## **Reviewer 2**

The authors present design information, along with select performance evaluation information, for a new cloud collector which they call BOOGIE. The collector is designed based on inertial plate impaction behind 3 rectangular jets and features an air sampling rate in the range of several existing collectors. For example, the air sample rate, as pointed out by the authors is slightly higher than the CASCC2 but remains well below the air sample rate of the original CASCC. The authors use CFD to estimate droplet collection performance and compare sample collection efficiency via comparison to optically measured cloud LWC and sample composition to measurements from two widely used samplers, the CWS and CASCC2. Overall, the manuscript is well organized and the new collector design and evaluation should prove of interest to AMT readers. There are, however, several items in need of attention before the manuscript should be considered for publication including one critical error that influences several analyses.

We would like to thank the reviewer for his review and the high regard in which it holds the development work that was carried out. We answer to his/her comment below in blue.

## Major items:

1- I was surprised to see a jet impactor operating using a rotary bladed fan since jet impactors often have a relatively high pressure drop and the performance of these fans can strongly decrease at increasing pressure drop. It appears that this issue is mitigated by the relatively high drop size cut of the collector (10 um). The manuscript would be improved by a discussion about the pressure drop generated in the collectors and how this relates to the ability to collect small droplets. Why was a 10 um size cut targeted rather than a more conventional value around 5 um? Was this a design choice or just how things turned out?

We initially developed the collector based on the geometry from the CWS we used at the puy de Dôme station. Therefore, we decided to maintain the distance between the inlets and the impaction plate, and the size of the inlets where the air is drawn is also conserved. The idea was to benefit from a validated, easy-to-clean collection system, particularly for measuring the biological component. We also duplicated the number of air inlets (3) and placed the impaction plates in a vertical position. Secondly, the suction system effectively uses a rotary bladed fan (like the CASCC) to aspirate the cloud droplets into the collector. We carried out several experiments in the field to test its collection rate; these tests were conclusive.

Concerning the pressure drop inside the collector, of course it exists. We decided to use this rotary bladed fan as it allows the air to be drawn into the collection system at high-flow rate. This type of fan is also used by the CASCC2 sampler for the same reason we presume. Moreover, even if there is a pressure drop (flow rate at 600 m<sup>3</sup>/h in theory vs. 433 m<sup>3</sup>/h measured experimentally), the volume of air is still large, so collection rate of water is effective. The dilemma relies in:

- we draw in high volumes of air, but the cut-off diameter of the sampler may be higher;

- we use systems with smaller cut-off diameters, but we must increase the sampling time to get enough volume for analysis.

After testing the collector under real conditions, we then decided to carry out CFD simulation to study how the droplets were collected in the system. This theoretical calculation enabled us to analyze how the air circulates inside the system and to assess which particles were most efficiently collected. By this way, we demonstrated that few turbulences occur around the collection plates and by injecting droplets we add the possibility to estimate a theoretical cut-off diameter around 10 µm, which was reassuring. There are different ways to calculate a cut-off diameter using for example fluid mechanics (i.e., Stockes number as described in Demoz et al., 1996 and used for example recently in Du et al., 2023). But, to our opinion, it is quite difficult to compare our estimates by simulations to these calculations. Moreover, in the CFD simulations, we only consider a constant flux of the air leaving the collector since it is impossible to reproduce the fan's rotational movement.

In the new version of the manuscript, we experimentally evaluated the exit velocity of the air flow even if it is difficult to measure as the reviewer mentioned below. We decided to do that since the measurement of the air inlet velocity presented also some limitations. By this we estimated the air flow rate that is needed to estimate the collection efficiency of the collector. To understand how the air flows inside the collector and given that estimating the air flow in the sampler suffers from certain limitations, we decided to carry out numerous CFD simulations, considering different air flow velocity from 2 m/s to 10 m/s at the outlet.

