Dear reviewer,
Please find the point by point answer to your questions
To facilitate the discussion I adopted a colour code:

- In red are the questions
- Blue are the answers
- Green are the modifications or additions in the article

Thank you for taking your time for the review
Kind regards

Major comments

Droplets generated by means of piezo-elements might be charged, giving rise to electro-scavenging forces which can affect the collection rate: see, for example, Pranesha et al. (1966) and Ardon- Dryer et al. (2015). In addition, the aerosol neutralizer leads to a Boltzmann distribution where the most common charge state other than neutral is a single charge. Therefore electro-scavenging forces, due to spurious charges, cannot be ruled out unless drop charges are measured. In addition, the droplets are falling in a subsaturated environment (77% RH) and could therefore evaporates giving rise to phoretic forces.

The authors should clarify these important aspects in order to highlight their results.

In order to better introduce the various mechanisms involved in the collection of the aerosol particles by drop (and droplets) we added in the first section after the introduction (Theoretical description of washout) a paragraph describing all the mechanisms involved in the collection of the aerosol particles by raindrops. (p4, line 1-30).

“Several mechanisms are usually considered to simulate the collision between aerosol particles and droplets. We recall them briefly, however, a more exhaustive review can be found in the literature (Pruppacher et al., 1998; Chate, 2005; Ladino et al., 2013; Ardon-Dryer et al., 2015). The three main mechanisms leading to this collection are Brownian motion, inertial impaction and interception. Small particles with a radius on the order of the mean free path or smaller are very sensitive to the collision of air molecules and scatter from streamlines of the flow due to Brownian motion. For large particles with a diameter greater than 1 µm, their inertia prevents them from following the streamlines of the flow and they impact the drop on its leading edge. Aerosol particles with a diameter smaller than 1 µm and much larger than the mean free path of the air molecules follow the streamlines of the flow around the drop. They might nevertheless enter in contact with the drop because the streamlines are approaching the drop at a distance smaller than the radius of the aerosol particle. For particles with diameter between 0.2 µm and 1 µm, there is a minimum collection efficiency called the “Greenfield Gap” (Greenfield, 1957). For these particles phoretic forces are expected to be the most efficient mechanisms. Thermophoresis and diffusiophoresis are respectively linked to thermal and water vapour gradients. The side of the aerosol particles exposed to the warmer air is impacted by molecules whose kinetic energy is higher than that of the molecules impacting the colder side of the particles. As a result, thermophoresis results in a force whose direction is the opposite of the thermal gradient. Similarly, particles exposed to a water vapour gradient are exposed to molecular collisions with a dissymmetric kinetic energy since water vapour molecules are lighter than air molecules. In the atmosphere, diffusiophoresis thus results in a force whose direction is the opposite of the water vapour gradient. Electro-scavenging could also have an important contribution when both droplets and aerosols particles are electrically charged, resulting in an attractive (or repulsive) force when they have opposite (or the same) polarity. Moreover, Tinsley et al. (2000, 2006) theoretically showed that electrically charged aerosol particles can induce an image charge on droplets that results in a short range electrical attraction that increases collection efficiency even with neutrally charged droplets.

For each of these elementary mechanisms, theoretical expressions of the elementary collection efficiencies have been derived (Table 1).

Table 1. References of theoretical expressions for the calculation of each collection mechanism

Elementary mechanism	Reference
Inertial impaction	Slinn (1977); Park et al. (2005)
Interception	Slinn (1977); Park et al. (2005)
Brownian motion	Slinn (1977); Park et al. (2005)
Diffusiophoresis	Waldmann (1959); Davenport and Peters (1978); Andronache et al. (2006); Wang et al. (2010)
Thermophoresis	Davenport and Peters (1978); Andronache et al. (2006); Wang et al. (2010)
Electro-scavenging	Davenport and Peters (1978); Andronache et al. (2006); Wang et al. (2010)
Image forces	Tinsley and Zhou (2015)

Finally, the droplet total collection efficiency can be theoretically deduced by adding all these elementary collection efficiencies together. The use of these theoretical models seems justified for cloud droplets since they have very small Reynolds numbers. However, there are many uncertainties concerning raindrop size range."

Once all the mechanisms described, we explain that we want to compare to Beard (1974) model, and thus to minimize all the mechanisms he did not considered in his simulations. (p7 line 2-6).

