

Reviewer 3

Vaitilingom et al. present the design of a new bulk cloud water collector (BOOGIE) which has been tested and applied at the Puy de Dome station and will be used for studies of biological and chemical composition. The new collector is well-motivated by shortcomings of existing designs especially with regards to sampling rates and ease of operation for biological applications and its characteristics and comparisons with other designs are reported and discussed. I have a number of comments and suggestions the authors might want to consider, and I recommend publication after these have been appropriately addressed.

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 comments

1- The intercomparison of solute concentrations (inorganic ions, formaldehyde and H₂O₂) between different collector designs is at the core of the collector evaluation, because this is what these instruments are made for in the first place. Unfortunately, this part of the manuscript is a bit weak. Only two samples were taken in parallel with the CWS, which has been in operation for more than 20 years at the station, and three with the CASCC2, another popular design. Based on such low statistics, it is difficult to robustly judge on the comparability of cloud water solute concentrations. I wonder these are really the only available data to compare? Given that the first BOOGIE application took place 8 years ago already, I would have expected a larger database for comparison. In case there is more data, I would suggest to include it to the manuscript, which would much strengthen the intercomparison section.

We agree with you comment of course. Unfortunately, we did this intercomparison in 2016 when the BOOGIE was under evaluation. We did not perform other inter-comparisons between the samplers at the puy de Dôme station. There are several reasons for that. The first most important point is that these intercomparisons of cloud collectors is currently underway in the frame of the European ACTRIS network. Measurement campaigns at various sites (Mont Schmücke, Germany; Puy de Dôme, France; Mont Sonnblick, Austria) have been carried out over the course of 2023 and 2024 to intercomparison the collectors deployed on a European scale: their collection efficiency under various environmental conditions and the resulting chemical water composition. This work is currently being finalized, and without revealing too many conclusions about it, it highlights the good performance of our collector. The second point is that we do not own the CASCC2 collector since it belongs to our colleagues from the IRSN institute, in France; so we could have only performed intercomparisons between the BOOGIE and CWS collector that is not sufficient to our point of view. Finally, since the BOOGIE collector presented high collection rates, over the last few years, we have prioritized the use of the BOOGIE collector vs the CWS to sample clouds at the puy de Dôme. The section in the paper on chemical composition intercomparison is admittedly rather preliminary and will in future be conducted within the ACTRIS network. We hope these arguments convince you.

To strengthen the section on the efficiency of collection, we added 4 cloud events (i.e., 19 samples) to the studied database.

2- The intercomparison of ion concentrations is discussed in a bit of an optimistic way. Deviations of up to a factor of about 2 ($Df < 0.5$) are assessed as “good comparability”, but it remains unclear what this positive assessment is based on. Would systematic deviations in this range not pose a problem to the long-term trends of concentration data at the station? How do the deviations compare to earlier collector intercomparison exercises, for individual ions as well as overall? In a few extreme cases, deviations up to factors of 3 and 6 were observed between BOOGIE and CASCC2, but these are hardly discussed.

Instead, it is implied that CASSC2 concentrations “appear” slightly higher” and only “at first glance”. I’d recommend to revise this section in a more detailed and more critical way. Overall, the results seem to call for more systematic intercomparisons with a larger number of samples.

The intercomparison of ion concentrations is a delicate point to interpret. Globally, most of the chemical compounds measured in this study were similar from the three samplers. It is the case for formaldehyde, hydrogen peroxide and some inorganic ions, as well for the microbial parameters (ATP/ADP). For the most concentrate ions (*i.e.*, the major ones contributing to the Total 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 μM) and potassium (<8 μM).

This sentence have been added :

“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 μM , respectively (Figure S12). For the most concentrated ions, such as ammonium (over 150 μM) and nitrate (over 50 μM), their concentrations are comparable between samplers.”.

Here, we compared the difference between BOOGIE and CWS, and BOOGIE and CASSC2. The variability between CWS and CASSC2 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.

