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Characterization and operation of a multi-channel Condensation Particle Counter (mc-CPC) for aircraft-based measurements, S. Richter et. al

Author comments to reviewer #2

The comments of the reviewer are depicted in black and italics.

Our answers to the reviewer comments are written in green color.

Changes in the revised version of the manuscript are given in red color.

Referee comment #2

We thank the reviewer a lot for the thorough reading and the valuable comments and questions, which will help to improve this manuscript. We especially appreciate the proposed changes to the mc-CPC design that will increase the performance.

Excellent work in characterizing and correcting the performance of the CPC during non-ideal cases, such as unknown/uncontrolled sample flow and varying external and instrument pressures. There's a lot of normalization that is happening that probably warrants a summation of error bars to understand the compounding corrections applied to the field dataset.

The summation of the error bars including the uncertainties of the normalization factors is given in Appendix C.

The equation determining NPF is maybe overly strict, but also seems arbitrary as it's currently written. Considering this is the major focus of the instrument, I think this needs better justification or characterization. I think the combination of the aforementioned error analyses and Poisson variance would provide a stronger basis for quantifying NPF.

See below for our reply, why we think that using our fairly conservative NPF criteria is preferable when taking also systematic uncertainties into account. We are trying to reduce these uncertainties for future aircraft measurements, and then we will consider to switch to the Poisson statistics criteria for determining the presence of NPF events.

Without the inlet pressure control being appropriately designed and characterized, the instrument may not accurately indicate the total particle number concentration, but is sufficient for NPF quantification since common-mode errors of larger particle loss is subtracted. This can be easily remedied in the next instrument update, to improve the amount of information received. Similarly, the second channel can be put to better use gathering additional information instead of redundant with the first channel.

We fully agree. We are eager to a) reduce the inlet losses by using a properly designed pressure control unit, b) obtain more information on the aerosol size distribution (from other instruments running in parallel) and therefore being able to determine the size-dependent losses, and c) changing the cut-off settings for the redundant channel. The redundancy for channels 1 and 2 was only intended for the initial operation to gain confidence about the reliability of the absolute measurements.

L020: Why was flight 4 selected when it was operated at a different pressure from typical?

We decided to show this flight in particular because we think that it is most suitable as a proof of concept. Our main goal was to present a flight that includes a distinct NPF event, which was only observed in a subset of flights. Furthermore, we wanted to show a flight that also includes non-ideal measurement conditions (flow fluctuations) to gain a better understanding of the data quality. Here, Flight 4 was the best choice although the pressure setting was indeed not the typical 250 hPa.

L089: Suggest changing "gets mixed" to "mixes" for succinctness.

Thanks, we changed it to "mixes" as suggested.

L090: Suggest changing "enable" to "causing" or "activating", showing causality.

We changed "enable" to "activating".

L101: Critical orifice is not shown in Figure 1 flow diagram. Is it within the Channel block? What is the role of the pump valve if not for flow control?

Yes, the critical orifice that is mentioned in this paragraph is indeed located inside the channel block. The pump valve represents a safety measure and can be either open or close. As it is located outside the channel block it is also shown like this in Figure 1. We added the following to line 106 of the revised manuscript:

(…) which is located downstream of the detection cell inside the channel block.

L221: Correcting for inlet orifice particle loss isn't necessary if only subtracting channels for an NPF measurement, but it is critical if trying to observe total particle concentration, especially in the lower Stratosphere, where mode size is typically accumulation mode. During ACCLIP, the integrated UHSAS concentration often exceeded the NMASS concentration in the Stratosphere due to losses in the pressure control orifice. Losses can be reduced with an appropriately designed expansion section after the orifice.

We agree that the absolute ambient particle number concentrations cannot be derived from our measurements, only lower limits. This is something we plan to improve for future flights and therefore we are grateful for your suggestions about including an appropriately designed expansion section after the orifice. To make this clearer we added the following to section 5, line 667 of the revised version:

As we could not perform a quantitative particle loss correction because of the unknown size distribution, the measured concentrations represent lower limits of the ambient aerosol concentration. Nevertheless, as all channels are subject to similar particle losses due to their common inlet, the identification of NPF events should not be affected strongly. Furthermore, the general concentration range and relative trends of the total concentration are well represented by the measurements.

L241: Eqn. 2 values seem arbitrary. Some of the cited papers use different values for uncertainty. Recommend testing analysis from Williamson et al., 2019 with your dataset based on Poisson counting statistics.

Thanks a lot for the suggestion. We reproduced the formula you suggested and determined the NPF criteria for our data with the Poisson counting statistic. When comparing channel 1 and 3 we typically get σ_{diff} in the range of 100-200 #/cm^3. For our data this means that a large part of the research flight would have been classified as an NPF event, although the difference between both channels could have been caused by systematic flow differences. We believe that the Poisson approach is feasible for very robust data sets that are hardly affected by systematic errors. For the mc-CPC this is unfortunately currently not the case as the data quality was influenced by the flow fluctuations in our system. Even the channels with the same ΔT are subject to some deviations (see Fig. 10). The systematic variations resulting from instrument uncertainties could falsely indicate NPF events. Therefore we would rather keep the more conservative approach. We decided to use the 30% as an NPF threshold because the uncertainty of the individual CPCs was determined to a maximum value of 22% (Line 241). By setting the NPF criteria to 30% we wanted to avoid misclassification. If we can be confident that systematic errors like a drift of the flows is excluded, then we will change to the Poisson approach in the future.