"The objective of these modifications is also to be consistent with the hypothesis of the Beard (1974) model, which considers only drag and gravitational forces on the aerosol particles. The modifications are thus intended to minimise electro-scavenging (discussed in sections 2.1 and 2.3), diffusiophoresis (discussed in section 2.3 and Appendix 1) and thermophoresis. Both the drop generator and aerosol chamber are described in the following sections. "

-Concerning electro-scavenging:

We minimized electo-scavenging by checking that the drop are not electrically charged. The drop charges are measured with the help of a Faraday pail connected to an electrometer (Keithley model 6514; Sow & Lemaitre, 2016). Any electrical charge on the drop could not be measured even with the high sensitivity of the electrometer (10 fC) and even integrating the measured charge on a large number of drops. It should be noted that this generator is completely different than microdrop Technologies and MicroFab Technologies), first the piezo element is not in contact with the fluid. Second the all hydraulic system is grounded.

We add the following paragraph in section 2.1 (P7 line 23 to p8 line 2).

“Classical piezoelectric drop-on-demand systems may produce electrically charged droplets (Ardon-Dryer et al., 2015). However, we want to limit electro-scavenging as Beard (1974) did in his simulations.

Thus, the net charge of each drop produced by this system has been measured with the help of a Faraday pail connected to an electrometer (Keithley model 6514; Sow & Lemaitre, 2016). Any electrical charge on the drop could not be measured even with the high sensitivity of the electrometer (10 fC). This might be explained by the fact that unlike classical piezoelectric drop-on-demand systems (such as those of microdrop Technologies and MicroFab Technologies), the piezoelectric transducer in our drop generator is not in direct contact with the liquid (Figure 2).”

Finally the aerosols particles are neutralized. Thus they have a Boltzmann charge distribution.

-Concerning diffusiophoresis:

To be sure that during our experiment (with 77 % relative humidity) diffusiophoresis is a second order mechanism, we compared the elementary collection efficiency it induces compared to beard (1974) model. Diffusiophoresis was calculated with the model of Davenport & Paters (1978). This comparison is presented in figure 12.

We added the following tex in section 2.3(p13, line 34):

Furthermore, at this high relative humidity, diffusiophoresis is not expected to contribute significantly to the collection efficiency, even close to the minimum of efficiency. Indeed, the contribution of diffusiophoresis calculated with the model of Davenport and Peters (1978) for our experimental conditions (relative humidity, air temperature and drop size) is 2.5 x 10^{-4}, which is smaller than the collection efficiencies predicted by the Beard model (Figure 1).

Line 73. " *The Slinn model (1977) does not reproduce this increase efficiency, leading to errors of several order of magnitude*...". Lai et al (1978) measured scavenging of aerosol particles by falling water droplets. They compared their results against Beard and Slinn models. Concerning the Beard model the authors stated that "... *his estimation of the collection efficiency is more than one order of magnitude lower than our experimental results*..." and concerning Slinn model *"...hardly predicts the qualitative features of our results, it intercepts the range of values obtained in this work with submillimeter droplets* ...". Therefore Lai et al. (1978) concluded that the Slinn model at least intercepts their experimental data. The authors should explain this different conclusion.

This point is discussed together with next question. Nevertheless on point is still strange for us, because when we compare Beard (1974) and Slinn (1977) model, we more or less, find the same results for aerosol particles with stokes number greater than 0.05, however Lai find some differences. May be Lai (1978) added diffusiophoresis to their calculations as it seems that their experiments are dominated by diffusiophoresis. However we don’t have many details on that point on the publication from Lai *et al*. (1977).

Line 164 *"...the Slinn model underestimates by two order of magnitude the measured collection efficiency for submicron-sized particles*". The figure below shows, among others, the collection efficiency from Lai et al (1978), present manuscript, Beard (1974) and Slinn (1971), Quérel et al., (2014). The collection efficiencies are given as particle aerodynamic diameter. It can be seen that the experimental points from the present manuscript are in agreement with Beard (for D < 1 micron); however the collection efficiency from from Slinn model (taken from Fig. 6 of Lai et al. paper) is in better agreement with Lai and not with the manuscript authors (see previous comment). Figure 1The experimental points from Lai et al., were obtained with AgCl with a particle density of 5.6 gcm-3, larger than the particle density used in the manuscript (1.3 g cm-3). Droplet rear particle capture is mainly due to inertial effects (Brownian diffusion is too weak in this particle size range) therefore, in order to compare results from different papers, the collection efficiency should be compared against aerodynamic particle diameter or even better against the Stokes number.

