

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 diffusiophoresis). 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.

Thank you for this general comment

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.

We did our best to enhance the English writing

- 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.

We totally agree. Indeed, both articles are strongly linked and the reader needs to read both articles to deeply understand all the experimental details. Nevertheless, we did our best to remind the main information required to understand each article. The few verbatim are useful to avoid the readers to pass from an article to the other. Note that we added the subsection with the quote so that the reader can easily look for the information in the other paper.

Besides, we previously wrote a single paper but it was too long and completely unclear. Indeed, we really felt it is necessary to separate paper. The main reason is that the method of CE measurement (and uncertainties) are completely different between Part I and Part II. Moreover, as it is frequently the case in the literature, we preferred to focus on the influence of only one parameter in each article:

1. Pranesha, Kamra, (1996, 1997a, 1997b).
2. Wang, Pruppacher, (1977) & Wang, *et al.* (1978).

Finally writing a single article would take around 50 pages.

Pranesha, T. S., & Kamra, A. K. (1996). Scavenging of aerosol particles by large water drops: 1. Neutral case. *Journal of Geophysical Research: Atmospheres*, 101(D18), 23373-23380.

Pranesha, T. S., & Kamra, A. K. (1997a). Scavenging of aerosol particles by large water drops: 3. Washout coefficients, half-lives, and rainfall depths. *Journal of Geophysical Research: Atmospheres*, 102(D20), 23947-23953.

Pranesha, T. S., & Kamra, A. K. (1997b). Scavenging of aerosol particles by large water drops: 2. The effect of electrical forces. *Journal of Geophysical Research: Atmospheres*, 102(D20), 23937-23946.

Wang, P. K., & Pruppacher, H. R. (1977). An experimental determination of the efficiency with which aerosol particles are collected by water drops in subsaturated air. *Journal of the Atmospheric Sciences*, 34(10), 1664-1669.

Wang, P. K., Grover, S. N., & Pruppacher, H. R. (1978). On the effect of electric charges on the scavenging of aerosol particles by clouds and small raindrops. *Journal of the Atmospheric Sciences*, 35(9), 1735-1743.

- 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.)

-

You are right, we updated the figures by adding the modelled CEs. See response for the specific comments.

Specific comments

1.27: Replace “the neutralisation” with “zero”. **We updated following your comment.**

1.30: Replace “correlation of Kraemer and Johnstone (1955)” with “prediction of Kraemer and Johnstone (1955)” (and elsewhere in the text). **We updated following your comment.**

1.34: Replace “on” by “on”.

The end of the abstract was updated following the comment of the other reviewer.

1.49 Replace “is mainly depending” with “mainly depends”.

We updated following your comment.

1.71-74: Please clean up this sentence.

The sentence becomes:

“For this purpose, the modelled CEs with electrostatic forces need to be experimentally validated before the incorporation in cloud models. Especially, the analytical expression for electrostatic forces used in numerical studies (Jaworek et al., 2002; Tinsley et al., 2006 ; Tinsley and Zhou, 2015 ; Dépée et al., 2019) has to be confirmed by measurements.”

1.114-115: Please clean up this sentence.

The sentence becomes:

“The first part of the paper describes the experimental setup. Afterwards, the method to evaluate the CE is detailed.”

1.121: Remove “a”.

We updated following your comment.

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).

(We did the same correction in Part I and Part II, see respond in Part I.)

We added trajectories outside the cross section of the droplet. Note that all trajectories are collected and we didn't show uncollected trajectories. Indeed, the CE is 27 for these parameters (Dépée et al. 2019) and if you neglect the Brownian motion here you have $CE=x_{max}^2$ where x_{max} is the max distance from the vertical droplet axis when the AP is collected due to the electrostatic forces (Dépée et al. 2019). So $x_{max}=5.2$ and we didn't want to plot until this value on the X or Y axis to see uncollected AP trajectories. We wanted the same scaling between the panel E and D.

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

OK, note that we added another sentence.

“Droplets fall at their terminal velocity (≈ 25 cm/s) into a chamber through an AP flow of 1.5 l/min. The flow velocity is 1.3 cm/s and the AP transfer time in the collision chamber is almost 80 s.”

1.144: Replace “go out” with “pass out of”.

We updated following your comment.

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⁻³)? 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?

We updated the Figure 2 according to your comment in the review of Part I. We also updated the Figure A.1.

