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Reviewer #2 comments

Dépée et al., *"Laboratory study of the collection efficiency of submicron aerosol particles by cloud droplets. Part I - Influence of relative humidity"*.

-        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 II - Influence of electric charges"*.

-        The present paper presents a measurement of the collection efficiency of aerosol particles by sedimenting raindrops under various relative humidity (RH) conditions. The measurements are made in the so-called Greenfield gap region around 50-500 nm aerosol radii since both diffusion and inertial impact are minimized. In this size range the collection efficiencies are at a minimum (1E-3-1E-2) since aerosol particles barely deviate from streamlines that carry them around a falling droplet. Here the collection efficiencies are especially sensitive to thermal and diffusion effects that arise under reduced RH conditions due to droplet evaporation.

-        Aerosol wet scavenging is an important process for atmospheric aerosol and cloud microphysics. The experiments are very carefully executed and modelled. The precision of the measurements (shown in Fig. 9) is impressive and clearly demonstrate the influence of RH on collection efficiency. The measurements also agree well with their model predictions which, in turn, agree well with those of a previous model (Wang et al, 1978), albeit with the inclusion of additional physical processes. These measurements are probably the best experimental data that have been obtained so far on this mechanism for aerosol scavenging. They experimentally confirm the validity of the models, whose predictions now have increased confidence. In summary, I have no hesitation to recommend that the paper be published in ACP, after responding to the comments below.

Thank you for your general comment

General comments

-        My main recommendation is that the paper needs significant work to improve its flow and clarity before publication. Sufficient information is contained in the paper but it is hard to find and the reader is forced to go forwards and backwards many times to find critical details that appear in the wrong place. Several figures also need improvements.

For a deep understanding of this work, it is true you need time to read carefully every section and sometimes go forwards and backwards. It comes from the fact that we wanted to explain everything and sometimes, we had to make links/quotes between subsections. Indeed, it is not only true for this paper but for both papers. If you want to understand everything you need to read both papers since they come from a same study. At the beginning we wrote one single paper of 50 pages but the reader would have been lost between the different set up and method to compute the CE. Thus, we separated into two parts and to prevent part II from doing plagiarism or repeating part I, we added some quotes « More details can be found in Dépée et al. (2020).. ».

-        There appear to be many examples of a literal translation from French text into English, which leads to incorrect grammar. For example, in the Fig. 1 caption there is the following sentence:

We did our best to correct this impression

"From Figure 1 A to F, the considered effects are the Brownian motion (A), the inertial impaction

(B), the interception (C), the diffusiophoresis (D), the coupling of thermophoresis and diffusiophoresis (E) and the thermophoresis (F) are highlighted."

It should read:
"The panels indicate the effects of Brownian motion (A), inertial impact (B), interception (C), diffusiophoresis (D), combined thermophoresis and diffusiophoresis (E), and thermophoresis (F)."

We took this remark into account.

English frequently does include the definite article "the" before nouns, in contrast with French. This mistake occurs throughout the manuscript. I recommend that the authors ask a native English speaker to edit the manuscript.

Specific comments

l.45-47: This sentence should be removed; human (non-)survival does not depend on atmospheric caesium-137.
We replaced the word « survival » with « health ».

l.57-59: The collection efficiency needs to be defined here - not just 10 pages later – since it is key for all the discussion in the introduction.

We added the definition at the end of the paragraph:
« It is the ratio between the AP number (or mass) collected by the droplet over the AP number (or mass) within the volume swept by the droplet, for a given AP radius. Another equivalent definition is the ratio of the cross-sectional area inside which the AP trajectories are collected by the droplet over the cross-sectional area of the droplet. ».

l.66: Replace "phenomenon called" with "known as".
This remark is taken into account.

Fig.1: Use a consistent colour scheme for all panels: red = impact, black = no impact, blue = no impact (second AP). This is an important figure but it has to be stared at for a long time before the many numbers are interpreted and understood. Try to simplify the labelling in the panels to show the key information (the different processes and the AP in each trajectory) and move non-key details to the caption.