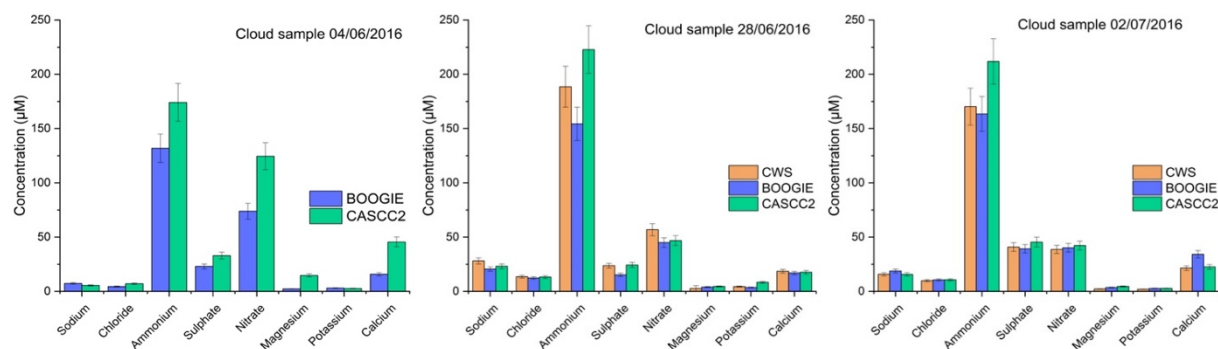
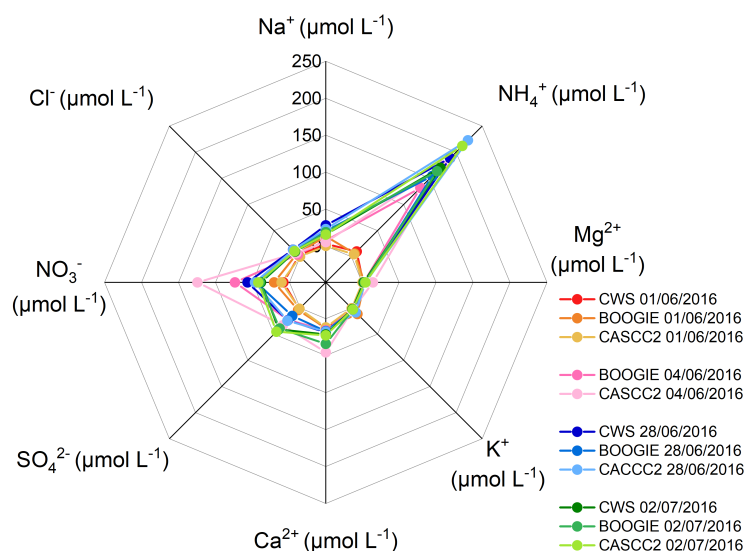


Figure S12. Histograms presenting the concentrations of anions and cations for the three cloud samples collected using CWS, BOOGIE, and CASSC2 in parallel.

Following the reviewer’s comment, we tested new figures to highlight the differences between collectors. For example, we proposed the radar plot but it seems not really adequate as it is difficult to read.



Radar plot describing the differences between the collectors for the 3 cloud events.

We prefer the histograms proposed in the SI (Figure S12).

We used to see variable concentration even for a same cloud event with identical collector. An example was given in the SI text, see Fig S13.

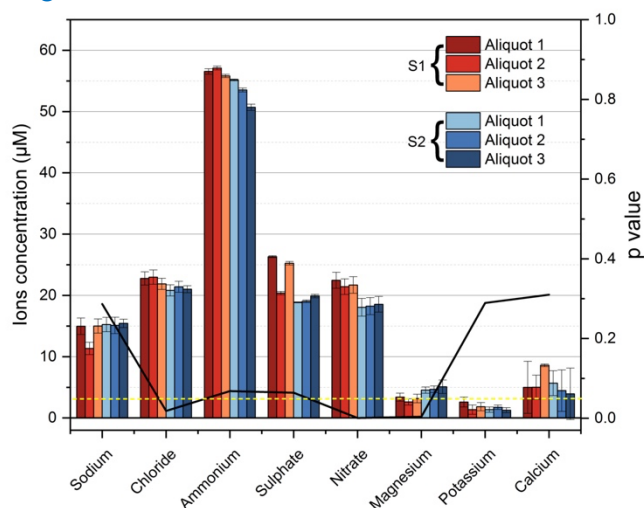


Figure S13. Histograms presenting the concentrations for a specific cloud sampled on 08/07/2021 at PUY with two BOOGIE collectors. This time, three aliquots were analysed twice (error bars) using ion chromatography. p-values are indicated with the black line and the yellow dashed line indicates the threshold of $p = 0.05$.

More generally, inherently linked to the collection method, the composition of the water sample is not a perfect representation of the real cloud water composition. Through the impaction process by all passive and active samplers, a bias is added. Several parameters can affect the cloud composition: the collection temperature, the matter of the body collector and the bottle, the time delay between the impaction and its flowing inside the vessel, the variation of air/liquid volume of the collection bottle. These conditions affect the liquid/gas exchange, even the microorganisms will modify the chemical composition by their metabolic activity during the sampling time. For all these reasons, we consider the difference of composition in our study acceptable even for the less concentrate compound.

