulomb force between charges and 2) the short-range force due to an induced image charge distribution on the raindrop (independent of its net charge state) by the charged aerosol. The latter force is always attractive and dominates when the aerosol is within a few droplet radii of the droplet surface, whereas the former dominates at greater distances and is either attractive (for unlike charges) or repulsive (for like charges). These mechanisms are collectively known as electroscavenging of aerosol particles.

- Cloud droplets – even in warm clouds – are frequently charged. Moreover, aerosol particles also may become charged at the few  $10e$  levels due to the evaporation of charged droplets or space charge effects at the edge of cloud layers in Earth’s electric field. So electroscavenging is likely to be an important microphysical process for atmospheric aerosol and cloud microphysics. However, although it has been fairly extensively treated theoretically, there are very few experimental data on electroscavenging. There was considerable experimental effort on the subject in the 1970-80’s, but with limited experimental control and resulting measurements that differed by several orders of magnitude.

- The experimental measurements reported in this paper are therefore essentially unique. The experiments are carefully executed. The authors have demonstrated in Part I of their laboratory study that their experimental apparatus is capable of precise measurements of the raindrop-aerosol capture efficiency (CE) (in the former case, measuring the effects of thermophoresis and diffusiphoresis). The authors have already published a theoretical model that includes electroscavenging (Dépée et al., 2019), which agrees well with a similar model previously reported by Tinsley and co-authors. As for the Part I paper, I have no hesitation to recommend that the Part II paper also be published in ACP, after responding to the comments below.

## **General comments**

- I recommend that the authors apply the same general comments that I made for the Part I paper also to the present paper. In particular the authors should ask a native English speaker to edit the manuscript for poor English grammar and lengthy sentences.

- There is a lot of cross-referencing with the Part I paper, and many parts (main apparatus description, derivation of the collection efficiency, ...) where texts (and some figures) are repeated almost verbatim. I leave it to the editor and authors to decide if it is best to keep these as two separate papers or to make the Part I and Part II papers as two large chapters of the same paper.

- The most surprising aspect of the paper is that the authors have made some unique and impressive measurements *and* have developed a careful theoretical model yet there is only one figure (Fig. 10) where they compare their measurements with their model. Why do they omit any comparison of their model with their data in the other two figures (Figs. 7 and 10) where they are presented? Without this comparison it is hard to get confidence that their data do indeed verify the short range image force attraction. (I will pick this point up below.)

## Specific comments

l.27: Replace “the neutralisation” with “zero”.

l.30: Replace “correlation of Kraemer and Johnstone (1955)” with “prediction of Kraemer and Johnstone (1955)” (and elsewhere in the text).

l.34: Replace “on” by “on”.

l.49 Replace “is mainly depending” with “mainly depends”.

l.71-74: Please clean up this sentence.

l.114-115: Please clean up this sentence.

l.121: Remove “a”.

Fig.1: See the comments I made for Part I. I suggest you add some more trajectories to panel D that show aerosol particles outside the geometrical path of the raindrop, which are attracted into a collision (or not).

l.141: Indicate the AP (air) flow velocity and transfer time in the chamber.

l.144: Replace “go out” with “pass out of”.

Fig. 2: You do not show in Part II but we see from Part I that the APs are introduced into the sheath region of the laminar flow down the tube, whereas the droplets fall down the centre of the tube. The APs have charges between 25e and 150e. What is their number concentration (cm<sup>-3</sup>)? I could not find this anywhere in the text and it is an important number. The APs will form a space charge in the tube that pushes them to the walls and away from the central region where the droplets fall. How big is this effect? Does it influence your estimate of the AP number concentration seen by the falling droplet?

Fig.2: You estimate the mean AP number concentration in the main tube with the HEPA filter. Charged aerosol will have higher losses in the main tube and in the pipes leading to the neutralizer. How big is this loss and is it corrected for when estimating the mean AP number concentration in the main tube?

l.175: Replace “the atmospheric one” with “one atmosphere”.

l.181: Replace “So,” with “In this way”.

l.182: Replace “was” with “were”.

l.198: replace “get” with “are”.

l.210: Replace “varying” with “varying”.