We changed the colour code as you suggested in Figure 1 and we updated the caption:

« APs trajectories computed with the extended Dépée et al. (2019) model for a 50 µm droplet radius (A) and AP with various radii (a) and densities (ρ_{AP}). The air temperature (T_{air}) and the air pressure (P_{air}) are respectively -17°C and 540 hPa. The panels indicate the effects of Brownian motion (A), inertial impact (B), interception (C), diffusiophoresis (D), combined thermophoresis and diffusiophoresis (E), and thermophoresis (F). Red trajectories result in an AP collection. In Figure D and F, the gradients are equivalent to a relative humidity of 0.01 % (when there is no gradient the equivalent relative humidity is 100 %). $\rho_{v,s}$ and $\rho_{v,air}$ are the vapor densities at the droplet surface and in the bulk air, respectively. »

We know there are some parameters in this figure and it can be confusing. However, we believe letting parameters available can be helpful for readers who want to deeply understand the modelling and reproduce the same simulations. Note that in the theoretical paper Dépée et al.

(2019), there is no simulation like this. Thus it is new and helpful to understand the work and the true physical effects involved in the AP collection.

Note that we also changed this figure and caption in part II.

l.144: "component" not "part". **We guess your comment was « part not component » so we updated.**

Fig. 2: This is a key figure but it is extremely poor. I suggest you re-draw it to show the components in schematic box form and not a mixture of 3D images and over-detailed objects. It is completely unclear which way the AP and air flow proceeds, whether gas is entering at the top or bottom (it is both), etc. The gas lines are too thin and the gas directions should be made very clear, and their purpose indicated. Even the piezoelectric droplet generator is unlabelled… Why is all the unimportant structure around the drift tube included in the figure? I had to go forward several pages and then come back to this figure + text to understand the figure. Fig. 1 should be understandable on its own without having to do this. The authors of course know what this figure means; but the reader does not.

We re-draw this figure. We simplified everything and add colour code for the different key parts of In-CASE. We deleted every images or objects and we added flowrates. The new caption is : "Figure 2 In-CASE setup to study the influence of relative humidity. Colours represent different functions. Red – upward argon flow against AP pollution in the droplet impaction cup. Purple – AP (and Argon) evacuation toward the HEPA filter. Orange – AP, generation, selection and neutralisation. Black – surplus evacuation and DMA flowrate control. Brown – droplet radius measurement. All the key features of the setup are detailed in Table 2. "

Note that we didn't explain the injection head and the AP/droplet separator since they are little simplified in this figure. We ask to the reader to go few pages later for more details. We know it can be boring to go forward and backward but we chose to provide details on the validation of each component of the experiment. This is done step by step.

Nevertheless, we changed the plan of the section in order to have the description of the injection head, the collision chamber and the AP/droplet separator just after the Figure 2. So, the reader does not have to go a lot of pages later to have a complete description of the In-CASE set-up.

Previous plan :
1.1   Overview
1.2   AP generation
1.3   Droplet characterisation
1.4   In-CASE chamber
1.4.1   Injection head
1.4.2   Collision chamber
1.4.3   In-CASE's bottom stage

New plan :
1.1   Overview
1.2   In-CASE chamber
1.2.1   Injection head
1.2.2   In-CASE's bottom stage
1.2.3   Collision chamber
1.3   AP generation
1.4   Droplet characterisation

Fig. 2: I suggest you add a table summarizing the key features of the apparatus (dimensions, flow rates, residence times for droplets, residence times for Aps, temperature, RHs, etc.). Most of these are in the text but they are scattered in several places.

This comment is taken into account. We replaced the table 2 (which has been deleted following your comment) by a table with key features of the In-CASE set-up.

Section 1.3.2: You state the droplet charge is 0±600e after the inductor plate neutralizer. How stable is the droplet charge neutralisation with the change of geometry (setup *ex situ* to operation *in situ*)? A small change in geometry or surface charging of an insulator could affect the electric field at the tip of the piezoelectric droplet generator and, in turn, the droplet charge. If I look at your Fig. 6 in Paper II, the droplet charge is about 3000e per 1 V on the inductor plate, so you are sensitive to nearby stray potentials of only a few 100 mV.

This comment is completely true. Different geometries were investigated in order to ensure that the charging law is stable over time, even after disassembling/cleaning/reassembly cycles. To this aim a monobloc unit was 3D printed, in order to fit, by construction, the Electrostatic inductor and the piezoelectric injector.
Moreover, the inductor plates are made of stainless steel, and are periodically cleaned with a precise protocol to keep the surface of the plate clean and free of rust.
Finally a stabilised power supply, with a precision of 1×10-2 V, is used to minimise the uncertainty on the electric field produced by the inductor.

