Public justification (visible to the public if the article is accepted and published):
This manuscript contains and important and well-conducted evaluation of the performance of water-based condensation particle counter (CPC) at low pressure. This CPC will be part of an instrument package on the IAGOS measurement suite flying on commercial aircraft. The IAGOS data will be broadly used to improve understanding of aerosol particles in the free troposphere, and is unique in its temporal and spatial coverage. As such, this work is eminently appropriate for publication in AMT, and it is important that the instrument's performance be well documented.

The referee reports of the original manuscript all expressed concerns regarding the clarity and presentation quality of the results. The authors have moved and expanded some technical aspects of changes to the instrument's behavior at low pressure to the supplemental information, retaining a brief description of this topic in the main text.

While the manuscript is improved, it still suffers from a lack of clarity, typographical errors, and some technical issues. While many of these were pointed out by the referees and corrected by the authors, many remain. Because of this, the manuscript is not yet ready for publication.

I point out some of remaining issues below.

1) Typographical errors. There are a number of mis-spellings, incomplete sentences, and other errors in the manuscript. A thorough copy editing job is essential, and an English spell checker must be run to identify some of these errors.

2) Clarity. There are several sentences that are not clear. Some examples (but not all of the cases):
a) Lines 21-22 say, "the D50 cut-off diameter did not differ significantly for particle sizes around 10 nm, whereas. . . .". I think the meaning is, "the D50 cut-off diameter of ~10 nm did not vary substantially as a function of pressure, whereas the D90 cut-off diameter. . . ." (D90 is not defined, by the way.)

You Are Right! We have clarified the points in the text.

b) Line 45, "only reach best performance at low pressure levels". I think the meaning is that butanol CPCs perform best at higher pressures, closer to sea level.

This was originally aimed for fluorinert. We changed the text accordingly.

c) Lines 84-85. "The diffusion losses are assumed to similar for all instruments." Besides missing the "be", this sentence implies an assumption, but in the next sentence we learn that the tubing lengths to the various instruments were adjusted inversely with flow rate, so that diffusion losses were the same for each instrument; no assumption needed.

We deleted "assumed"

d) Lines 118-122. This is a non-sequitur; a statement that is not in the context of the preceding narrative. An exponential fit function? To what? How is the Wiedensohler function related to Equation 1? This is very confusing. I believe the first fit function is for the steady-state charge distribution, but this is not at all clear in the text.

EQ1 is a parametric exponential function, that is used to fit the cut-off curves. We have claryfied the points in the text

e) Lines 208-210. The first part of this sentence says that airborne measurements are not likely to encounter fresh soot, then the second part describes cases where airborne measurements will encounter fresh soot.

The overall likelihood to find FRESH soot at crusing altitude (11-13km) is low. But we have observed and identified fresh aircraft plumes onboard our CARIBIC Aricraft by parallel CPC and NOy measurement. These aircraft plumes are visible in time series as 1-2 sec peaks.. We have clarified this in the text.

f) Lines 154-155. If I didn't understand how DMAs operate, I would have no idea what is being discussed here. Please elaborate that you chose to operate the DMA at flow conditions that would allow selection of particles as small as 3.5 nm; as a result, the maximum size that could be selected was 138 nm.

We add the flow condition with 6 L/min sheat flow and the 8.8 cm tube length. We have clarified this in the text.

3) Scientific/Technical issues. There are some scientific/technical aspects of the manuscript that could be improved, including:
a) Please use a logarithmic axis for all the plots using diameter on an axis. This would allow the greater data resolution at smaller diameters to be clearly seen, and most aerosol parameters naturally scale logarithmically.

We changed those in the manuscript

b) The function used to fit to the detection efficiency curves does not describe the data very well in Fig. 4b. Have you considered trying the error function, which is the integral of the lognormal distribution and thus physically based rather than purely parametric? A good reference describing this is Deshler et al., JGR, 2019, doi:10.1029/2018JD029558.

To be consistent with prior studies, that characterised Butanol CPC instruments, we use this exponential function by Wiedensohler. We redid the fitting function

c) Does Fig. 5 include data from all diameters, or is the variation in measured concentration for one diameter? Does this range of concentrations encompass the likely range to be observed by the IAGOS package (i.e., from prior measurements in the free troposphere)? I'm curious if there is a roll-off in detection efficiency at higher concentrations. And might there be a roll-off in D50 as well? Remembering that ambient concentration at high altitude will be less than sea-level values, did your measurements cover the expected range? If not, that might be a topic for future studies.

