Response to Comments by Anonymous Referee #1

This manuscript by Lampilahti et al. presents a new method to calculate the particle growth rate (GR) during new particle formation (NPF). The particle GR is a key metric to understand the gas-to-particle conversion during NPF and the calculation from the particle number size distribution (PNSD) is still a considerable source of uncertainty when mechanistic insights into NPF are inferred from ambient aerosol data. This has two reasons: 1) considerable instrumental uncertainties in the sub-10 nm size range 2) the usage of GR calculation methods which highly depend on the user or are sensitive to the measurement uncertainties. In that sense, this manuscript presents a considerable step forward for the community working on analyzing NPF as no fully automated method to infer the GR from the PNSD has been presented so far. The introduced maximum cross-calculation method is shown to be robust compared to the other methods when a comparing the results for Hyytiälä Finland. However, the manuscript in its current form has some shortcomings, which should be addressed such that this paper encourages the involved community to use that new method in the future and therefore I can only recommend publication in AR after the following points have been addressed:

We thank the reviewer for the comments. Please see our responses below in blue text color.

Major comments:

The authors show in this manuscript that the new maximum cross-correlation method performs similar (providing the same medians and variance) to other methods when applied to a dataset from Hyytiälä, Finland. What is missing from this manuscript (and what should be part of an introduction into any new method) is the testing of its performance against data where the true GR is known! The central point of this manuscript is not only to show that the new approach reproduces probably the same errors than the other methods have but to show that it probably can also retrieve the actual GR of an NPF event. Inclusion of a synthesized dataset (with instrumental uncertainties imposed) and the application of the new method to it should be the number 1 point of the results. I am 100% confident,

that this group of authors has access to such synthesized NPF event data and it is therefore not too much additional work to include.

The second major point relates to the fact that the authors analyze 14 years of Hyytiälä data, but then do not make use of the already existing analyses of these datasets. As far as I know, already analyzed Hyytiälä NPF event classification and GR data should be available to that set of authors for the same 14 years they now present as being analyzed by the maximum cross-correlation method. It is unclear to my why Figure 5, only contains randomly selected days where the GR is re-analyzed with the maximum concentration method, while there should be the entire GR dataset for 14 years be available. There are two specific things I'd like to see in a revised manuscript: 1) Correlation plots of the full 14-year dataset for all instruments (ion-GR, particle-GR NAIS, particle-GR DMPS) between all GRs calculated from the maximum cross-correlation method and GRs calculated from other methods, whenever both methods returned a value. 2) The background/signal differentiation histograms when only data from manually classified NPF events (or use the ranking method or whatever) are used. Does the background population completely vanish if we only use values from "classified" NPF events (should be Appendix Figure).

We made major revisions to the results section with a focus on validating the method.

1) We investigated the sensitivity of the method in response to varying input parameters (normalization*, number of bins, maximum allowed time lag, smoothing window width) (Figures 2 and 3).
2) We simulated a NPF event with known GR and compared the performance of our method and the conventional methods (Figure 4)
3) We compared our results from Hyytiälä against a pre-existing GR dataset (2010-2019) (Figures 7 and 8)
4) We included a low nanorank vs high nanorank comparison to the GR distribution plot (Figure 5). At high nanorank values the background was reduced.
5) We compared the days with valid GRs against NPF event classification and nanoranking (Figure 9)

From the previous version Figures 5, 6 and 7 were now not included as they did not contribute so much to the method's validation.

*We added another input parameter (gamma) which controls the degree of overlap correction when the particle concentration time series are cross-correlated. We selected the gamma based on the condition that the GR should not change if the size range is divided into smaller and smaller bins.

Minor comments:

Line 35-42: Stolzenburg et al., 2023, Rev. Mod. Phsy. is the most recent and complete review on particle growth and also compares different methods with each other. It should be included here.

We included the reference. We also found the terminology used in the article useful (e.g. representative diameter, representative time) so we adopted some of the terms in our manuscript.

Line 37: In the above mentioned review, there is a long discussion about a third approach using the full evolution of the PNSD combined with the general dynamics equations. These methods do disentangle the different contributions to GR but suffer from other challenges. They should be mentioned here, because especially those methods would have the chance to also be fully automated (in an ideal world).

We added a discussion on dynamic methods to the introduction.

Line 63: What is the difference between these methods to the one presented in Lehtipalo et al., 2014. This should be clarified here.

We added the following clarification: "The difference between finding representative time lags and representative times is subtle but relevant since in practice the methodology can be different and one may be more amenable to automation than the other."

Line 67: I would also add that one of the advantages of the size channel based methods is that they do not require perfect knowledge on the absolute concentrations of the PNSD (i.e. the inversion correctness). In fact, they sometimes can even be run on raw data. The same applies to the new maximum cross-correlation method and should be mentioned here.

We added this to the advantages

Line 100-101: I'd prefer $\bar{N}_1$ (with bar above) for the mean notation, as this is far more common.

There seems to be a bug in the pdf conversion, the bar should be above. We changed the symbol to Greek letter mu to avoid trouble.

Line 109: To judge the robustness of the method, it would also be interesting how the results change when a different averaging is applied. Especially if the method is transferred to other environments, this parameter might need to change. In addition, it is known that rolling averages can skew the results of the appearance time method, so it would be interesting to see what changes when a static, time resolution reduction (i.e. block averages) is used.