The droplet charger is stable since we verified through few experiments if we get the same charging law at the end. Note that during all experiments, the plate was taken off from the 3D printing, cleaned, stocked carefully. Moreover, the tests was not performed during the same day, sometimes few weeks later! The same protocol was performed for these tests than during CE campaigns. The housing made with a 3D printer (Figure 9, right) was the same between *in situ* and *ex situ* experiment. In Fig 6, in Paper II, the four different colours are for 4 different experimental tests performed at different days, after different disassembling/ cleaning/reassembly cycles.

In Fig 6 paper II, we wrote the resulting correlation from the 4 tests but actually the correlations for the separated tests are equal. Indeed, the discrepancy of the linear coefficients ($Q=-3.2\times10^{3}$x U-8.4$\times10^{3}$) was less 1.5%. So, the « stability of the droplet charger » in the uncertainty of the droplet charge is neglected compared to the method of droplet charge evaluation detailed in Appendix B part II.
Besides, the correlation $Q=-3.2\times10^{3}$x U-8.4$\times10^{3}$ (Fig 6, part II) is gathering about 70 measurements performed over 4 campaigns of experiments, with a disassembling/cleaning/reassembly cycle between each of them. The high coefficient of determination 0.999 shows the high reproducibility.

Fig. 6: 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. How do you ensure good radial mixing of the AP – which is an important assumption in your determination of the CE? What is the flow rate and velocity of the air down the tube (I did not find this in the text; it may be there but this is another example of information that the reader should be able to find in a table or other easily identified place).
As referred in the section 1.2.1 :
« The APs are inserted from the sides of the entire circumference through a flat torus inlet. This injection principle is based on the CLINCH experiment (CoLision Ice Nucleation Chamber, Ladino et al., 2011) which ensures a laminar flow and a great spatial APs mixture in the collision chamber inlet. »

Considering the geometry of the aerosol injection in the collision chamber, the initial radial velocity at the injection the chamber is between 2 cm/s. It induces a vertical displacement for the aerosol jet to reach the centre of the collision chamber of about 4 cm. These 4 centimetres are considered to be the uncertainty of the effective height of interaction between droplets and APs ($u_{H_{eff}}$) which is finally 4 %.
We corrected this value from previous version of the article

l.302: The minimum RH is only controlled by the efficiency of the dryer to remove water from laboratory air. How stable is the minimum RH when the lab RH varies and perhaps also the efficiency of the dryer?

We didn't see a RH variation related to a saturation of the dryer. Indeed, 2 diffusion dryers were set in series (TSI 3062 dimensioned to dry a maximum flow of 4 L/min). The maximum flow we investigated is 1.5 L/min (part II) which is far less than the maximum flow rate provided by constructor. Moreover at the end of each experiment, each of them was regenerated in a proofer for at least 8 hours.
So during 1 experiment the two dryers were more than enough. Concerning the lowest RH considered during experiment, we observed after 5 hours of experiment an increase of 1% maximum of the relative humidity. This increase is attributed to the droplet evaporation (even weak) during the experiments which last few hours.

Finally before every experiment, especially for the lowest RH, we spent at least 15 min to purge the collision chamber by passing through it a dry air at high flow rate (more than 10 L/min).

Section 1.4.2.2: Include some numbers here: terminal velocity, transit time in tube.

we added in brackets this two parameters :

« The corresponding terminal velocity ($U_{A,\infty} \approx$ 25 cm/s) is computed from Beard (1976). The residence time of the droplet in the chamber (≈4 s) is computed considering these two changes. »

l.313: Replace "vaporization" with "evaporation".
This remark is taken into account.

l.344: Replace "fittings" with "fits".
This remark is taken into account.

l.367: You state the AP terminal velocity is about 1E-2 cm/s. For your largest AP (160 nm diameter), I estimate the terminal velocity is about 1E-4 cm/s.

You are right, we did a mistake.
The larger wet AP is 393 nm for 93% of RH. The AP density is 1150 km/m3 (as referred in table 2) taking into account the hygroscopicity of AP. With the air density rho_a= 1.2906 kg/m3 and the dynamical viscosity of na=1.7388e-05 kg/m/s, the AP terminal velocity is :

2*(393*10^-9)^2*9.81*(1150-rho_a)/(9*na)= 2.2244e-05 m/s ≈10-3 cm/s.