Yes, Fig. 5 include data from all Diameters.

Observation from Butanol CPC instrument during CARIBIC measurements rarely exceeded 6000 cm^{-3} particles, therefore we did not observe a roll-off.

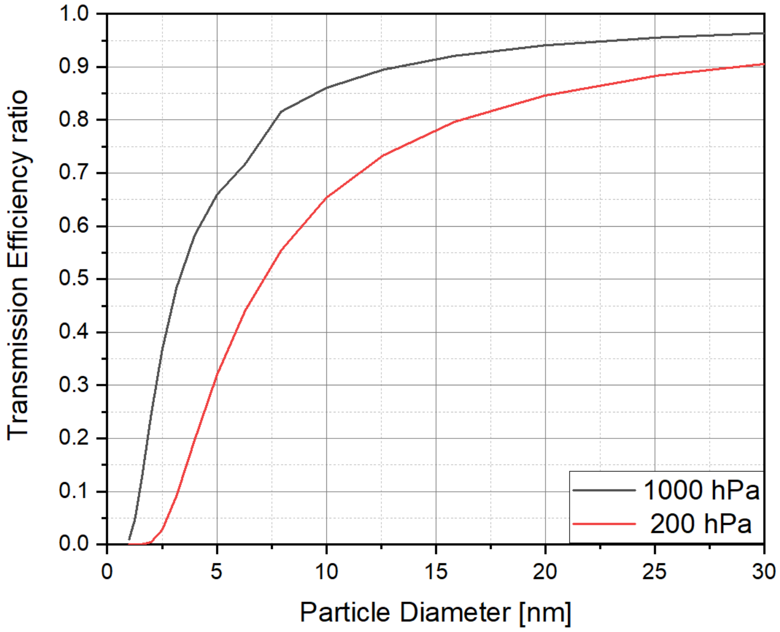
d) In the Conclusions and Recommendations, it is mentioned that "we were able to have a look at 5 units to verify that this was not an artefact of a single unit." This is extremely important, because the data from multiple instruments on multiple aircraft will likely be combined into a single dataset. If you have quantitative data regarding instrumental variability, please show it, even if it is limited. It is very important to understand how large the unit-to-unit variability might be.

We did not run the instruments in parallel to give a quantitative statement for the cut-off diameters. This, however, will be done in future studies, with the goal of SOP (Standard operation procedures).

e) The discussion of diffusion losses of particles is still muddled. Please clearly indicate whether the fall-off in detection efficiency with decreasing diameter at low pressure is due to diffusion losses within the instrument, or if it is more likely caused by pressure-dependent changes in detection due to smaller pulses. I also suggest including a figure showing the calculated diffusion losses in the IAGOs configuration using the one meter tubing length mentioned in the manuscript. You could calculate the losses at 300, 250, and 200 hPa over the entire detected size range. This would be very informative for researchers who will use these data in the future.

We will include a Figure for 200 hPA and 1000 hPa and the equations in the SI. We used Hinds, W. C., Aerosol Technology: Properties, Behavior, and Measurement of Airborne Particles. Wiley: 1999.

22. Baron, P. A.; Willeke, K., Aerosol measurement: principles, techniques, and applications. 2001.



Aerosol sampling is an important issue, when aerosol measurements claim to cover a total range of aerosol properties and size ranges. Particles can be lost in the sampling line by impaction, sedimentation or diffusion. For very small particles, the diffusion is the most important mechanism for particle losses. Those can be illustrated by diffusion losses in a cylindrical tube under laminar flow. The first thing to recognize is the Stokes`s Law. This fundamental force F describes the total resisting force a spherical particle experience by moving through a medium.

$$F = 3\pi\eta v d$$

With η is the viscosity of the medium, v the velocity of the particle and d the diameter of the particle. The Cunningham correction Cc factor has to correct an important assumption of this force. The assumption of the Stokes`s Law is, that the relative velocity of the gas at the surface of the sphere is zero. This assumption becomes an issue for small particles at the range of the mean free path.