Now for the AP penetration, you are completely right we didn't explain about that but we performed tests to evaluate the penetration. It was finally corrected in the AP number concentration. Note that the penetration is evaluated from the outlet to the AP charger to the outlet to the neutraliser (Figure 2 part II). We assumed the AP deposition was the same through the chamber even if it can be larger at the AP injection or in the AP/droplet separator.

In section 2.2 where we define the equation for the CE, we changed the end:

“

$$C_{m,AP} = \left(1 + \frac{1 - P_{InCASE,a,q}}{2}\right) \frac{m_{AP,tot}}{\Delta t \times Q_{In-CASE,c}} \quad (4)$$

The mean AP mass concentration is corrected considering the penetration ($P_{InCASE,a,q}$) in the collision chamber which depends on the AP radius (a) and charge (q). This parameter was estimated during *ex situ* experiments where the setup was the same as Figure 2, the only difference being a Condensation Particle Counter (CPC) set after the AP neutraliser and the AP charger to measure two AP number concentrations - 1 and 2, respectively. The penetration is then defined as concentration 1 over concentration 2. Thus, the measured penetration accounts for the AP deposition due to electrostatic forces on the wall of the collision chamber as well as in the pipes from the AP charger to the HEPA filter and the humidifier (Figure 2). The measured penetrations are presented in Table 3. It is observed the penetration decreases when the AP charges (q) increases and the AP radius (a) decreases since the electrical mobility of APs is larger. During experiments, the AP number concentration was ranged from $3 \cdot 10^4 \text{ cm}^{-3}$ (for $a=100 \text{ nm}$ and $q = -10 \pm 1 |e|$) to $2 \cdot 10^3 \text{ cm}^{-3}$ (for $a=250 \text{ nm}$ and $q = -90 \pm 9 |e|$). As a reminder, the pipes are anti-static and connected to the ground (as well as the collision chamber) so there is no charge accumulation due to AP deposition during experiments. Thus, the penetrations presented in Table 3 are assumed to be constant over time. Note that the AP deposition was neglected in Part I (Dépée et al., 2020) since the penetration was almost 100 % when the APs are neutralised.

Table 3 Measured penetration for the experimental conditions.

Dry AP radius (a)	AP charge (q)	Penetration ($P_{InCASE,a,q}$)
100 nm	$-10 \pm 1 e $	94.7 %
	$-20 \pm 2 e $	86.0 %
150 nm	$-11 \pm 1 e $	96.5 %
	$-30 \pm 3 e $	86.2 %
200 nm	$-10 \pm 1 e $	97.0 %
	$-34 \pm 3 e $	88.8 %

	$-71 \pm 7 e $	78.2 %
250 nm	$-22 \pm 2 e $	94.1 %
	$-52 \pm 5 e $	89.6 %
	$-90 \pm 9 e $	81.8 %

“

As you can see, here the CE calculation is not the same as Part I.

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?

The questions are addressed in previous answer. The penetration was evaluated from the outlet of the AP charger to the outlet of the AP neutraliser (Figure 2 part II). We assumed the AP deposition was the same during the transfer in the In-CASE chamber, pipes, etc. As a reminder, the pipes are anti-static and connected to the ground so there is no accumulation charge during experiments.

Note that, during the penetration tests, we didn't notice significant AP deposition in the anti-static pipes. So that, the APs mainly deposit in the collision chamber. It was also observed when we cleaned the chamber, there was more fluorescein on the wall when we measured CE with electric charges on APs than when the APs were neutralised.

l.175: Replace “the atmospheric one” with “one atmosphere”.
We updated following your comment.

l.181: Replace “So,” with “In this way”.
We updated following your comment.

l.182: Replace “was” with “were”.
We updated following your comment.

l.198: replace “get” with “are”.
We updated following your comment.

l.210: Replace “varying” with “varying”.
We updated following your comment.

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.

In this Figure, we updated the mid panel. We replaced it by a cross section of the housing made with a 3D printer. Note that we also updated the Figure 9 right of Part I.

We saved the other panels since it gives few points of view to understand the setup. Moreover, the injection head is presented here, the only time in this paper. We replace '3D printing' in the two papers by "housing made with a 3D printer".

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

We updated following your comment.

1.262: Replace "the double" with "doubly-".

We updated following your comment.

1.266: Replace "more" with "greater".

We updated following your comment.

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

This is the definition of the collection efficiency (the purpose of the article) so we feel it is essential to remind the definition.