But as mentioned by the reviewer for a long-term trends of concentration data at the station, a full comparison between the cloud samplers is required. We work on it, a special effort is being made on these aspects at the European level: 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>).

3- There are a number of inconsistencies for some quantitative results between abstract, main part and conclusions, which I will give below. In general, I'd suggest to critically check that correct and consistent information is given everywhere throughout the manuscript. Also, the wording and sentence structure would benefit from a critical check here and there.

Yes, we totally agree with your comment. We carefully check this in the revised version of the manuscript. We hope this is now correct. Concerning the wording and sentence structure, before the submission, we opted and paid for a high-standard language editing service to check the writing. We have already made this effort, so if the reviewer ask for additional corrections, we think that the journal might be able to take on this task.

4- Fig. 4 and its discussion are a bit misleading. Based on the data points, I believe the authors have forced their regression through zero, which means their slope, intercept and coefficient of determination are incorrect. Forcing through zero should only be done for very good reasons. In this case, the intercept could actually be informative and should not be artificially removed. With a simple linear regression, I would expect a slightly negative intercept and a slope close to 1. Given that small unrecovered amounts of water from inner surfaces or even slight evaporation inside the collector seem plausible, I would not expect an intercept of 0. Also, the collection efficiency does not seem to decrease at higher LWC, based on just the data points. This seems to be an artifact from the applied regression through zero. Please carefully check and revise accordingly.

Yes, we forced the regression through zero. Following the reviewer comment regarding the possibility of some artefacts (small unrecovered amount of water from inner surface or possibly slight evaporation inside the collector that we cannot avoid), we changed the regression. The intercept is now equal to -0.02, a negative value as envisaged by the reviewer and the slope equal to 0.92.

Concerning the relationships we described (decrease of collection efficiency at higher LWC_{mes}), this is true, there is no clear tendency. We changed the text accordingly.

5- L357ff: From the explanations, it seems outlet velocity (not inlet velocity) together with inlet surface area has been used to calculate the air volume flow rate. This would be wrong and would need to be corrected throughout the paper, including any discussions and conclusions that might change with a corrected air flow rate.

The inlet velocity is estimated by measuring the outer flow. Understanding the questioning of the reviewers, we have experimentally evaluated new flow rate measurement during the revision time delay, and this is presented in a new section (3.2 Evaluation of the air flow inside the BOOGIE collector). Consequently, with a new air flow, we recalculated the collection efficiency of the BOOGIE collector and modified the discussion accordingly.

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: "[...] Therefore, 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 $433 \text{ m}^3 \text{ h}^{-1}$ 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^{-1} . Based on this velocity, the Darcy-Weisbach formula and the Moody diagram (with a relative roughness of $2 \cdot 10^{-5}$), 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 $433 \text{ m}^3 \text{ h}^{-1}$, 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^{-3} , 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^{-1} , by dividing the flow by the inlet cross-section of $10.9 \cdot 10^{-3} \text{ m}^2$. This average velocity differs from the measured velocity at inlet (14 m s^{-1}) 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.

Further issues

- The title should be revised as it is uncommon to include two colons. Also, it becomes clear at second glance only, why oxidants and metal elements are mentioned. These are not measured and discussed in the manuscript, but seem to be part of the collector name. This could be made clearer, e.g. like this: “Design and evaluation of a new cloud water collector for the analysis of biological, ..., and metal elements (BOOGIE). Actually, nowhere in the paper it is explained where the name BOOGIE originates from.

We simplified the title of the paper following your comment. Now, the title is shorter: “Design and evaluation of BOOGIE: a collector for the analysis of cloud composition and processes”. At the end of the introduction, we explained why we decided to name the collector BOOGIE. This refers to the all the elements that can be measured after the collection of the water: “Biological, Organics, Oxidants, soluble Gases, inorganic Ions and metal Elements”.

To be honest, the collector's name “boogie” is a tribute to an “entity” that has been important in cloud-related activities on the Puy de Dôme site. Of course, linking this acronym to the description of what the collector does can be questionable. Our aim was not to oversell what our collector can do, since it only collects water! Collecting cloud waters among various environmental conditions is only done with the objective to perform analysis in the lab helps that are crucial to assess the effect of clouds on atmospheric chemistry.