$$F = \frac{3\pi\eta v d}{C_c}$$

The Cunningham correction factor can be calculated using the mean free path χ by

$$C_c = 1 + \frac{\chi}{d}[2.514 + 0.8 \exp\left(-0.39\frac{d}{\chi}\right)]$$

Then the diffusion coefficient ξ for laminar flow can be calculated considering the temperature T by

$$\xi = \frac{1.38 \cdot 10^{-23}[\frac{J}{K}] \cdot T \cdot C_c}{3\pi\eta d}$$

With this the dimensionless diffusion parameter μ can be calculated with the length of the tube l and the air flow rate f.

$$\mu = \xi\frac{l}{f}$$

Finally, the loss fraction can be described by

$$Loss = 0.819e^{-11.5\mu} + 0.0975e^{-70.1\mu} + 0.0325e^{-179\mu}$$

The final line loss is the product of two settings. Here, the main aerosol sampling line with a flow rate of 4 l/min for the final instrument package and a length of 1 meter till the splitting point, where the MAGIC-LP has a flow rate of 0.3 l/min and 30 cm distance.

f) What is the volume of the mixing chamber and its e-folding flushing time for the flows used? This information was suggested by one of the referees.

The volume of the mixing chamber is 500 ml with a flow rate of 10 l/min. This leads to a flushing time of roughly 3 sec and an e-folding time of 1.8 sec for 63%

g) Lines 127-129. The detector offset appears here without explanation Perhaps say, "when the internal instrument pressure decreases, laser power is automatically increased. As a result, increasing background scattered laser light effectively increases the baseline voltage of the detector, causing the particle pulse detector threshold to. . . ." And I stop the sentence here because I don't understand what these sentences are trying to say. Please make it very clear what is going on with this detection circuitry, and refer to the SI for more detail. I still don't understand if the baseline change with laser power is a deliberate, controlled shift, or if it is just the result of more scattered light.

We changed the text by: The first variable is the laser power which is adjusted to compensate for variations in droplet size as a function of the operating pressure. With decreasing pressure levels, droplet growth is affected, making the droplets smaller. The laser power automatically increases as the internal instrument pressure decreases to compensate for the smaller droplet size. The second variable is the detection threshold voltage which is adjusted to compensate for variations in background scattered light (i.e., measured light with zero particle counts) as the laser power varies, since the background light scattering on molecules increases with increasing laser power […]The manufacturer's settings were not optimized for operating pressure down to 200 hPa. For 250 hPa, we found, that the required laser power was increased so high by the firmware to compensate for the smaller particle sizes, thus the electronics could not determine the baseline voltage correctly. By adjusting the values for the laser power, detector threshold and offset, we solved this issue thus, the MAGIC LP-210 is now able to operate even below 250 hPa. The new settings are applicable for the complete pressure range without change. Further explanation on this is given in the SI.

4) Copy editing. There are some fairly egregious errors that should not have made it into a submitted manuscript.

a) Figure numbers have changed, but were not updated in the text.

We changed those in the text.

b) The butanol CPC is sometimes referred to as a "Sky-CPC" and sometimes as a "G-CPC". It took me a while to figure out that this was the same instrument. Please be consistent with nomenclature.

We will check to be consistent

c) The atomizer is referred to as a nebulizer in Fig. 1 (which is much improved from the initial submission).

We used Nebulizer and Atomizer as synonyms. We check this to be consistent.

d) What is PID? Also, in Supplemental Information Fig. 1, please make sure all variables are unambiguously defined.

We update the text with a short explanation: proportional-integral-derivative controller

e) Line 124. "Low Pressure" has already been defined as part of the instrument name.

We deleted this for the Result section, but keep it in the Conclusions

With these changes made, and a thorough overall job of editing, this manuscript, with its important results, should be suitable for publication in AMT. I look forward to a revised manuscript.

Additional private note (visible to authors and reviewers only):

This manuscript has many technical lapses that should have been fixed prior to submission. I don't expect perfect English usage from a non-native speaker, but I do insist that the meaning of each sentence is clear, which is still not the case. I appreciate the moving of some of the more technical content to the SI, and the other changes made in response to the initial referee reports (and I thank them all for their detailed and thorough reviews). I strongly recommend that one of the more senior authors take a firm editorial hand with this manuscript and ensure that the intent of each sentence is clearly communicated. With this effort, the paper should soon be ready for publication in AMT.

There is some great information in this manuscript, and I look forward to a revision soon.