Regarding the pressure drop inside the collector, we add in the new version of the manuscript this paragraph:

"The pressure drop in the BOOGIE impactor can be estimated from the fan and flow characteristics. Since the flow rate has been calculated at 433 m<sup>3</sup> h<sup>-1</sup>, the pressure drop compensated by the fan is estimated at 220 Pa, and consequently the pressure drop in the impactor is around 210 Pa. The variation in density is less than 0.0025 kg m<sup>-3</sup>, i.e. a variation of less than 0.25%. The flow can be considered incompressible, and conservation of flow-volume can be used. The average velocity at the BOOGIE inlet is estimated at 11 m s<sup>-1</sup>, by dividing the flow by the inlet cross-section of 10.9 10<sup>-3</sup> m<sup>2</sup>. This average velocity differs from the measured velocity at inlet (14 m.s<sup>-1</sup>) due to the velocity profile at the slots. The measurement corresponds to a maximum velocity."

The pressure drop is therefore rather low. Compared to atmospheric pressure, the variation is less than 0.25%, which has no influence on drop collection (no change in velocity, additional evaporation, etc.). The flow rate of 600 m<sup>3</sup>/h is where there is no pressure drop. This is the maximum flow rate supplied by the manufacturer. To our opinion, there is no relationship between cut-off diameter and pressure drop. The pressure drop is linked to pressure losses, due to air friction. The collection effect is linked to the difference in inertia between the air and the water droplets. It's linked to the air speed and its changes in direction. The more abrupt the changes in air direction, the less the drops (especially the larger ones) follow the air's trajectory and can collide with surfaces. High velocity induces more pressure loss and therefore a greater drop in pressure.

References :

Demoz, B. B., Collett, J. L., and Daube, B. C.: On the Caltech active strand cloudwater collectors, Atmos. Res., 41, 47-62, https://doi.org/10.1016/0169-8095(95)00044-5, 1996. Du, P., Nie, X., Liu, H., Hou, Z., Pan, X., Liu, H., Liu, X., Wang, X., Sun, X., Wang, Y.: Design and Evaluation of ACFC—An Automatic Cloud/Fog Collector, Atmosphere, 14, 563, 2023.

2- It is strange that the authors often present results in terms of their relationship to the exit velocity. Exit velocities behind a rotating fan are difficult to measure. Further, the relationship between flow entering the collector and flow exiting the collector depends on the pressure drop through the collector. 3- Lines 164-165: The authors' statement here about the theoretical flows entering and exiting the collector violates the principle of conservation of mass. The entering and exiting volumetric flow rates should be the same when adjusted for pressure drop through the collector. I think the theoretical ratio here is meant to refer instead to velocities.

Responses to comments 2 and 3:

Initially, we experimentally measured the air velocities of our collector at the system inlet, i.e. in front of the slots. We did this on the 3 air inlets and at different heights. We also did this by modulating the intensity of the fan. This is shown in figure S6 in the manuscript. By this way, we characterized the air flow inlet velocities for different fan intensities. This helped us to estimate the air flow entering the system that is a crucial parameter to investigate the collection efficiency of our new collector.

Next, we carried out numerical CFD simulations, keeping the geometry of the collector in its entirety. To reproduce how the air enters our system we need to constrain the air outlet flow on the rear face of the collector to reproduce the air inlet flow measured experimentally at the system inlet. We simulate air outlet flow at the fan considering the surface area of the fan; we do not reproduce the fan's rotation. We initially to not attempt to experimentally measure velocities behind the rotary fan. We did several CFD simulations to numerically estimate how each size class of droplets were collected. Regarding the areas of the slots and of the fan, based on the mass conversion, there was a theoretical ratio of 1.6 ratio between the inlet air volume flow and the air outlet volume flow. As highlighted by the CFD simulation, the air inlet velocity at the slot level is highly heterogenous. This led us to question the relevance and robustness of air velocity measurements at the slot level.

Therefore, we decided to also measure the outlet air flow velocity even if, as pointed by the reviewer, it is difficult to measure. By this way, our objective was to give a more robust estimate of the sampler airflow that is a crucial parameter to estimate the collected LWC ( $CLWC_{exp}$ ) (eq. 1 in the manuscript). This will also help us to validate the simulated values performed by CFD regarding the inlet/outlet velocities.