- Results of the collector intercomparison should be summarized in a more quantitative way in the abstract and conclusion sections. The purely qualitative and subjective statement of “comparable” concentrations is not very informative. To not lengthen the abstract further, the introduction on the importance of cloud water composition could be shortened.

We modified the abstract following the reviewer comment: we shortened the introduction of the abstract and added results from the collector intercomparison. We also suppress in the conclusion the paragraph describing the different collectors used for the intercomparison experiment because it is redundant. We prefer not to add more details in the conclusion regarding the collector intercomparison. We made this choice to avoid redundancy.

- L28: Doe you mean “lightweight” or “simple” or “easy to operate” mobile sampler?

Yes, this sentence is unclear. Following your comment, we added “easy to operate”.

- L33 and L346: The mean collection rate is inconsistent between abstract and results section. Please check and correct.

Yes, this is true. The good collection rate is $100 \pm 53 \text{ mL h}^{-1}$. This has been corrected.

- L60: What does HAP mean? Make sure to introduce all abbreviations at first occurrence.

We are sorry, HAP is the French abbreviations. We modified by “polycyclic aromatic hydrocarbon (PAH)”.

- L103: delete first “and”

Done.

- L112: Was the sampler indeed obtained from Kruisz et al. or built according to their design?

We modified this following your comment.

- L256: Do you mean “More information on this analysis is given in ...”?

We modified the text following your comment.

- L306: When you say “classes of particles”, do you mean just different sizes? Or has anything else been changed for the different “classes”?

Yes, you are right. We injected droplets from different sizes of droplet in the collector. We modified the manuscript to avoid any confusion.

- L310-311: Incomprehensible sentence, please rephrase.

This has been modified, thanks.

- Fig. 3 would be easier to understand, if droplet sizes were given directly in the legend, rather than arbitrary “class” numbers. Also, I’d suggest a more compact y-axis label without uncommon abbreviations, e.g. “number collection efficiency” and “mass collection efficiency”

Yes, we have modified the figure following your comment.

- L315ff: It is not always clear which type of collection efficiency is discussed (number vs. mass). Please make sure to correct accordingly.

This is true. We modified the text accordingly.

- L316: At velocities below 5 m/s, the collection efficiencies of large droplets are not always > 50%. Please check and revise sentence.

Yes, this is true. This has been corrected following the comment.

- L319: Does the given average collection efficiency not strongly depend on the droplet sizes which were simulated? If more larger sizes were simulated, the average collection efficiency would be different. Please clarify what can be learned from this calculated average efficiency?

The average collection efficiency depends on the simulated droplet sizes. We have chosen the different sizes based on the size distribution encountered in clouds. The largest droplets present a diameter of 20 μm . Above all, the average efficiency shows the importance of the largest droplets, both in mass and number, in the total collection.

Of course, if we add supplementary droplet size the average collection efficiency should be modified. This change would be most noticeable in term of mass collection efficiency, as the largest droplets would make a significant contribution to the total mass. But this simulation performed were only done to evaluate which droplet sizes are the most efficiently collected. The average collection efficiency is shown in the figure for information only, from our point of view.

- L324-328: This section is difficult to understand and partly redundant (50% cut-off at 10 microns). Please revise.

Yes, this is true. We modified the text following this comment.

- L335: Why are the theoretical efficiencies assessed as “good”? Are there any comparisons with other data that would substantiate this judgment?

Yes, these conclusions are not supported by quantitative data, such as comparisons with other collectors. Therefore, we modified the sentence as following:

“However, the performed simulations indicate that the new BOOGIE collector is able to collect cloud droplets, which also confirms that the distance between the air inlet slots, and the outlet fan is adequate because it is beneficial for air flow stabilisation.”

- L340ff: Please check for repeated, i.e. redundant information.

This has been modified accordingly.

- L342: It would help the reader to give a brief summary of the main characteristics of the sampled events.

This has been added in the manuscript following this comment.

- L354: In the section before, the optimal velocity is given as 10, not 8 m/s. Please check and correct.

This was modified in the new version of the manuscript.

This point has been removed; a new section has been added to estimate the flow measurement « 3.2 Evaluation of the air flow inside the BOOGIE collector ».

Now the new sentence is : “To evaluate $CLWC_{\text{exp}}$, we estimated in section 3.2, the sampled air flow experimentally at $433 \text{ m}^3 \text{ h}^{-1}$ ($7.22 \text{ m}^3 \text{ min}^{-1}$).”

- L359: In L164, the total inlet surface is given as 11.088E-3, please check and correct.

