

Atmos. Chem. Phys. Discuss., referee comment RC2 https://doi.org/10.5194/acp-2021-99-RC2, 2021 © Author(s) 2021. This work is distributed under the Creative Commons Attribution 4.0 License.

Comment on acp-2021-99

Anonymous Referee #2

Referee comment on "Aerosol formation and growth rates from chamber experiments using Kalman smoothing" by Matthew Ozon et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-99-RC2, 2021

The manuscript by Ozon et al. describes the application of a new theoretical method (FIKS, Fixed Interval Kalman Smoother) for deriving new particle formation and growth rates from size distribution measurements. The manuscript that introduces the theoretical framework of the applied method is currently under review (Ozon et al., 2020). In the present paper the FIKS method is applied to artificially generated size distribution data and data from the CLOUD experiment for three different chemical systems (sulfuric acid + ammonia, highly-oxygenated organic compounds and iodic acid). The data used for FIKS are taken from the DMA-train, which provides time-resolved particle concentrations for seven different size channels (between 1.8 nm and 8 nm, see Stolzenburg et al., 2017). From these data the FIKS method yields the new particle formation rate (J at 1.7 nm) as a function of time and the particle growth rate (GR) as a function of the particle size. These results are compared with the J and GR derived from other (established) methods. For the derivation of the new particle formation rate the particle number concentrations measured with the Particle Size Magnifier and the Scanning Mobility Particle Sizer are used (Dada et al., 2020), whereas for the derivation of the growth rate the INSIDE method with the DMAtrain data are used (Pichelstorfer et al., 2018). Overall, the inter-comparison between the results from FIKS and the other methods show good agreement. The FIKS method has the benefit of providing an uncertainty range for the derived quantities. Ozon et al., further demonstrate that the method can be used to optimize the settings of the DMA-train in terms of the set size resolution such that size distribution can be reconstructed by the proposed method with high accuracy. This is very important as such a guideline can improve the data quality in further experiments.

Overall, I agree with the authors that the development of sophisticated data evaluation methods as the one presented here, is important and lacks somewhat behind the instrument development. Therefore, I highly favor the publication of the present study and have only a few suggestions for further improvement and clarification (listed below). What I find, however, somewhat problematic is the fact that the method paper (Ozon et al., 2020) is not finally published yet. In this respect, I would also like to mention that I did not review the method itself but only its application in the present study. Therefore, I

think that it would be appropriate to wait for the final publication of Ozon et al. (2020) before the present manuscript can be published in ACP.

Specific comments:

 Page 1, line 33/34: The authors mention here that "potentially crude approximations" can be made when formation and growth rates are derived. It would be good to explain what approximations are meant here.

Page 2, line 72/73: I think, the data from the DMA-train can also be used to derive new
particle formation rates (from the smallest size channel) and that these data need to be
included in the further analysis and discussion. This could show whether the differences
in J from the PSM and the FIKS method arise mainly due to the use of data from two
different instruments or from the different methods.

Page 7, line 218: Here it is mentioned that the number of the size channels is 32. Some discussion should be included how this choice affects the outcome of the results. Can a larger number of size channels improve the agreement between the FIKS method and the other methods?

 Page 11, line 335: The discrepancy between J from the PSM and the FIKS method is a factor of ~2.5, i.e., it is significantly larger as for the sulfuric acid/ammonia and the iodic acid system. I would like to see some further discussion on the possible reasons for this difference. References:

Dada, L., Lehtipalo, K., Kontkanen, J., Nieminen, T., Baalbaki, R., Ahonen, L., Duplissy, J., Yan, C., Chu, B., Petäjä, T., Lehtinen, K., Kerminen, V.-M., Kulmala, M. and Kangasluoma, J.: Formation and growth of sub-3-nm aerosol particles in experimental chambers, Nat. Protoc., doi:10.1038/s41596-019-0274-z, 2020.

Ozon, M., Seppänen, A., Kaipio, J. P. and Lehtinen, K. E. J.: Retrieval of process rate parameters in the general dynamic equation for aerosols using Bayesian state estimation, Geosci. Model Dev. Discuss., 2020, 1–32, doi:10.5194/gmd-2020-236, 2020.

Pichelstorfer, L., Stolzenburg, D., Ortega, J., Karl, T., Kokkola, H., Laakso, A., Lehtinen, K. E. J., Smith, J. N., McMurry, P. H. and Winkler, P. M.: Resolving nanoparticle growth mechanisms from size- and time-dependent growth rate analysis, Atmos. Chem. Phys., 18(2), 1307–1323, doi:10.5194/acp-18-1307-2018, 2018.

Stolzenburg, D., Steiner, G. and Winkler, P. M.: A DMA-train for precision measurement of sub-10 nm aerosol dynamics, Atmos. Meas. Tech., 10(4), 1639–1651, doi:10.5194/am 634 t-10-1639-2017, 2017.