We added in the manuscript the following paragraph in the new section "3.2 Evaluation of the air flow inside the BOOGIE collector" describing the experiment we conducted to estimate the sampler air flow: "[...] we designed an experiment to measure the air flow at the collector outlet. The airflow rate at the fan outlet was measured using the following procedure. A 3.5 m long PVC pipe with an internal diameter of 154 mm was installed after the fan outlet. This diameter enables the entire flow to be measured without reduction, thus limiting the additional pressure losses generated by the addition of the pipe. A hot-wire anemometer was installed in the tube at 3 m from the fan. The large distance/diameter ratio (greater than 19) minimizes disturbances (high turbulence and vortex rates) as the air passes through the axial fan.

The flow velocity profile is measured every 5 mm along the diameter. Flow rate is calculated by summing the average velocity for each ring by the ring area. The flow rate was estimated at 433 m<sup>3</sup> h<sup>-1</sup> at 90% of the fan speed. The average velocity in the pipe is found by dividing the flow rate by the cross-sectional area, which corresponds to a velocity of 6.5 m s<sup>-1</sup>. Based on this velocity, the Darcy-Weisbach formula and the Moody diagram (with a relative roughness of 2 10<sup>-5</sup>), the pressure drop in the pipe is estimated at 10 Pa. As a result, the addition of the pipe has little influence on the flow rate.

The pressure drop in the BOOGIE impactor can be estimated from the fan and flow characteristics. Since the flow rate has been calculated at 433 m<sup>3</sup> h<sup>-1</sup>, the pressure drop compensated by the fan is estimated at 220 Pa, and consequently the pressure drop in the impactor is around 210 Pa. The variation in density is less than 0.0025 kg m<sup>-3</sup>, i.e. a variation of less than 0.25%. The flow can be considered incompressible, and conservation of flow-volume can be used. The average velocity at the BOOGIE inlet is estimated at 11 m s<sup>-1</sup>, by dividing the flow by the inlet cross-section of 10.9 10<sup>-3</sup> m<sup>2</sup>. This average velocity differs from the measured velocity at inlet (14 m s<sup>-1</sup>) due to the velocity profile at the slots. The measurement corresponds to a maximum velocity.".

In the previous version of the manuscript, based only on the air inlet measurements, we did calculations that were false as pointed by the reviewer due to the non-respect of the conservation of mass. Note that, in the CFD calculation, of course we do not violate the principle of conservation of mass.

We therefore re-calculate all the collected LWC ( $CLWC_{exp}$ ) and we redid the figure 4 that present experimental collected LWC vs the measured LWC. The collection efficiency (in %) were also recalculated and are indicated in Table S3 in the Supplementary Information. We also decided to include in this study additional clouds collected at the puy de Dôme, notably in spring 2024 during an international RACLET measurement field campaign as part of the ATMO-ACCESS program. 4 cloud events corresponding to 19 samples were added to our analysis.

4- I am somewhat surprised that some fraction of droplets impacted on the vertical plates are not blown off the sides of the impaction plates, pulled by the airflow around those plates. The collection efficiency results suggest this is not, however, a major issue. Can the authors comment on what prevents this from happening? Does the CFD simulation suggest there is a quiescent stagnation zone near the plate surface that prevents the drops from being pulled toward the plate edges?

For the calculation of droplet collection, in the CFD simulation, when a droplet collides with the plate it is assumed that it sticks on the surface et does not re-evaporate under the effect for example of the air flow. We analyzed the air flow velocity and the flow static pressure (Pa), relative to the ambient pressure to the collection system (i.e., plate) (Figures below). As pointed out by the reviewer, it can be noted that there is a zone close to the surface of the plates where the velocity is very reduced, possibly resulting in a zone of air stagnation close to the surface of the plate, which could prevent the droplets from being carried towards the edge of the plate. On the other hand, it can be noted that on the edges of the plate, the air flow, even if it is reduced compared to the velocity at the level of the inlet of the collector, could potentially induce the droplets to be blown out or even evaporated from the surface. This could be one of the explanations of the higher cut-off diameter we estimate for our collector. But this phenomenon is also surely the same for other cloud collectors where the air flow goes around the surface of collection.



5- There are a number of issues about the use and description of the CASCC2 that need to be clarified in a revised manuscript:

- The normal CASCC2 as described by Demoz et al. has a polycarbonate body, Teflon collection strands, and a Teflon collection trough. Was the CASCC2 borrowed for this work modified to metal construction?

