Reviewer replies: Reviewer #2

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 [2, 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 [4, 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 [1, Dada et al., 2020], whereas for the derivation of the growth rate the INSIDE method with the DMA-train data are used [3, 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 clari_cation (listed below). What I find, however, somewhat problematic is the fact that the method paper [2, 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 [2, Ozon et al. (2020)] before the present manuscript can be published in ACP.

We thank the reviewer for his/her thoughtful replies and are happy that he/she is convinced about the importance of this manuscript. We are happy to inform that the method paper has been published. Please see link: https://gmd.copernicus.org/articles/14/3715/2021/gmd-14-3715-2021.html.

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.

We removed the statement about potentially crude approximation, because “rather simple regression or balance equation approaches” is exactly what we mean.

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.

We have added the nucleation rates estimated with the DMA-train data and the traditional method for all three cases. In the sulfuric acid case and the iodic acid case, the nucleation rate values are quite similar to the two other approaches, supporting the claim that we should be quite confident about the estimations. The observation that in all test cases the direct DMA-train-based estimates and their credible intervals are somewhat in the middle of those given by FIKS and the PSM, hints that the differences between the FIKS and PSM estimates are caused by the use of different instruments and the differences in the estimation 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?

We agree that this needs a short discussion and added the following statement to the manuscript: "We use a resolution of 32 bins from 1.7 to 10 nm for the FIKS to keep the computational effort low. We tested also 16 to 64 size discretization bins, but higher resolution required additional adjustments in the size-correlation of the covariance given in Eq. (8), which would result in significant differences compared to the original work of Ozon et al. (2020) without providing significantly more accuracy."

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.

We agree with the reviewer that the organics experiment shows the largest discrepancies and this should be further clarified. There is a plausible reason for the deviations in the organic experiment compared to the other two: In the organic case, there are the lowest number concentrations in the chamber for all three experiments. These are already close to the limit of detection in the DMA-train and hence our uncertainties are not well described by the Gaussian assumptions inherent to the FIKS, but Poisson statistics are needed. Further, we know that our calibrations for organics both for the PSM and for the DMA-train are quite uncertain, and derived with different approaches. For the PSM we use the method of Dada et al. (2020) by comparing with the NAIS, while for the DMA-train we used a lab-based calibration with beta-caryophyllene particles (Wlasits et al., 2020) which are not exactly equal to the alpha-pinene particles. Hence, it is not surprising that there are some deviations caused by the instruments and not the methods, which can be seen from Figure 4b) where we also included the traditional nucleation rate approach using DMA-train data (see earlier comment). We thus added: "Also the formation rate retrieved with the method from Dada et al. (2020) but using the DMA-train data is significantly higher, but in-between the two estimates. The possible deviation has hence two plausible reasons: The instrumental differences can be caused by different calibration procedures for the DMA-train and PSM (Dada et al. (2020) direct cross-calibration using NAIS versus Wlasits et al., (2020) using beta-caryophyllene ozonolysis surrogates). And the methodological differences could arise from the very low counting statistics in the DMA-train during this experiment compared to the other two, which will cause the inherent Gaussian assumption of the FIKS to fail."