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.

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.

- 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:

"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)."

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.

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.

l.66: Replace “phenomenon called” with “known as”.

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.

l.144: “component” not “part”.

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.

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.

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.

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 indentified place).

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?

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

l.313: Replace “vaporization” with “evaporation”.

l.344: Replace “fittings” with “fits”.

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.

Fig.9: Remove the colouring so the details of this figure can be seen (as in the upper part of the figure).

l.379: Replace “usual” with “standard”.

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.

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.

l.439: Replace “the ones (single charged)” with “those with single charge,”.

l.442: Remove “into”.

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.

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.

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.

Fig. 10: Replace “EC” by “CE” in the x axis label.