L328: What was the DMA sample flow? Relevant to understanding transfer function width (and thus horizontal error bars in the sizing).

The DMA sample flow was 0.7 lpm and this was added to line 339.

Here we need to clarify that the DMA bandwidth error at FZJ was determined with a sample flow of 1 lpm. We changed this accordingly in the revised manuscript.

Fig 4: Can you clarify how the error bars are calculated? Is it the standard deviation from the varying particle number concentration measured during the time period of each test point? Is the standard deviation of the TSI reference CPC considered, since it will fluctuate as well. In Poisson statistics, need to sum variances together.

We calculated the efficiency error by using the standard deviation of the respective mc-CPC channel and the TSI reference instrument and combining these two errors by error propagation.

$$\frac{\Delta(eff)}{eff} = \sqrt{\left(\frac{\sigma_{mcCPC}}{N_{mcCPC}}\right)^2 + \left(\frac{\sigma_{TSI}}{N_{TSI}}\right)^2}$$

After checking all figures and the error bars once more, we noticed a minor inaccuracy of the depicted figure and in the text. We thus have updated Fig. 4 and also corrected the error bars using the above mentioned formula. We also added a part in the description of Fig. 4:

The error bars in a) represent the combined uncertainty of the counting efficiency, derived from the standard deviations of the two aerosol instruments.

L472: This would not be a factor for sub-100 nm particles if an expansion section was included.

Thanks a lot for this suggestion. The mc-CPC could really benefit from an expansion section after the pressure-reducing orifice. We are planning to realize such an improvement in the future.

Fig 7: I think it would be nice to see the effect that pressure has on the plateau efficiency, rather than the normalized plots.

This is a very valid point, as from Fig. 7 one cannot directly see the decrease in counting efficiency with increasing dp ($p_{external}$ - p_{CPC}). Still, in Table 4 these changes are summarized and the raw data can also be found in Appendix F Figure F1 in the revised version. As we decided to only show the normalized data for consistency, we would rather keep this form of presentation.

Table 4: Is Ch3 operating temperature a typo, or did you operate Ch3 at dT=36C? If so, why did you calibrate the counting efficiency outside of its operating spec? And why is D50 of CPC3 relatively high for the same dT?

This is indeed a typo. Ch3 was operated at a ΔT of 15°C throughout all measurements. Therefore we changed it in the table from "ΔT = 36°C" to "ΔT = 15°C".

Fig 9: How can you have N11-N15 ~= 1000 /scm3 and consider it "no NPF"? This circles back to my comments on Eqn. 2. The whole section from 12:30-13:45 looks strongly related to NPF.

Yes, we also consider that the deviation between N11-N15 of 1000 scm^{-3} arises likely from NPF. This was stated in the paper as follows: "However, in the second part of the flight, the differences between the channels increase significantly, and the NPF criteria indicates NPF events". But we agree, that it is better to indicate the exact time, when we assume NPF to occur. Therefore we have changed the sentence and also added time stamps for the short NPF events:

Line 673: (…) was only fulfilled for a few seconds (e.g. 11:33).

Line 675: However, in the second part of the flight and especially in the time from 12:30 to 13:45, the differences between the channels increase significantly, and the NPF criteria indicates NPF events.

The NPF criteria shown in Fig. 9c was updated as we had used channel 2 and not channel 1 as stated in the description. We updated the plot accordingly and implemented the new figure into the revised manuscript.

L627: Is the inlet pressure reduction orifice properly sized for 200 hPa operation? Maybe you can go smaller and switch at a higher altitude to reduce amount of bypass flow the pump has to accommodate.

This is a very good suggestion that we will consider it for future campaigns. For this instrument we had a switchable orifice, but the second one was even larger than the one we used.

L641: The diffusion losses of 10-13 nm can be corrected since you have that information from subtracting the two channels and their relative contribution to the total concentration.

When we assume that the average size of the N_{11-15} channel is 13 nm, then the diffusion losses can be calculated to 22%. We state this loss in the caption of Fig. 9 now, but we did not apply this as a correction factor in order to keep the N_{11-15} data directly comparable with the N_{11} and N_{15} data.

Instrument suggestions for future development: Designing and characterizing an appropriate expansion chamber at the inlet pressure control orifice will allow the first channel to (closer) measure a total number concentration. Now that the first and second channel have been tested *together, I think the first channel alone can be trusted, so the second channel can be used for additional information such as a third cut size or a non-volatile measurement (if inlet orifice is characterized through Accumulation mode).*

Thanks again for the very helpful remarks. We greatly appreciate your comments and suggestions for the instrument improvement. For the next adjustment of the mc-CPC we will consider all these ideas.

We added a sentence to the conclusion:

Line 727: Another aim is to use all three channels at different cutoffs to gain more information about the air masses and potential NPF events.