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.

We initially wanted to write only one article and, in this section, we focus on the differences between the articles:

Following the comment of the other reviewer, we reminded the calculation of the uncertainties in this section – we do not just refer to the Part I section. In this new section, we emphasise the difference with Part I when another relative uncertainty is added in the calculation – the relative uncertainty of the AP penetration in the In-CASE collision chamber. Since it is neglected in Part I, it is another difference between the two papers. We also mentioned the potential AP pollution during the experimental protocol which can change our results and explain some difference model/measurements. Because this uncertainty is difficult to precisely quantify, we increase the low uncertainty for the CE measurements smaller than 10^{-4} .

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

We updated following your comment. We defined the variables in the two papers.

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.

In Figure 2 from Tinsley et al. (2000, see below) you can find the following figure (P parameter vs the normalised radial distance) where the P parameter is the addition of the short-range attractive term and the coulomb inverse square term in Equation (10) of Part II. As you can see, at low distance AP/droplet the sign is minus since the short-range attractive term dominate. At

large distance the coulomb inverse square term dominates since the sign is negative or positive following the charges have unlike ($K=Q/q < 0$) or like signs ($K=Q/q > 0$), respectively.

Nevertheless, it is not because it is always attractive at low distance following the electrostatic forces that the AP will be collected. Indeed, it is a balance between a lot of effects – Brownian motion, inertia, airflow around the droplet, drag forces, electrostatic effects, thermo and diffusiophoresis. Note that the dynamical effects as well as the Brownian motion do not only depend of the radial distance but also the orthoradial component. Thus, it would have been too complicated in one single figure to completely understand the different contributions for the experimental conditions. Moreover, we cannot plot the same curves as Tinsley et al. (2000) considering our experimental conditions since we have almost 70 values of the K parameter (through 10 AP charges and 7 droplet charges).

That's why, we did the choice to give the simplest explanation of the two terms in the electrostatic forces in introduction. As stated in this paragraph, a complete theoretical study of the different contributions in the AP collection can be found in Dépée et al. (2019).

Paragraph from the introduction:

“Finally, the analytical expression of the electrostatic forces is the addition of two Coulomb forces between the AP and the two-point charges inside the droplet. The factored expression can be found in equation (10) and further details can be found in Tinsley et al. (2000). It consists of two terms. The first one is the Coulomb inverse square term which prevails in the AP collection for large enough AP electrical mobilities or electric charge products ($q \times Q$), attractive (Figure 1, D) or repulsive (Figure 1, E) depending on whether the AP charge (q) and the droplet charge (Q) have unlike or like signs. The second term is referred to the short-range attractive term and dominates for weak electric charge products or for small AP electrical mobilities (Figure 1, F) and is always attractive (due to the charge distribution at the droplet surface with opposite sign to the AP charge (q)). A detailed study of their contribution can be found in Dépée et al. (2019).
“

Tinsley, B. A., Rohrbaugh, R. P., Hei, M., & Beard, K. V. (2000). Effects of image charges on the scavenging of aerosol particles by cloud droplets and on droplet charging and possible ice nucleation processes. *Journal of the atmospheric sciences*, 57(13), 2118-2134.

