

## Referee #1:

The present paper focuses on improving the counting statistics of sub-10 nm aerosol particles using a DMPS system with a modified Airmodus A20 CPC. They further found that improving counting statistics significantly reduces the uncertainty with the estimation of new particle formation and growth rates. I found it very important and interesting for aerosol size distribution measurements. The manuscript was well-written and structured. I would suggest publishing it on AMT after a minor revision.

We thank the Referee for their thoughtful comments which helped to improve the manuscript. Please find our detailed responses below in blue color and changes to the manuscript in red.

Specific comments:

1) Line 92. Why do you choose 2.5 lpm? Since a higher inlet flow rate indicates higher counting statistics, can you increase even higher?

The referee is indeed correct, that higher flow rates than 2.5 lpm would provide even better counting statistics. However, increasing the flow rate results in two challenges. First, the necessary supersaturation for activation of small particles becomes more difficult as the inlet air might not be fully saturated within the current design of the A20. Kangasluoma et al. (2015) indeed showed that the activation efficiency starts to decrease already at these flow rates. Further optimization of the CPC design would therefore be required to achieve even higher flow rates at similar activation properties. Second, higher flow rates would also require higher sheath flow rates in the DMA to maintain the same resolution. While DMAs with much higher sheath flow rates exist, they often provide access only to much more limited size-range as the voltage to diameter relation depends on the absolute value of the sheath flow. With voltages being limited to the order of 10 kV to avoid arcing, this sets natural limits to DMAs with high sheath flow rates, good resolution, and a broad accessible size-range, which in turn limits the available flow rates for a detector downstream of the DMA.

We tried to clarify these two points by adding the following sentence on line 92:

“While higher detector flow rates would result in even better counting statistics, it would require adjustments in the CPC design to achieve similar particle activation due to lower supersaturations and would also result in a lower size resolution for the DMPS system if the sheath flow rate remains constant (higher sheath flow rates would in turn reduce the dynamic size-range of the DMPS).”

2) Section 2.4 uncertainty in CPC measurements. As so many uncertainties are calculated (e.g., counting, measurement, total...) and discussed later in the results section, it is sometimes difficult to follow for readers. I suggest making a summary table listing all uncertainties, including the formula, use of purpose and values for exemplified experiments (e.g., 28th March, 5th and 6th May).

We agree with the referee that an overview of the results for the three measurement days would be beneficial, and we added the requested Table to the manuscript in the results section.

3) Line 142-143 and Figure 2. Why do you choose these certain number ranges?

We agree with the referee that this deserves a more detailed explanation. We choose to use a count interval such that it assures good balance between a narrow enough interval (keeping the relative error of the width below 5 % and enough counts per bin such that the resulting count distribution of the TSI is fitted well. To explain this we added the following sentence to the manuscript:

“This approach of choosing finite count intervals from the Airmodus A20 data instead of just using a single count value is due to the otherwise limited statistics which would not allow for solid fits of the corresponding count distributions of the TSI 3776.”

and later we added:

“(…) and the finite width of selected counts in the interval range (with the relative error due to this kept below 5% by our interval selection  $N_2=1.05 \cdot N_1$ )”

Line 170, ...fits sigmoidal functions to the rise of the measured signal.. of which parameter (number concentration)?

Yes, the sigmoidal fits can be applied even to the raw number concentration as they are independent of the absolute magnitude (see Lehtipalo et al., 2014). We clarified this at this point and removed it from the sentence on line 189-191:

“(…) fits sigmoidal functions to the rise of the measured raw number concentration (the approach is independent of the absolute magnitude of the signal and hence the inversion procedure, see Lehtipalo et al., 2014) in each size channel separately.”

“The generated input data (counts) were used to directly calculate  $GR_{3-6}$  and  $GR_{6-10}$  as the appearance time method can be performed on the raw signal ~~and is independent from any inversion procedure (Lehtipalo et al., 2014).~~”

5) Line 180-182. What does CS mean in equation (8)? Give some details on how to calculate  $GR_{3-6}$ . It would be helpful to have exemplified fitting plots to derive GR and J3 for non-NPF background readers.

We thank the reviewer for pointing out that lack of definition here. CS is condensation sink and the abbreviation is now introduced in the text such that it can be understood in the equation. For the detailed explanations of GR and J, we are convinced that Section 2.5 provides the necessary details, and the exemplified plots are already given in Fig. 6a-c.

6) Line 186. Why do you choose 28 March as an example?

We agree that this deserves a short explanation. We chose the 28<sup>th</sup> March because it is classified as a strong NPF event with good data coverage and an average growth rate such that the nucleation mode persists for a significant time. We therefore added the following to the manuscript:

“The 28<sup>th</sup> March is chosen as the example day as it is a typical class-1 NPF event day with a strong nucleation rate, but not much higher than average GR, such that the nucleation mode persists over long enough time in the sub-10 nm range to investigate the effect of improved counting statistics in full detail.”