Yes, it has a metal construction. The modified CASCC2 from IRSN has a metal body, stainless-steel collection strands, and a metal collection trough. This is now indicated in the manuscript: "This collector has a metal body, stainless-steel collection strands, and a metal collection trough." (section 2.3.2).

- The sampling performance of the CASCC2 depends on the fan used to pull the airflow. The analysis in Demoz et al. is based on a 115VAC Nidec-Torin TA700 fan operated at 60 Hz. What fan was used in the CASCC2 operated in the current study? The fan performance will change even with the 220 V/50Hz version of the TA700 fan. In particular, the flow rate will decrease with a lower frequency voltage supply. This should be considered in the performance analyses.

The fan that is used for the modified CASCC2 is an "4100 NH5 - S-Force ebm-papst, 24 V dc, 119 x 119 x 38mm, 45W" that was operating a frequency of 60 Hz allowing to induce an air velocity in front of strands equal to 8.6 m s<sup>-1</sup> that has been measured by a thermal anemometer at different levels. With the surface area of the collector inlet, we were able to estimate the air flow entering the collector at 348 m<sup>3</sup> h<sup>-1</sup>. In Demoz et al. (1996), two factors have been proposed to estimate the fraction of the air that actually induced the sampling of the droplets: the volume fraction of the air sampled (86%) and the volume fraction of the collected ambient droplet distribution (95%). This leads to a final sampled air flow of 284 m<sup>3</sup> h<sup>-1</sup>. We modified the text to explain this in a more descriptive way (section 2.3.2).

- The CASCC2 is sometimes operated with a downward facing inlet excluding rain. Was that used here? No, the CASCC2 used in the study was not operated with a downward facing inlet to exclude the collection of rain.

6- Lines 357-360: The authors make a critical error here in the calculation of the BOOGIE air sampling rate. They should not assume that the exit velocity matches the velocity through the impactor slots. Rather, they should use conservation of mass and assume that the exit volumetric flow rate matches the total volumetric flow rate entering the collector, adjusting for the pressure drop through the collector. Their incorrect assumption of matching velocities introduces a critical error into the collector sampling flow rate that will bias many of their subsequent analyses. This error needs to be fixed, sample flow rate corrected, and derived LWC and collection efficiency also corrected. This is a critical error and must be fixed!!

Yes, we totally agree with this. See responses to the questions  $n^{\circ}$  2 and 3 where we described the corrections we performed regarding this error. Sample flow rate was corrected as well as the derived LWC and the collection efficiency.

7- The same error discussed in item 6 appears to have been made for considering the CWS flow rate requiring similar correction and updating of results.

Yes, we totally agree. This has been corrected following your comment. We recalculated the collection efficiency based on the corrected airflow.

8- The authors should introduce and define the concepts of isokinetic and non-isokinetic sampling much earlier in the paper and address them when discussing collector performance. They should also be clearer in discussing when and how the BOOGIE collector was aligned with respect to the wind in the collection efficiency evaluation.

Yes, we totally agree with your comment.

First, the collector was installed in front of the wind at the beginning of the experiment. For this we used the wind direction data that is available at the station. During the sampling we regularly checked the wind speed direction, and we modified the orientation of the sampler accordingly.

Regarding the concepts of isokinetic and non-isokinetic sampling, this is related to the wind intensity that can modify this condition. For safety reasons, of course, we avoid installing the collectors when the wind speed is too high (more than 50 km  $h^{-1}$ ), and also because of this problem of non-isokineticity during the sampling.

In Table S3 you will find the information relative to the wind speed measured during the sampling of the 21 studied cloud events. The average values during the duration of sampling can vary from 0.2 to 14.3 m s<sup>-1</sup> (0.7 to 50 km h<sup>-1</sup>). Only the clouds collected during the 02/07/2016, the 25/04/2022, the 03/04/2024 and the 15/04/2024 present value above 8 m s<sup>-1</sup>, respectively at  $12 \pm 1.4$  m s<sup>-1</sup>,  $11 \pm 1.2$  m s<sup>-1</sup>,  $13.9 \pm 1.3$  m s<sup>-1</sup> and  $14.3 \pm 1.4$  m s<sup>-1</sup>. Those values are closed or higher to the air flow velocity of the sampler (14 m s<sup>-1</sup>). For these 4 events, the collection of droplets has been performed under possibly non isokinetic conditions. A problem with the orientation of the collector in strong wind condition can lead to significant gaps in collection efficiency. We cannot rule out the possibility that at some point the collector may not face the wind, leading to a reduction in collection efficiency, or that it may face the wind at very high intensities, leading to sampling in non-isokinetic conditions and inducing collection efficiencies more than 100%. This is clearly seen in these 4 events, which show highly heterogeneous collection efficiencies. A discussion on this point is implemented in the new version of the manuscript (section 3.3.2).