So, we changed « and the AP terminal velocity is about 10-4 cm/s » by « and the AP terminal velocity is less than 10-3 cm/s ».

Fig.9: Remove the colouring so the details of this figure can be seen (as in the upper part of the figure).
Figure is redrawn without colours

l.379: Replace "usual" with "standard".
This remark is taken into account

Section 1.4.3.2: You ran fluorescein (the AP detection material) through the droplet generation system for this test. Did you confirm that there was no residual contamination of fluorescein in the droplet generation system before doing the actual CE experiments? Even a very small contamination could interfere with your measurements.

Yes, the piezoelectric generator was cleaned several time with water to be sure no pollution remains on it and we tested the water by fluorescein analysis. For the droplet generator, we turn it on during few hours with pure water and high pressure (to get a water jet) to clean it. At the end, we tested the water several times at the outlet of the piezoelectric generator by fluorescence spectroscopy. All of the measurement were below the limit of detection of our system. It was thus considered free from any fluorescein contamination.

l.421: C_m,AP is first defined here, ½ page after the equation where it first appears. Please define new variable immediately – at least descriptively – immediately after the equation where it first appears.

Ok we reminded the CmAP definition right after it first appears.

l.439: Replace "the ones (single charged)" with "those with single charge,".
This is done

l.442: Remove "into".
This remark is taken into account

Section 3.1: The droplet charge is 0±600, so all droplets are, in fact charged with several 100 e of either sign, on average. The AP have a Boltzmann distribution so roughly 50% have ±1e and 13% have ±2e charge.

You state that this has a negligible influence on the CE but it is a sufficiently important effect that you should expand this discussion with a more quantitative justification that charges can be neglected.

we added details :

"In all experiments, the droplet charge is 0 ± 600 elementary charges with a radius of about 50 μm. Since the AP charge distribution is similar to a Boltzmann distribution, an AP charge of more than 5 elementary charges is thus highly unlikely. Consequently, it is assumed that the contribution of the electrostatic forces on the CE is of second order and these effects were then neglected. Indeed, Dépée et al. (2019) numerically evaluated the contribution of the electrostatic forces on the CE for a droplet of 50 μm radius with 1000 elementary charges and 5 elementary charges on the AP. For these extreme values, they showed that the electrostatic forces increase