7) Line 187-188. By altering the measured counts in each size-channel for each measurement time according to their underlying uncertainties. Plot out the time series of the uncertainties or give numbers (e.g., avg. +/- std)

We agree with the reviewer that it would be nice to see the variation of the uncertainty in each channel over the course of a day and hence added a new Figure to the Supplement, which shows the relative uncertainty as a surface plot for each diameter and its evolution during the 28<sup>th</sup> of March for all three simulation cases (Airmodus A20 counting uncertainty only, TSI 3776 counting uncertainty only and TSI 3776 total uncertainty). We added the following sentence at the end of Section 2.5 to the manuscript:

“The relative uncertainties for each size-distribution evolution measurement (in time and size) used as input for all three Monte Carlo simulations are shown in Fig. S2 in the Supplement.”

8) Line 222-224. How much does the chemical composition influence the cut-offs? Give a number if available.

Wlasits et al. (2020) showed that the variation of the  $d_{50}$  cutoff diameter between different seed materials for the unmodified Airmodus A20 is up to 4 nm, and up to 1.5 nm for the TSI 3776. We thus modified the text as follows:

“(…), which can be more than 3 nm difference for the  $d_{50}$  cutoff diameter between different seed materials for the unmodified Airmodus A20 (and only 1.2 nm maximum variation for the TSI 3776) (Wlasits et al., 2020).”

9) As shown in figure 4, what is the meaning of the numbers: 0.94-70.0 and 0.89-14.0?

These numbers represent the slope and y-axis offset of the fit, but the reviewer is correct that this should be written differently to be understandable. We further added an explanatory sentence to the Figure caption.

10) Line 236-237. Why the formation rate is more robust even though the used GR3-6 is less consistent between the A20 and TSI 3776? It would make sense if the GR term is not the dominant term. But in lines 268-269, the author demonstrated that the dominant term for formation rate is the growth term.

It is indeed an interesting remark and the reason for this is not entirely clear. The J calculation seems to have some buffering against the GR fluctuations which either comes through the less dominant terms (CoagS,  $dN/dt$ ) or the number concentration  $N_{3-6}$  which also needs to be included in the growth term. However, our findings are somewhat consistent in that sense, as also in the MC simulations a 16% relative uncertainty on the  $GR_{3-6}$  value only translated into a 13% relative uncertainty on the  $J_3$ . N, GR and CS are all highly linked quantities. Fig. 5a shows that  $GR_{3-6}$  derived by the A20 is often faster when above  $3 \text{ nm h}^{-1}$  (where the GR term in the formation rate calculation is more dominant), but at the same time Fig. 4 shows that number concentrations measured by the A20 are slightly lower. This might be one reason why  $GR \cdot N_{3-6}$  and hence  $J_3$  (dominated by  $GR \cdot N_{3-6}$ ) fluctuates less between the two instruments. We added that little speculation to the text.

“However, as shown in Fig. 4, the modified Airmodus A20 measured slightly lower concentrations compared to the TSI 3776, while  $GR_{3-6}$  was measured higher by the Airmodus A20 for values above  $3 \text{ nm h}^{-1}$ . Therefore, in these cases with a high growth term ( $\frac{GR}{\Delta dp} N_{dp}$ ) possibly dominating the formation rate calculations due a fast growth rate ( $>3 \text{ nm h}^{-1}$ ), the lower  $N_{3-6}$  might compensate for the higher  $GR_{3-6}$  reducing the fluctuations between the two instruments. In addition, the other terms ( $\frac{dN_{dp}}{dt}$  and  $CoagS_{dp} N_{dp}$ ) in Eq. (7) might also buffer the higher GR due to lower  $N_{3-6}$  values in that case.”

11) Line 241-242, Figure 6 is not well described and explained, add more details if you think it is important otherwise delete it. In Figure 6 (c), formation rate for A20 or TSI 3776? In Fig.6 df, why the distributions of A20 are always narrower than TSI?

We understand the reviewer’s confusion here and apologize that our references to that Figure were wrong. Fig. 6 is indeed already described, but it was referenced as Figure 7. We adjusted that. We also added the information that we used the 3776 for Fig. 6c. In Fig. 6 d and f, the distributions of the A20 are narrower than those of the TSI due to the better counting statistics. This was already explained in the text.

12) Line 253, please clarify how you derive the statistical uncertainty, refer to the table in comment 2.

We added the information to the Figure caption.

13) Figure 7 (a), any explanations on why is the distribution of GR3-6 bi-modal?

As already mentioned in line 280 we attribute the bimodal distribution to problems in the automated fitting. The bimodal distribution could perhaps be reduced requiring strict goodness of fit conditions for an MC result to be accepted, but it is beyond the scope of this work to find an extremely robust automated GR fitting method.

14) Lines 277-281, please clarify the relative uncertainties, refer to the table in comment 2.

As requested by the reviewer, we added the new Table 1 to the manuscript.

15) Out of curiosity, is it possible to compare A20 with NAIS as NAIS is good with nano-particles down to 0.8nm?

Kangasluoma et al. (2020) showed that NAIS in particle mode (which only extends to down to 2.5 nm) still shows significant discrepancies in total concentration measurements compared to other instruments in the sub-10 nm range. It is therefore not ideally suited to calculate formation rates. Growth rates derived from NAIS have been compared extensively with other approaches (e.g. Gonzalez-Carracedo et al., 2022, ACP) showing similar scatter as the two DMPS approaches. As the NAIS measures current and not particle counts, its underlying uncertainty treatment needs to be different and is out of scope of the manuscript.