This has been changed in the new manuscript.

- L373 and elsewhere: LWC_{meas} , not LWC_{mes}

This has been modified.

- L396: PVM-100

Done.

- L397: The acquisition rate of the PVM could easily be increased, but I doubt it is really a relevant factor here.

We used the LWC measured by the PVM to estimate LWC_{meas} that allows calculating the sampling efficiency of the collector. Recording the LWC only every 5 minutes may lead to errors in the estimation of the real LWC during sampling. Indeed, the LWC could vary significantly between two measurement points. Of course, this depends on the conditions encountered during the sampling. We would prefer to leave this point to our discussion of potential sources of error in assessing the efficiency of collecting.

- L398: Why is CLWC_{exp} “intrinsically an estimate”? In the previous section it is explained as a value from measured velocities and geometries.

Yes, this is true. The sampler airflow is not an estimate since it is calculated using measurement of velocities and geometries (surface of the entry slots). We modified the text accordingly.

- L417/418: Harmonize between “radius” and “diameter”

We modified accordingly.

- Table 1: Rain intensities during the events should be included as well. The information on possible rain drop contamination is given late in the discussion only, but is quite a relevant information.

Unfortunately, rain is not measured at PUY with a pluviometer. In the case of rain during the sampling, we indicate this event in the text : “In the case of the 4th June cloud, the appearance of fine rain during sampling could possibly explain the higher of collection efficiency observed for all collectors, as we did not observe conditions such as strong winds that could disrupt the sampling.”.

We modified the legend of the table 1 as suggested : “** Fine raining event before the end of sampling”.

- L438: Not sure if it is really advisable to use collected water volumes as a metric for cloud LWC.

Yes, we agree. We delete this sentence.

- Fig. 5: What does the “analytical error” of 10% represent and how was it determined? In the Supplement, “accuracy” is mentioned, but precision or repeatability would be more appropriate metrics here. Is it really correct that this value is the same for minor ions with low concentrations and for major ions with usually larger peaks?

The uncertainty of measurement varies from 6 to 10% for these ions analysis. The analytical error is obtained by the calculation of the standard deviation on two analyses of the same sample. The values obtained are comparable to those obtained by the standard deviation from 3 replicates of calibration curve using a standard solution (Multi Ion cation and anion IC standard solution, Specpure™ (Dionex)). Even for the low concentrations of magnesium or potassium measured, these values were largely higher than the limit of quantification.

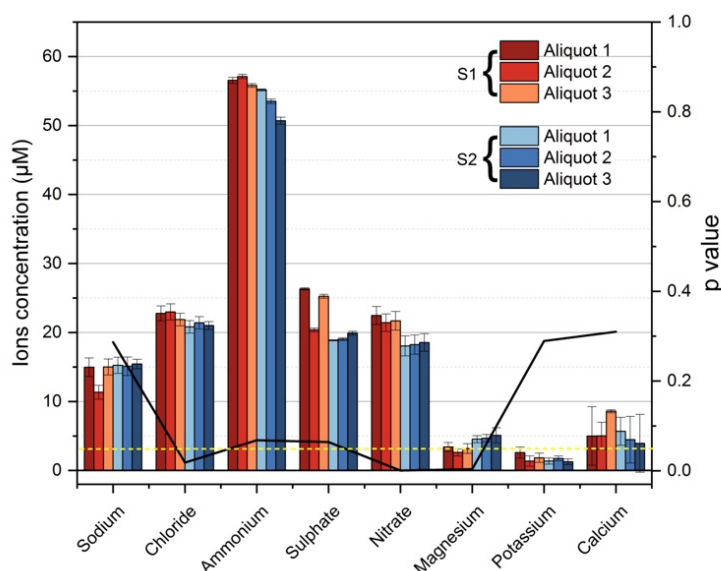
- Fig. 6 could go into the Supplement, as its discussion is brief and does not add much. Instead, Fig. S13 could be shown in the main manuscript as it presents a different aspect of the evaluation, i.e. comparability between two identical BOOGIEs. It might even deserve its own subsection.

This Figure 6 is useful and presents the concentrations of formaldehyde and hydrogen peroxide of three cloud event with the three samplers; the discussion is brief because the interpretation is straightforward. Their concentrations were similar between the samplers and this information is relevant.

Yes, as suggested the Fig S13 has been added to the main text.

- In Fig. S13, why are some of the aliquots much different from the other two of one and the same sample? Measurement precision from repeated IC analysis seems to be in the low percent range and can thus not explain these deviations. How much do these “inconsistent” aliquots statistically impact the between-collector comparison?