9- Figure 3. The authors should add drop sizes to the legend. In addition to the model results in Figure 3, the authors should present modeled collection efficiency (e.g., at 10 m/s) vs. drop size. This is how collector performance is typically shown. Finally, the authors should use jet velocities rather than exit velocities as the independent variable in Figure 3.

Yes, we agree that the drop size is missing on the Figure. This is not indicated in Figure 3. For the Figure 3, we prefer to present the result for different outlet velocities because we measured it experimentally and the air inlet velocity is heterogenous at the slot level. We apply the constrain on the air outlet velocities to perform the CFD simulation.

10- The authors should more clearly point out that drop composition within a cloud has been experimentally shown to vary with drop size (e.g., Bator and Collett (1997) Cloud chemistry varies with drop size. J. Geophys. Res., 102 (D23), 28071-28078) and that differences in the composition of collected samples may therefore be expected if the lower cut size for the collector changes. They kind of dance around this concept but never explain it clearly.

Yes, of course, we completely agree that the cloud composition is variable and droplet size dependent. Most of the chemical compounds measured in this study were close between the three samplers. For those which present different concentration, it could be partly due to the difference of size cut-off diameter between the samplers. We have already mentioned this point in the manuscript as one of the causes of this composition difference :

"These three samplers present specific designs and surfaces of collection (plate for BOOGIE and CWS vs strands for CASCC2), leading to different estimated cutoff diameters (around 12  $\mu$ m for BOOGIE, 7.5  $\mu$ m for CWS, and 3.5  $\mu$ m for CASCC2) and possibly to small differences in the chemical composition of the samples."

However, the real cut-off diameter for the 3 samplers and more generally for all samplers are only estimation; moreover, for the BOOGIE collector, we use CFD simulation to estimate the cut-off diameter that has not been performed in the same way for other samplers. For this reason, we think it is more prudent to not only explain the difference of chemical composition with the difference of in the cut-off diameters.

This issue about the size cut-off and the chemical composition is a major item for our scientific community. The idea of the present paper was not to make an in-depth comparison of the measurements obtained between different collectors but to mention that the measurements obtained were consistent with each other. The main objective of the paper is to present the collector and its efficiency. Moreover, within the framework of the European ACTRIS network, a special effort is being made on these aspects: intercomparison campaigns between collectors are being conducted (in which BOOGIE has been deployed) to compare their collection efficiency, and intercomparisons between cloud chemistry measurements are also being carried out. This work is still in progress, but initial results confirm that our collector is efficient, and indicate that laboratory measurements (mainly ions) are comparable between collectors, even if some bias may appear. This work will be promoted within ACTRIS (see The Centre for Cloud Water Chemistry (CCWaC): https://www.actris.eu/topical-centre/cis/centre-cloud-water-chemistry-ccwac).

11- Figure 5: I wonder if these comparisons would be better represented by scatter plots of concentrations measured in the collector pairs. As presented, it is unclear if larger differences might simply reflect measurements of species at very low concentrations.

For the most concentrate ions (*i.e.*, the major ones contributing to the Totat Inorganic Content - TIC), they are comparable as indicated in the paper for sulphate, nitrate, chloride, and ammonium. The compounds presenting the higher variability are the less concentrate ions such as magnesium (<15  $\mu$ M) and potassium (<8  $\mu$ M).

This sentence have been added in section 3.4 :

"A large variability of a factor 3 to 6 was observed for magnesium and potassium ions, but they also had the lowest concentrations, below 15 and 8  $\mu$ M, respectively (Figure S12). For the most concentrated ions, such as ammonium (over 150  $\mu$ M) and nitrate (over 50  $\mu$ M), their concentrations are comparable between samplers."