Sensitivity test to the smoothing window width was added (see summary of major revisions). Adding more smoothing does skew the results towards lower GRs.

Line 123-131: It is not fully clear to me how the approach in creating more size increments is facilitated. Is the PNSD first inverted for all data and then somehow resized? I.e. how many original channels are e.g. in that window between 2-3 nm and how many increments are later used? I.e. do you obtain a higher size-resolution than the original data with that approach?

In our version of the algorithm the size range is divided into n equally sized bins on log scale. The concentrations are interpolated in linear piecewise manner to the diameters that divide the bins.

Having more bins than there are size channels in the size range will result in tau_max=0 evaluations and no GR output. We added to the Methods section 2.1 in the revised version that one should not exceed the number of size channels when dividing into bins. The log distance between size channels in the NAIS data is at most 0.03 and in the DMPS data 0.05. So between 2-3 nm there are 6 size channels (we divide it into 2 bins).

Line 133: In my opinion the method should be called maximum cross-correlation method throughout the manuscript as maximum correlation method could be misleading/ doesn't describe exactly what the method does.

We changed the naming and abbreviated it as MCC in the revised text.

Line 147-149: That sentence seems to be broken. "To ensure" what?

Removed "to ensure"

Line 149-151: Again, how many original size channels are in these ranges?

Number of channels between 2-3 nm in NAIS: 6
Number of channels between 3-7 nm in NAIS: 12
Number of channels between 7-20 nm in NAIS: 15
Number of channels between 3-7 nm in DMPS: 7
Number of channels between 7-20 nm in DMPS: 9

We added this information to the revised manuscript section 2.2 describing Hyytiälä dataset

Line 173-178 and 183-184: As said in the major comment: Here should be a comparison to the classical NPF event day characterization.

See the summary of major revisions above

Line 192-194: Probably the right argument. But assuming these fast GRs are really there, would it make sense to then probably just tolerate e.g. one of the tau_max,i to be below zero to still capture some of these?

With the new normalization using the exponent gamma=0.25 the shift in the GR distribution is less pronounced. Some of the shift is from bias towards smaller GRs caused by the previous normalization where gamma=1 (see revised Figure 2). The bias accumulates when the size range is divided into more bins. We tried accepting some invalid tau_max,i and it did indeed bring some extra days, but the difference was so small that in our opinion it did not justify making it the default option.

Line 211-214: As the new method can calculate GR on more days, it would be very interesting to see how this corresponds to a event classification scheme (major comment 2)

See above

Line 216-218, Figure 5: It is important to see this comparison across different instruments and also probably different methods. Moreover, as said in major comment 2, the authors should make use of the full Hyytiälä datasets available to them.

See outline of the major revisions above. The instruments in the pre-existing GR dataset matched the ones we used (NAIS ion mode and DMPS). The method in the pre-existing dataset was maximum concentration, unfortunately no other methods were used.

Line 240-241: Again Stolzenburg et al. (2023) probably provides the to-date most complete overview of GR datasets and should be referenced here.

Citation added, see below

Line 245-250: Feels a bit off here, as this more or less is Figure 8, but Figure 6 is not yet discussed here. Should that be later in the manuscript?

The discussion on median GRs and their size dependent behavior is now in text where we discuss Figure 5. We only describe the NAIS total particles shortly. This is the modified paragraph:

"The median GRs in Figure 5 showed an increasing trend with particle size, a pattern commonly observed across various environments (Kerminen et al., 2018; Stolzenburg et al., 2023). In the same size range the median GRs found for the positive ions, negative ions and total particles (from DMPS) were similar, which is also supported by previous observations (Gonzalez Carracedo et al., 2022; Hirsikko et al., 2005; Manninen et al., 2009; Yli-Juuti et al., 2011). The GRs calculated from the NAIS total particles were higher compared to the other GRs in the same size range. This might be linked to the NAIS measuring higher concentrations in total particle mode compared to other instruments (Kangasluoma et al., 2020)."

Line 253-255, Figure 6: My honest opinion: This Figure is not very interesting as it doesn't provide any new insights (except that the new method can perform across different seasons). If the authors want to save space as they need to include the comparisons with synthesized data or the full Hyytiälä dataset, they can remove it. In addition, the caption is too short and should explain more what is in that Figure.

This figure was removed

Line 260-268, Figure 7: I would love to see the median days also for the "edges" of the GR distribution, i.e. the very fast and very slow growth cases, as these might be the most interesting, where deviations from the classical "banana-type" picture might appear.

We found this figure to be too much on the side of interesting results than method validation. Therefore to keep the manuscript coherent we decided to leave it out.

Line 296-297: Why only ion GR from Gonzalez-Carracedo? The DMA train data are especially useful in the sub 3 nm range, where other instruments often perform worse. This comparison should be shown, as this is one of the cases where the method might reach its limits (I don't think so, but it needs to be included).

We did not calculate the GR from the DMA train data using our method in the sub-3 nm size range. We only used NAIS ions, therefore in order to compare "apples to apples" we only included the NAIS ion data from Gonzalez-Carracedo in the comparison.