Fig. 5: This is a very poor figure. The “3D printing” is a black blob with no detail. It conveys no information. And what is a “3D printing”? If you mean that the piezoelectric droplet generator is installed in a housing made with a 3D printer, then state that. I suggest a simple line schematic cross section should be provided to replace the three objects in this figure.

Fig. 6: Replace “Charging low of the electrostatic inductor colors” with “Droplet charge versus electrostatic inductor voltage. The colours”.

l.262: Replace “the double” with “doubly-“.

l.266: Replace “more” with “greater”.

Section 2.2: This is a verbatim copy of Section 2.1 in Part 1. Please see earlier comments.

Section 2.3: Another verbatim section (which simply refers to the Part I section). These are examples that argue the two parts should be combined since it should not be necessary to read a separate paper for this information.

Eq.5: The variables in this formula are undefined. It is not sufficient to refer to a separate paper to define the variables.

Eq.6: I suggest you add a figure to show the relative importance of these two force terms – the Coulomb term and the image charge term - versus radial distance, under the experimental conditions of the present paper. If the other dynamic forces can also be indicated for comparison, so much the better.

l.326: What is the meaning of the ambiguous word “global”? If it indicates “mean” then use “mean”.

l. 341: Replace “repulsing” with “repelling”.

l.341: Replace “the fact that” with “whether the”.

l.343-347: The short-range attractive force needs to be pointed out in Fig.7. I assume it is the small rise in CE at positive  $q \times Q$ ?

Fig.7: Add a vertical axis/line at  $q \times Q = 0$  so the key transition from attractive to repulsive Coulomb force can be seen.

Fig.7: State the droplet size in the caption.

Fig.7: Dark blue and black points are indistinguishable. I suggest you use different symbols for the three AP charges and then colour the points with a rainbow legend according to droplet charge.

Fig.7: Please add your theoretical curve to this figure from Dépée et al., 2019. Does it pass through your measurements? If not, please explain the discrepancies. Do you predict the small inflection in the CE as  $q \times Q$  goes from negative to positive?

Fig.7: Concerning the 3 points in the bottom right hand corner, are you capable of measuring CE at  $1E-4$  and below? Figure 10 would suggest not. One of the points disagrees with other points at higher CE values but the same  $q \times Q$ . The error bars on these 3 points look unrealistically small.

l.371-372. Please clean up this sentence.

l.374-379: You highlight the fact that these are the first experimental data to show the short-range attractive image charge forces but you do not provide any quantitative comparison in Fig.8 with your detailed model (Dépée et al., 2019). Please correct this.

Fig.8: Instead of the lines joining the points, please add curves showing the predictions from your model (Dépée et al., 2019) – including the uncertainties in the residual Coulomb force due to the  $0 \pm 600e$  charge on the droplets.

Fig.8: State the droplet size in the caption.

Fig.9: I suggest this figure (and Fig.10) is better plotted as Measured CE (y) versus Modelled CE (x). The model (if calculated properly) has no errors but the measurements do have errors and so they are better shown on the y axis. The CE data then appear above or below (or in agreement with) the theoretical prediction. The dashed line should be labelled “Measurement = Model”.

Fig.9: I don't understand what a “Parity plot” means. Better to label this “Measured versus modelled collections efficiencies according to Kraemer...”.

l.435: Replace “compared” with “comparable”.

l.453-470: This seems like a very long-winded paragraph that could be replaced by a brief sentence: “The lower detection limit on our experimental collection efficiencies is  $1E-4$ ” (or whatever is the correct number).

Fig.10: Please follow the same axis convention as Fig.9 (Measured CE (y) versus Modelled CE (x)).

Fig.10: Add a second curve to show the modelled collection efficiency without any charge effects so we can see the relative importance for the CE of charge compared with dynamic effects.

Fig.10: Please indicate in the caption what the error bars indicate. They are clearly underestimated and do not represent the full errors for each point. Please indicate the magnitude of the systematic errors either on a few representative points or else quoted in the caption.

Fig.10: State the experimental conditions – or their range – in the caption.

l.525: Replace “got” with “have”.

l.560: Replace “considering” with “that include”.

Fig.12: This is another figure that uses 3D images but would be far clearer and more useful if it were replaced by a simple line schematic. Please indicate precisely where on the new figure the image shown in Fig.13 is obtained.