Becomes

Supplementary Materials from Dépée et al. 2019

| AP radius	Droplet charge/ AP charge						Droplet charge/ AP charge
	-100000/5	-10000/5	-1000/5	-100/5	0/5	100/5	0/0
0.004	2.22E+1	2.37E+0	2.97E-1	1.66E-1	1.47E-1	1.36E-1	1.32E-1
0.005	1.45E+1	1.53E+0	2.04E-1	1.16E-1	1.09E-1	1.01E-1	9.60E-2
0.008	5.89E+0	6.07E-1	9.26E-2	6.03E-2	5.70E-2	5.43E-2	5.00E-2
0.01	3.85E+0	3.96E-1	6.50E-2	4.45E-2	4.25E-2	4.10E-2	3.70E-2
0.03	4.71E-1	4.74E-2	1.37E-2	1.13E-2	1.12E-2	1.08E-2	9.46E-3
0.05	1.84E-1	1.93E-2	7.37E-3	6.46E-3	6.35E-3	6.29E-3	5.19E-3
0.08	7.82E-2	9.48E-3	4.46E-3	4.04E-3	4.03E-3	3.98E-3	3.22E-3
0.1	5.38E-2	7.05E-3	3.59E-3	3.33E-3	3.32E-3	3.31E-3	2.64E-3
0.3	1.07E-2	2.29E-3	1.63E-3	1.58E-3	1.57E-3	1.56E-3	1.34E-3
0.5	6.02E-3	1.61E-3	1.27E-3	1.24E-3	1.23E-3	1.22E-3	1.16E-3
0.8	3.59E-3	1.24E-3	1.07E-3	1.06E-3	1.05E-3	1.04E-3	1.02E-3
1	2.72E-3	1.09E-3	9.88E-4	9.85E-4	9.81E-4	9.77E-4	9.68E-4
1.3	1.85E-3	1.03E-3	9.59E-4	9.58E-4	9.57E-4	9.56E-4	9.55E-4
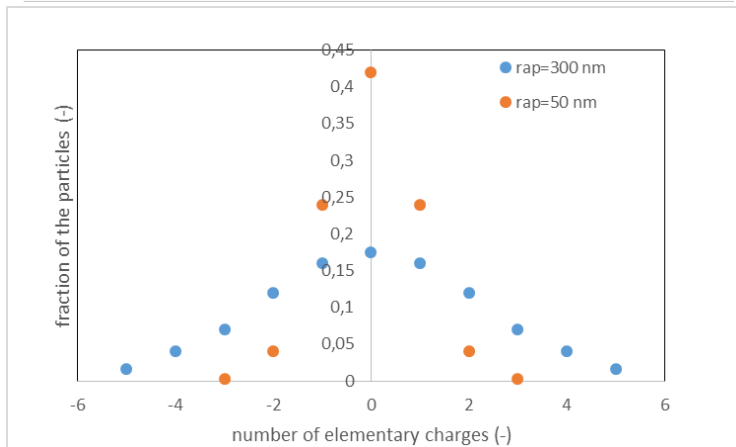
Fig. 9: These are the main results of the paper but they could be presented much better. It is confusing to make the reader look at a table (not a legend) to understand the explanation of the multiple curves in the plot. I suggest that you remove the dashed and dotted curves and leave only the solid curve giving the central prediction of the model. The remaining coloured bands then indicate the range of predictions of the model corresponding to the extreme experimental conditions. That will reduce the number of curves by a factor 3. I also suggest that you show only the top panel: your data and your preductions. Then, in asepearte figure, show the comparison of just the central predictions from your

model and from Wang et al. That way: a) Fig. 9 focuses on your measurements + model and b) the following figure focuses on the difference of your model with Wang et al.

We changed the figure :
- **There is only one panel now,**
- **We deleted the extreme experimental conditions and the previous table 2,**
- **We only kept the solid lines corresponding to the mean conditions for the three levels of relative humidity,**
- **Since we deleted the bottom panel but we compare our model and the one of Wang et al. (1978), we added dashed line corresponding to the Wang models in the same panel.**

We feel like this new figure is clearer and adding the Wang models in the same panel of our model is better for the comparaison of both model <u>with</u> measurements.

We also changed the caption:
« CE measurements for three levels of relative humidity - 71.1, 82.4 and 93.5 % - compared to the extended model of Dépée et al. (2019) (solid lines) and the Wang et al. (1978) model (dashed line). Squares are the CE measurements summarised in Table 3. For the modelling, air temperature and dropet radius are then the mean values of the three levels of relative humidity - T_air=0.26, 0.27 and 1.2°C - A=49.3, 50.8 and 48.8 μm - from the lowest to the highest, respectively. For RH= 100 %, the parameters are those from table 2. ».

We also changed the first paragraph of the subsection 3.2 :
« […] The key features of the experiments are summarised in Table 2. The measurements are compared to computed efficiencies using the models described in Wang et al. (1978) (dashed lines) as well as the extended version of Dépée et al. (2019) (solid lines). Note that the experimental conditions vary a little for the CE measurements at a given relative humidity level. For the modelling, air temperature and dropet radius are then the mean values of the three levels of relative humidity - T_air=0.26, 0.27 and 1.2°C - A=49.3, 50.8 and 48.8 μm - from the lowest to the highest, respectively. For RH= 100 %, the parameters are those from table 2. »

Fig. 9: Please add a table here that summarises the droplet and AP experimental conditions (droplet radius, AP wet radii, RH, etc.) rather than, for example having "A = 49.6±1.3 μm" in a huge bold font in the panel. This table would be helpful at many stages during the reading of this manuscript. The table should include the AP number concentration in the chamber (I did not find this anywhere in the text; it is frustrating for the reader to spend 5 minutes searching unsuccessfully for an important experimental condition). Also include in this table the distance between successive droplets falling through the chamber.

As previously mentioned, we replaced the table 2 (which has been deleted following your comment) by a table of key features.

Fig. 10: Replace "EC" by "CE" in the x axis label.
It was a typo, thank you