Comment on amt-2021-334
Anonymous Referee #2

Referee comment on "Regularized inversion of aerosol hygroscopic growth factor probability density function: Application to humidity-controlled fast integrated mobility spectrometer measurements" by Jiaoshi Zhang et al., Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2021-334-RC2, 2021

The authors apply different inversion methods to invert the aerosol GR-PDF from the measured signals from synthetic HFIMS signals. They found that for the few test cases, Markowski-Towmey's method generally outperforms other methods. By doing this, they convincingly improved the data inversion of HFIMS data and promisingly HTDMA data, which were mainly based on predefined size distributions or least square methods. This well-written manuscript is easy to follow. I recommend it to be published in *Atmospheric Measurement Techniques*, However, a major revision is necessary to convincingly demonstrate that the data inversion of HFIMS (and HTDMA) is improved. I feel that the authors are too optimistic about the representativity of their limited synthetic data on real laboratory experiments and atmospheric measurements. Further, this manuscript will have a broader impact on the community if its outcomes (e.g., inversion codes) can be readily used for HTDMA measurements. My detailed comments are given below.

Major comments:
1. More tests and/or discussions are needed to provide supports for the argument that Towmey's method outperforms other tested inversion methods. The three test cases are perhaps sufficient to show that Towmey's method is better than least square methods because the least square methods are notorious for solving ill-conditioned problems. However, the reason why Towmey's method is better than the Tikhonov regularization methods needs more clarification and/or data to support.

2. The authors need to show the performance of Towmey's method with at least one dataset from either laboratory experiments or atmospheric measurements. Estimating the measurement uncertainties with only the counting uncertainties typically underestimates the total uncertainties. Despite this, I am not concerned about the applicability of Towmey's to real datasets and its better performance of the than least square methods.

Minor comments:
3. Lines 30 - lines 115. The working principles of HFIMS are well summarized. However, they can also be removed or shortened to make space for more tests and discussion, as long as the inversion problem (e.g., Eq. 4) is clearly proposed.

4. Line 116, Eq. 4. Please consider adding an error term (\(\epsilon\)) to Eq. 4 and other related
equations to emphasize that the main challenge of data inversion is to deal with the uncertainties. The least-square methods are supposed to work pretty well if there is no error in the inversion problem as presented in Eq. 4.

5. Line 125. "The integration can be written as......". I recommend replacing "written as" with "approximated by". Discretizing a continuous distribution is also a step of inversion and there are inversion algorithms using improved discretization methods (e.g., Hagen and Alofs, doi.org/10.1080/02786828308958650).

6. As far as I am concerned, Towmey's method does not mathematically guarantee convergence of the inversion results. Optimized adjusting factor(s) are usually needed to guarantee that convergence without a great sacrifice of the computational expense. As a result, the convergence of Towmey's method for one dataset (e.g., synthetic data) does not guarantee its convergence for other datasets (e.g., laboratory experiments and atmospheric measurements). I recommend the authors address this very briefly in the main text. Considering broader applications of the inversion methods to HTDMA studies, I recommend the authors address this very briefly in the main text.