Due to the low concentration of cloud water constituents, we make sure to analyze the samples with the highest sensitivity for the instrument used (Thermo ICS 5000+). This goal is achieved by injecting a high volume of the sample (750 μL). On one hand, the limit of detection is below 0.1 μM for less concentrated ions, on the other hand, the high concentrated ions show peaks that are sometimes broad. In the context of a complex medium, with a baseline populated by other signals, such as those related to short chain carboxylic acids, integration can be sometimes difficult and produce sometimes a variability of the signal. This is the reason why we always inject the sample at least twice. In this plot, we reported the concentrations calculated for each single injection as a strict test for both the sampling error and the analytical error. If we consider the average of the three injections (Figure S13 in the SI that is now Figure 7 in the article) the difference between the two collectors is less visible.



- L487: In the Supplement, it says “duplicate” analyses, not triplicate.

The sentence in the text is: “Analyses were performed in duplicate or triplicate if the standard deviation of measurement exceeded 5%, the DL was of 0.07 μM (Vařtilingom et al., 2013).”

The analysis is performed in duplicate, and if the standard deviation exceeds 5%, a third analysis is performed.

- L504/505: These comparisons should be made in a more detailed way, i.e. ion by ion and making sure the metric for “difference” is really identical.

A more detailed comparison of the similarity and discrepancies between different collectors needs more samples, collected in different environments, to build a more robust dataset and perform consistent statistical study. This goes beyond the scope of this article. Nevertheless, this comparison is already ongoing in the ACTRIS CIS CCWAC working group and will be published in a near future.

- L505: Are these values consistent with what is discussed earlier?

We cannot focus mainly on the ionic composition of minor ions which present high variation. We agree with that, but these differences were also measured from other studies. As the one mentioned in the manuscript (Wieprecht et al., 2005).

Even during a same cloud event, the composition varies. The particulate, the gas and the liquid composition is heterogenous inside a cloud.

From our experience and from the literature where samplers were compared, yes we still agree that the chemical composition and microbial energetic states between the samplers are close in this study. The formaldehyde and peroxide contents are similar between the three samplers; the microbial energetic states are barely identical. About the 8 ions measured, 3 of them (also the less concentrated) show a high variability and 5 of them have close concentrations. Thus, globally, we consider that the composition are “comparable” and have a “good assessments” between the samplers.

- L506ff: The comparison of biological data might warrant its own subsection. Right now, it is a brief extension on the chemical comparisons, but a bit difficult to understand without more context.

The microbial measurement here gives only the metabolic energetic state. It was done to see if the collection process was stressful or not for microorganisms. The energetic state reveals a good level of microbial “health” with identical values from each collector, which indicate that the collection is not stressful from the three samplers. This information is valuable but does not deserve more discussion in this section.

- The conclusion section repeats much basic information and explanations from earlier sections. It could be improved by summarizing in a more compact way the core results and conclusions.

We agree with this comment and have modified this section. A paragraph has been deleted that presented the different collectors for example, because it was redundant.

- L559: The results presented before do not seem to support the statement of high collection efficiency of the CASSC2, it was actually the lowest one among the compared designs.

All the calculations regarding the collection efficiencies of the collectors have been updated following the reviewer’s comments. The CASCC2 present now the highest collection efficiency with the BOOGIE collector.

- L560: How is the applied CASSC2 model not affected by rain? The data of the cloud event with slight rain do not seem to support this statement.

The sentence was not complete from our first draft. The original sentence was: “This active sampler is a compact version of the CASCC, in which droplets are collected by impaction on a set of six rows of stainless-steel strings; it is highly efficient in terms of collection and is not affected by raindrops owing to its design by the using of a rain shield.”

It has been modified, but this paragraph summarizing the three collectors was effectively too long and redundant to the introduction. We agree with the reviewers 2 and 3 and this paragraph has been deleted. We add some precision about the CASSC2 in the M&M section :

“The collector body was stainless steel, the inlet contained the impaction rows, and the sample drainage was removed before each sampling for cleaning and sterilisation. A sterilised amber glass bottle was placed under the sample drainage during collection. The CASCC2 was also not operated with a downward facing inlet allowing to exclude the collection of rain. This cloud collector was not adapted for temperatures <0 °C because droplets freeze upon impaction on metallic strands. Note that an upgraded version of the CASCC family was specifically designed for supercooled cloud sampling, the Caltech Heated Rod Cloud Collector (CHRCC).”