Review to “Regularized inversion of aerosol hygroscopic growth factor probability density function: Application to humidity-controlled fast integrated mobility spectrometer measurements”, Atmos. Meas. Tech., December 2021

The authors test of different inversion methods for aerosol hygroscopicity measurements with a specific focus on the humidity-controlled fast integrated mobility spectrometer. The manuscript is very well written, and concise. As inversion techniques other than least-square approaches have recently gained some attention in the community again (Petters, 2021; Sipkens et al., 2020a, 2020b; Stolzenburg et al., 2022), this manuscript certainly is another interesting approach focusing on an important problem, the retrieval of the GF-PDF. However, with respect to the above-mentioned literature, I have the feeling that the authors need to add additional work to their manuscript, such that it significantly adds to the body of knowledge and I can recommend publication in Atmos. Meas. Tech. I give my detailed suggestions below:

1) The authors need to challenge their inversion technique with more difficult data. Using the same forward and inverse model is considered to be not sufficient in testing an inversion approach, leading to unrealistically good outputs (e.g. Colton and Kress, 2013). The authors need to run tests on their inversion where the forward and inverse model are different (i.e. assuming that the calibration of the FIMS is not perfect) and where statistics other than the counting error influence the result and their actual distribution can only be guessed (e.g. the standard deviation of an additional Gaussian error is different in forward and inverse model). Both cases are closer to real measurements and this would demonstrate the performance of the inversion algorithms under more challenging conditions.
2) The authors should at least present an application of their algorithms to real measurement data, such that the reader can judge how big differences we would expect in the retrieval of the GF-PDF for typical hygroscopicity measurements in the ambient air.
3) *Data availability*. I think it is nowadays almost standard that the authors publish their code openly along with the corresponding manuscript (again referring to Petters, 2021; Sipkens et al., 2020b; Stolzenburg et al., 2022). If the authors want to ensure reuse of their methods this is highly encouraged.
4) I understand that the manuscript focuses on the inversion for the FIMS, but this instrument is not available to many researchers doing hygroscopicity studies. In my opinion, it would be highly beneficial if the authors could also test their inversion on classical TDMA data.

Apart from the above, I have only a couple of minor comments:

1) L.153 ff.: Please note that Tikhonov regularization (and LSQ) implicitly assume Gaussian statistics on the measurement noise, but that the underlying statistics used to generate the measurement data is of Poisson nature in this manuscript (see also comment above on imposing an additional Gaussian uncertainty to challenge the methods under more difficult conditions). You can refer to Stolzenburg et al. (2022) here who developed a Poisson approach for regularization.
2) L.159: Please be aware that the matrices described here impose specific boundary conditions (Donatelli and Reichel, 2014). I assume that Dirichlet boundary conditions have been used. Please specify this, as it is often neglected in inversion procedures that the boundary condition is quite important for the shape of the solution.
3) L.161 ff.: Which algorithm was used to locate the L-curve corner?
4) L.242: The discussion about the computing time is difficult, as it crucially relies on the used device. I think it is better to make Figure 2d with relative computing times.

5) L.250 ff.: The measure smoothness is defined in Eq. (14) by the 2[nd] order derivative, so it is not surprising to me, that the 2[nd] order Tikhonov regularization achieves the best smoothness measure, as it optimizes for the same quantity.
6) L. 253: It is somewhat expected that the absolute values of ξ increase with more modes. The more interesting question is rather why it is not so significant for the Twomey algorithm.

References:

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