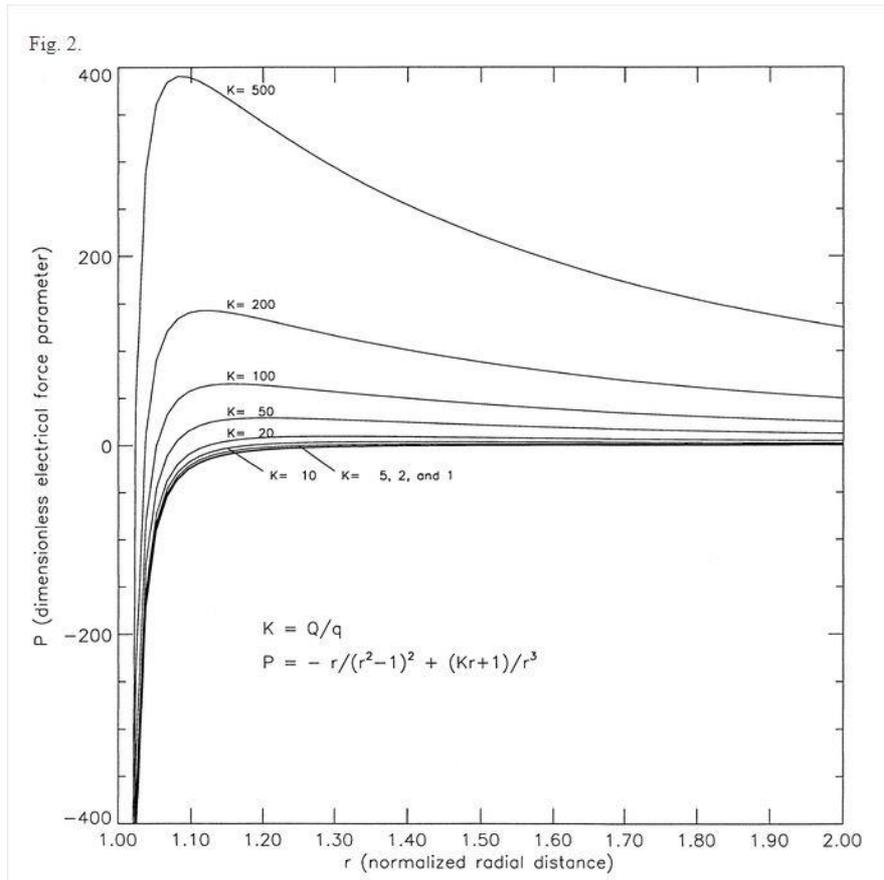


Figure 2 from Tinsley et al. (2000)

1.326: What is the meaning of the ambiguous word “global”? If it indicates “mean” then use “mean”.
We indicated “mean”.

1. 341: Replace “repulsing” with “repelling”.
We updated following your comment.

1.341: Replace “the fact that” with “whether the”.
We updated following your comment.

1.343-347: The short-range attractive force needs to be pointed out in Fig.7. I assume it is the small rise in CE at positive $q \times Q$?
We added the 4 curves for the 4 AP charges and the uncertainty range for the droplet and AP charges.

We updated the first paragraph of this section:

“The CE measurements for a wet AP radius of 432 nm are presented in Figure 7 as a function of the product of the droplet (Q) and AP (q) charges. The measurements are compared to the Dépée et al. (2019) extended model (solid line) for the 4 AP charges, considering the AP and droplet

charge uncertainties. There is a good agreement between model and measurements which indicates that the analytical expression of the electrostatic forces (equation (10)) reliably describes the observations.

Indeed, an important charge influence is measured [...]"

Yes you are completely right, the small rise in CE at positive charge product is due to the short-range attractive force as stated at the end of the section :

“For small positive charge products (approximately $0 \leq q \times Q \leq 10^6 |e| \times |e|$), an increase of CE with a factor of more than three is measured compared to the theoretical CE value without electrostatic forces. This fact truly emphasises the contribution of the short-range attractive term in equation (10) which attracts the APs toward the droplet even though the droplet and AP charges have like signs. Indeed, as previously stated, this term prevails for small charge products (Dépée et al., 2019).”

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.

We updated following your comment.

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

We updated following your comment.

As the paper Part I, we added a table 5 for the key features of the In-CASE set-up. We also mentioned the table in the caption to get the experimental conditions.

“measurement as a function of the product of the droplet (Q) and AP (q) charges for the wet AP radius of 432 nm and a droplet radius of $48.5 \pm 1.1 \mu\text{m}$. The experimental conditions are summarised in Table 5. Color code informs about the AP charge. The dashed line represents the theoretical CE value disregarding the electrostatic forces (given the air parameters 1°C , 1 atm, 95% of relative humidity). The solid line is the interpolation of the model (with the charge uncertainty range) for the respective CE measurements at a given AP charge.”

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.

We changed the blue color by the green color.

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?

We added the 4 curves for the 4 AP radii and the uncertainty range for the droplet and AP charges. As you can see, there is a good agreement between measurements and model and the small inflection that you describe appears in the modelling.

Fig.7: Concerning the 3 points in the bottom right hand corner, are you capable of measuring CE at $1\text{E-}4$ 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.

The In-CASE setup is capable of measuring CE at $1E-4$ and below. The only thing you have to do is to extend the duration of experiments. For these three points, the duration of experiment was almost 6 hours (with a concentration of about 10^4 part.cm⁻³).