We explored different hypothesis to explain the difference between Lai (1978) measurements and ours.

- First it seems that the particle they use have same geometric standard deviation as ours, thus the difference could not come from this point ("*For a particular aerosol the particular size distribution typically had a variance of about 10 % from the modal value*").
- Second, it seems that the particles they use are not neutralised by any system. However it seems that they are more or less neutralised. Indeed when they plot the influence of the drop charge on the collection efficiency (Figure 7 from their article) their measurements are symmetric with the ordinate axis.
- It seems that their drop are not exactly at terminal velocity. Indeed they used a 455 cm shaft to accelerate their drop however it seems from Wang, & Pruppacher, (1977b) that 6.5 m are needed to reach 99% of the terminal velocity. (we have 8m). but we think this is not the point that explain of the difference between Lai results and ours.
- The most convincing reason explaining this difference is the relative importance of diffusiophoresis. Indeed Lai performed their experiments in pure nitrogen. Thus their experiments seem to be driven by diffusiophoresis.

To compare our measurements with Lai's ones we need to plot them as function of the Stokes number as advised by the reviewer. Then we compared Beard (1974) and Slinn (1977) models together with present and Lai's results (figure 12). The analysis of Lai's experimental procedure shows that their aerosol chamber is filled with pure Nitrogen. Thus we had on the figure the contribution of diffusiophoresis calculated from the model of Davenport & peters (1978) in our and Lai experiment. This highlights that Lai experiments are dominated by diffusuiphoresis. This is the reason why they doesn't fit beard results.

We added : (p 20 l 15 - p 21 l 23)

"Furthermore, it is interesting to compare our measurements with the ones from Lai *et al.* (1978) since they are the only ones in the literature in the same drop size range. As the aerosol particles produced in these experiments are composed of silver chloride ($\rho_{AgCl} = 5.6\ g.cm^{-3}$), which is much denser than sodium fluorescein ($\rho_{C_{10}H_{10}Na_2O_5} = 1.3\ g.cm^{-3}$), it is more appropriate to plot all the collection efficiencies as a function of the Stokes number of the particle (St_{ap}).

$$St_{ap} = \frac{\rho_p U_\infty(D_{drop}) d_{ap}^2 C_{c,d_{ap}}}{9 D_{drop} \mu_{air}}$$

In this equation, μ_{air} is the dynamic viscosity of the air and ρ_p the density of the aerosol particles. This comparison is presented on Figure 12.

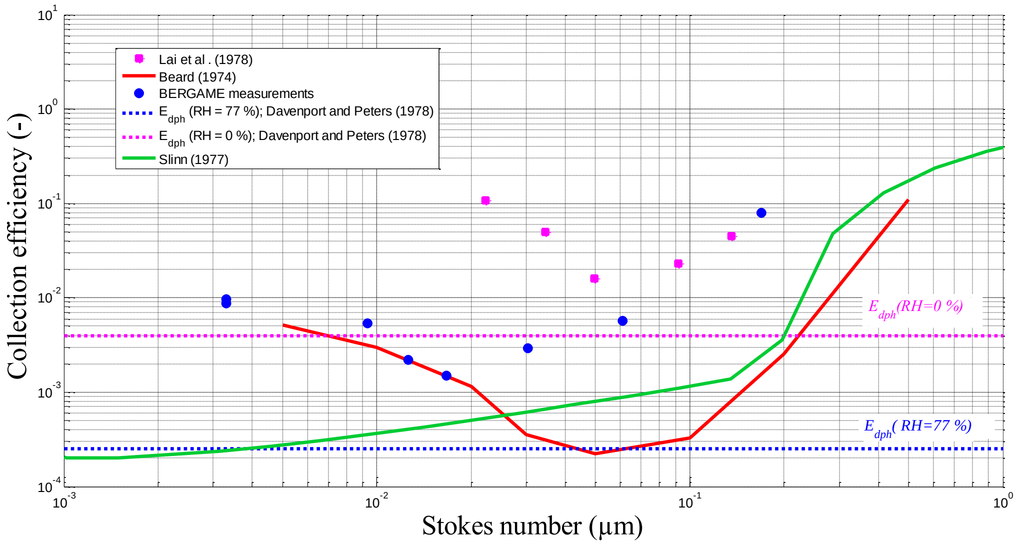

Figure 1. Comparison of our measurements with Lai *et al.* (1978). Both measurements are compared with the Slinn (1977) and Beard (1974) models. The contributions of diffusiophoresis are evaluated in both experiments with the model of Davenport and Davis (1978)