Here, we compared the difference between BOOGIE and CWS, and BOOGIE and CASCC2. The variability between CWS and CASCC2 is also high for the low concentrate ions.

By using the discrepancy factor (Df), we have probably artificially brought to the fore differences that are not the most significant because they result from the least concentrated ions. Looking at the Figure S12 in the supplementary information, these differences do not appear as pronounced.



Figure S12. Histograms presenting the concentrations of anions and cations for the three cloud samples collected using CWS, BOOGIE, and CASCC2 in parallel.

12- The authors occasionally allude to BOOGIE being capable of supercooled cloud sampling. Has this been evaluated? Are the impaction surfaces easily removable to retrieve accumulated rime? Are there

issues with the collector face or jet entrances riming and/or clogging? What about the fan? With drops up to 10 um transiting the fan I would think this could be an issue.



The BOOGIE collector could be deployed under supercooled condition. The impaction plates are easily recovered after sampling and the ice formed on the plates can be collected in bottles (see photo).

The collection period must be short to avoid clogging the slots and causing fan failure. This type of sampling can therefore be carried out, but we are not very confident about the measurements that will be made on this sample, particularly concerning volatile species, possibly not trapped in the ice formed. We avoid collecting supercooled cloud event, and even if we analyzed the sample chemical and biological composition from these events, these data were never considered in our data base.

In addition, the geometry of the collection surface changes over time,

leading to a bias in drop collection efficiency. Consequently, we sincerely prefer to avoid collecting under supercooled conditions for all these reasons. Regarding the reviewer comment, we do not find in the manuscript where we mentioned the possibility to collect with BOOGIE under supercooled condition. We only mention that the CWS can operate under this condition and that it exists an upgraded version of the CASCC2 was designed for supercooled cloud sampling: the Caltech Heated Rod Cloud Collector (CHRCC). We prefer to not add additional text relative to the possible sampling of supercooled cloud with the BOOGIE collector, to avoid any confusion.

## Minor items:

We would like to thank the reviewer for taking the time to look carefully at the small errors in the manuscript. We answer below.

- The manuscript does not adequately describe turbulent conditions in the collector. As far as I can tell, the only evaluation of turbulence is through the CFD modeling and that is not well described. There are also ambiguous claims in the abstract and conclusions about "few turbulences have been observed...". Were there any "observations" of turbulence and, if so, what do these claims mean?

Yes, we agree with the reviewer comment. Our conclusions are not effectively based on a turbulence calculation/assessment in the collector. Turbulence is considered in numerical CFD simulations, as mentioned in section 2.2. Of course, some amount of turbulence could certainly be observed, particularly around the collection zone. We have therefore chosen to delete these sentences to avoid drawing overly hasty conclusions.

- Line 103: change "where and their designs" to "where their designs" Done.

- Line 107: Change "Caltech University" to California Institute of Technology" Done.

- Lines 112-113: Her the authors refer to "The sampler". This is confusing as written. I first thought this was referring to the BOOGIE sampler but I think it actually refers to the CWS. Please rewrite this sentence to be clear.

Thanks, we rewrite this sentence following your comment.

- Lines 198-199: please specify here the optical instrument used to measure LWC (PVM-100). Done.

- Line 223: replace "resistance" with "drag" Done.

- Line 228: replace "strains" with "strands" Done.

- The authors should define the drop effective radius and clearly point out that this is not the mean physical radius.

Line 353: we clarify this point following your comment.

- Lines 396 and 397: Change "PWM" to "PVM." Why can't the PVM output be recorded at higher time resolution? It's just a voltage.

Done. For the time resolution, since this instrument is used to monitor the LWC over long period of time, the data acquisition frequency is 5 min for data storage reasons.

- Line 435: The sentence here is garbled. Please rewrite. Done.

- Line 531: Replace "in a radius" with "in radius" Done.

- Line 548: Replace "would present" with "would likely present" Done.

- Line 560: It would be worth pointing out that there is a version of the CASCC family specifically designed for supercooled cloud sampling: the Caltech Heated Rod Cloud Collector (CHRCC). This is discussed by Demoz et al. and other references.

We add this information in the description of the CASCC2 (section 2.3.2).