In Figure 10 the 6 data point which are not suitable are probably due to AP pollution in the droplet impact cup during the fluorescein analysis (take off the cup, change the room with the cup to analyse, etc). The truth is, the AP pollution remains the larger error at low CE measurements but it is really difficult to account for this pollution in the error bar calculation. We were not able to quantify this random pollution in our setup. It can be due to many things, air quality in the room where we analysed the fluorescein, pollution of beakers, syringes, etc..

Even if it is often the case, this is not because the measured CE is lower that the corresponding errors is greater. Without AP pollution, the error is mainly due to the fluorescein analysis and the two diodes corresponding to two scaling and precision. Indeed, in the fluorescein analyser you have two diodes, one giving more precision at low detected concentration (= mass for a given volume control) which quickly saturate when the concentration increases; the second with low precision at low concentration but which saturates for larger concentration than the first one. When you are near the limit of a diode precision, here you have a large error (until 30 %). But sometimes you have a larger error with a great measured mass since you are not using the same diode. Thus, you cannot only consider the CE value to state that the error has to be great or low. It is all about detected mass, experiment duration and diode used.

As a reminder, we stated in 3.2.3.2 :

“Note that the detection limit of the spectrometer is 10^{-15} kg (for the nominal analysis volume considered), which only represents ten APs with a dry radius of 250 nm deposited on the droplet impaction cup. Thus, it exists an important uncertainty in these CE measurements related to a possible contamination. This is difficult to quantify but the low uncertainties of the CE measurements smaller than 10^{-4} were increased in Figure 10. To reduce this potential pollution, it would be necessary to work in a cleanroom or increase the experiment duration to avoid detection problem. However, for these data points the experiment duration was almost 6 hours (without mentioning the preparation, the purging and the cleaning durations) and, beyond this duration, stability problems of the piezoelectric droplet generator were frequent.”

1.371-372. Please clean up this sentence.

We update the sentence :

“However, at a given AP radius, an increase of the CE is observed when the number of elementary charges on the APs is larger. As a reminder, this increase appears even though the droplet is neutral (or very weakly charged considering the charge uncertainty of 600 elementary charges).”

1.374-379: 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.

See next response.

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.

We deleted the lines joining the points according to your comment. Afterward, we added the modelled CE for every CE measurement. We did not interpolate the modelled CE because it would have been too busy and we did not perform enough simulations to get reliable interpolations (and performing other simulations need few weeks to be statistically reliable as stated in Dépée et al. (2019)).

The vertical error bar for the modelled CE consider the AP and droplet charge uncertainties as mentioned in the new caption.

At the end of the 2nd paragraph (3.2.2) we added a sentence :
“Here, the good agreement between measured (circle) and modelled (triangle) CEs confirms that the analytical expression of the short-range attractive term in equation (10) is reliable”.

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

As the paper Part I, we added a table 5 for the key features of the In-CASE set-up. We mentioned the table in the caption to get the experimental conditions. The new caption is :

“

CE measurement (circle) as a function of the AP charge (q) for the 4 wet AP radii (Color code). The respective modelled CEs are also presented (triangle). The droplet is neutral with a radius of $48.5 \pm 1.1 \mu\text{m}$. The experimental conditions are summarised in Table 5. The dashed line represents the theoretical CE value disregarding the electrostatic forces (given the air parameters 1°C , 1 atm , 95% of relative humidity). The vertical error bars for the modelled CEs consider the AP and droplet charges uncertainties. “

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”.

Since the scaling is smaller on the Y-axis than the X-axis, it is better to see the measurement and the corresponding errors on the X-axis. If we switch on the Y-axis the error bar will be smaller.

In the figure there are errors for the modelling because, as explained in 3.2.3.1 and 3.2.3.2 :
“The horizontal error bars are the measurement uncertainties while the vertical ones are the extreme theoretical CE values considering the extreme droplet and AP charges (by adding or subtracting the charge uncertainties).”.

Indeed, the uncertainties for the droplet and AP charges does not appear in the CE calculation so that they do not appear in the errors of the measurements. Nevertheless, these uncertainties exist. At the end of the calculation, the measured CE can be the one of the droplet and AP charges predicted as well as the charges more or less the uncertainties. Thus, we modelled the CE for the experimental conditions as well as the min and max charges to get the max and min CE modelling which can correspond to the measured CE.

On the figure 9 and 10, note that we deleted the label of the droplet radius and we added a table for the experimental condition of these experiments (as you recommended on the other paper).

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

We deleted "Parity plot" in the text and the captions.

l.435: Replace "compared" with "comparable".

We updated following your comment.