For particles with a Stokes number greater than 6×10^{-2}, the motion of the particles is driven by their inertia, leading us to expect to observe the same trends in our measurement and those of Lai *et al.* (1978). The comparison for Stokes number smaller than 6×10^{-2} is much less obvious. Indeed, for these particles, the measurements of Lai *et al.* (1978) indicate an increase in collection efficiency, while our measurements continue to decrease down to a Stokes number of 1.6×10^{-2}. At that point, the slopes of the increases of both collection efficiency measurements are similar, while the Stokes number decreases.

A precise analysis of the procedure for the aerosol particle injection in the experiments of Lai *et al.* (1978) indicates that the carrier gas is pure nitrogen without any subsequent humidification. As a consequence, it is reasonable to consider that their measurements were performed with 0% relative humidity. In order to compare the contribution of diffusiophoresis for both our experiment and that of Lai *et al.* (1978), we plot in Figure 12 the elementary contribution of diffusiophoresis (E_{dph}) to the collection efficiency. This contribution is calculated with the Peters and Davenport (1978) model for 0% relative humidity (as expected for the experiments of Lai *et al.*, 1978) and 77% (as measured in our experiments). From this figure it will be noted that for the experiments of Lai *et al.* (1978), the contribution of diffusiophoresis is more than one order of magnitude higher than in ours. Furthermore, while in our experiments the contribution of diffusiophoresis is smaller than the collection efficiency simulated by Beard (1974), the opposite is observed with Lai *et al.* (1978). Thus, it appears that the experiments of Lai *et al.* (1978) cannot be compared directly to Beard (1974)'s model, because they seem to be dominated by diffusiophoresis."

Line 478. "*The reasons for the increase in collection efficiency for particles smaller than 0.65 μm in diameter are not as easy*". This sentence is badly written since all scavenging models predict an increase in collection efficiency for submicronic aerosol particles due to Brownian and turbulent diffusion processes (among others Davenport and Peters 1978, Park et al., 2005). The authors should clarify that they are considering a limited size range where Brownian diffusion is not important.

In order to better introduce the mechanisms leading to the collision of aerosol particles with drops we added a paragraph that introduces all the mechanism (p4, line 1-30).
Moreover we added a small sentence to say that Brownian diffusion is not expected to play an important contribution in the particle size range investigated: (p 19 line 17-18)

"The reasons for the increase in collection efficiency for particles smaller than 0.65 μm in diameter are not as easy to figure out. Indeed particles of this size range are not expected to be affected by Brownian motion since their diameter is seven times bigger than the mean free path of the air molecules. "

Table 1. Since the input aerosol is not monodisperse, it is not clear how the authors report the collection efficiency for the particle sizes given in the table.
We add just below the table:
"In this table, the aerosol diameter (d_{aero}) is the median aerodynamic diameter of each particle size distribution measured using the APS or the ELPI."

Table 1. UR,E is the relative measurement uncertainty which is mainly due to the contribution of fluorescein uncertainty inside the aerosol chamber (0.08). The propagation of variances equation (line 612) gives about 0.17. Table 1 (first raw) gives 4.5 10-4 as the UR,E value. If it is the absolute uncertainty, then E times UR,E (8.8 10-3 times 0.17) gives 1.5 10-3 not 4.5 10-4 as reported. The authors should explain better the data shown in Table 1.

This is an error from me. The table has been modified to give the uncertainty with all the measurement discussed in appendix.

Minor comments

Line 30. *Beard (1974).* In the reference list there are two papers from Beard (1974). Modified

Line 107. Mircea et al. is not in the references list. Modified

The reference list is not typographically uniform. Modidied

Line 424. Mdrop becomes Mgtte in equation 6 and so on. Modified

The English language of the manuscript should be revised. The article has been rephrased and an English native speaker has correct the entire manuscript

References

Ardon-Dryer K., et al., ACP, 15, 9159-9171, 2015.

Davenport H.M., et al., Atmos. Env., 12, 997-1008, 1978.

Lay K.Y., et al., J. Atmos.Sci., 35, 674-682, 1978.

Park S., et al., J. Aerosol Sci., 36, 1444-1458, 2005.

Pranesha T.S., et al., JGR, 101,D18, 23,373-23,380,1996