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).

There is no clear limit of CE calculation in the present experiment. As far as the separation of droplet and aerosol is extremely efficient (100 % for the experiment duration considered in the study) the limit is not on the CE but on the mass collected in the impaction cup. It is possible to increase this mass by increasing the duration of the experiments. However as observed from figure 7, collection efficiencies seem to highlight vertical asymptotes, that would induced very long experiments for a low increased precision. For 6 data points, we suspect a post experiment contamination of the cup that induces overestimation of the collection efficiency). In present study we limited to 6 ours experiments. To represent this, we extended the lower limit of the uncertainties for the collection efficiencies bellow 10^{-4} . It would have been possible to increase accuracy with longer experiment (40 hours) or if the entire experimental setup were in a clean room in order to avoid this post experiment contamination.

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

As for Fig 10 we prefer this representation because it stretches more the experimental uncertainties.

Since the scaling is smaller on the Y-axis than the X-axis, it is better to see the measurement and the corresponding errors on the X-axis. If we switch on the Y-axis the error bar will be smaller.

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.

The aim of the Fig 7 and 8 is to observe the influence of the electric charge on the CE. So, you can find the theoretical curves without the electrostatic effects (and the modelled CE with electrostatic effects following your comment).

Fig 9 and 10 appears in the section "3.2.3 Comparison with existing models" where we only focus on the comparison of our measurements with the Kraemer and Johnstone prediction (1955) and our model. Thus, we did not add a curve with the modelled CE without electrostatic effects. Moreover, it is the 4 curves for the 4 AP radii (like in Figure 8) which have to be added vertically and horizontally and the figure would have been too crowded.

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.

You are right, we underestimated some point at very low CE ($\leq 10^{-4}$). As stated in 3.2.3.2, it is explained by the detected AP mass which is close to the detection limit of the fluorimeter. So, a post experiment pollution was probably involved. The problem is that this pollution is difficult to evaluate even if it can change a lot our measurements. Before your comment, we just omitted this uncertainty but now we did the choice to increase the low uncertainty of the CE measurements smaller than 10^{-4} .

This choice is explained at the end of the new section “2.3 Uncertainties”:

“As mentioned in 3.2.3.2, an AP pollution independent from the experiment (pollution during the spectrometry analysis, when the droplet impaction cup is extracted at the end of experiments, etc.) remains and should be considered in equation (5). Indeed, it can significantly increase the CE measurement, especially when the measured AP mass is close to the detection limit. It means, when the CE is below 10^{-4} for the experiment durations considered in the study. Instead of omitting this uncertainty which is difficult to evaluate, the low uncertainty for the CE measurements below 10^{-4} were increased until the end of the axis in Figure 7 and 10. “

Note that this choice is reminded in section 3.2.3.2 in the 2nd paragraph which states about a possible AP possible for the CE measurement below 10^{-4} :

“Note that the detection limit of the spectrometer is 10^{-15} kg (for the nominal analysis volume considered), which only represents ten APs with a dry radius of 250 nm deposited on the droplet impaction cup. Thus, it exists an important uncertainty in these CE measurements related to a possible contamination. This is difficult to quantify but the low uncertainties of the CE measurements below 10^{-4} were increased in Figure 10. To reduce this potential pollution, it would be necessary to work in a cleanroom or increase the experiment duration to avoid detection problem. However, for these data points the experiment duration was almost 6 hours (without mentioning the preparation, the purging and the cleaning durations) and, beyond this duration, stability problems of the piezoelectric droplet generator were frequent.”

For the vertical error bar for the modelled CE, it is explained in 3.2.3.1 and 3.2.3.2 :

“The horizontal error bars are the measurement uncertainties while the vertical ones are the extreme theoretical CE values considering the extreme droplet and AP charges (by adding or subtracting the charge uncertainties).”.

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

As Fig 8 and 9 we added the droplet radius following your previous comment. In the three captions we added “The experimental conditions are summarised in Table 5.” Where the Table 5 give the key features of the experiments. We added this table according to your comment for the part I paper.

1.525: Replace “got” with “have”. **We updated following your comment.**

1.560: Replace “considering” with “that include”. **We updated following your comment.**

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.

We didn't change this figure since we feel the left and mid panels offer different points of view and can be useful for people more comfortable with 3D view. The right panel of this figure looks like a cross section. So, we feel, everyone can understand this *ex situ* experiment by examining the panel of their choice.