Manuscript No.: egusphere-2026-848

Title: Technical Note: Deep-GF-PRM - A physics-informed deep learning framework for parameterizing aerosol hygroscopic growth factor probability density function

We thank the anonymous referees for their valuable and constructive comments/suggestions on our manuscript. We have revised the manuscript accordingly and please find our point-to-point responses below.

Comments by Anonymous Referee #3:

General Comments:

Huiping Lu et al. have published a nice pre-print of a technical note (egusphere-2026-848, "Technical Note: Deep-GF-PRM – A physics-informed deep learning framework for parameterizing aerosol hygroscopic growth factor probability density function") detailing their use of a neural-network-based machine learning algorithm to invert observations of aerosol hygroscopic growth factor distributions. The algorithm is compared to field data inverted using a Tikhonov regularization matrix. The paper is reasonably well-written and the presented algorithm reproduces the inverted data quite well. This paper is of interest to the community given the increased uptake of machine learning algorithms. The paper is also a good fit as a technical note of ACP or AMT.

After reading the paper I remain somewhat dubious that the fast nonparametric Tikhonov method, which does not require a great deal of computational power, will be inferior to the deep learning method. I recommend major revision to address this concern and to improve the clarity of the work. If the following points can be addressed by the authors I feel the paper would be suitable for publication.

<u>Responses</u>: We sincerely thank the reviewer for this incisive and constructive comment on our work. We have carefully addressed every comment and revised the manuscript accordingly. Point-by-point responses are detailed as follows, and all modifications are marked in the revised manuscript.

First, we would like to clarify that the non-parametric GF-PDF inversion method used in this study is the Twomey algorithm, not Tikhonov regularization. As mentioned in the manuscript, Zhang et al. (2022) systematically compared these two methods for GF-PDF retrieval and demonstrated that the Twomey algorithm yields lower inversion error. The Tikhonov regularization, on the other hand, depends on the regularization parameter, λ, which cannot be obtained reliably using existing methods in practice. We therefore selected Twomey inversion as the benchmark throughout this work.

Second, we did not claim that Deep-GF-PRM outperforms non-parametric inversion methods for accurate GF-PDF inversions. While both Twomey and Tikhonov inversion are computationally efficient for a single measurement, they have to process each sample individually and only yield a discrete GF-PDF spectrum without explicit modal information. To extract modal parameters from the discrete GF-PDF, additional parametric fittings are required, which are not only computationally inefficient but also introduce extra uncertainties, as demonstrated in the manuscript (Fig. 3f). In addition, this two-step workflow (inversion + fitting) becomes a practical bottleneck when processing large datasets. Deep-GF-PRM addresses this bottleneck through parallelized end-to-end prediction: it maps the instrument response directly to modal parameters, and processes multiple samples simultaneously via matrix operations. On a test set of ~27,000 samples, Deep-GF-PRM inference completes in ~0.1 seconds on GPU (~10 seconds on CPU), compared to several hours for the serial inversion + fitting workflow.

In addition, we have refined the Deep-GF-PRM model by restructuring the loss function, and the updated model performs better in GF-PDF MSE on the synthetic test set (from 0.187 to 0.09), and the agreement in GF_{mean} values improved correspondingly for both synthetic and field datasets.

MAJOR COMMENTS:

[1] The authors need to give serious consideration to the clarity of the writing. This applies at the paragraph level where machine learning terminology is used, and to the below comments [1a] and [1b].

[1a] The number of models described almost seems to exceed the number of models actually tested. Please consider including a table naming each model and providing a citation, brief description, what data the model is tested with, the figure showing the results, etc.

[1b] It would also be very helpful for the reader if the authors would include flowchart describing the methodology and comparisons between models.

Responses: We would like to clarify that the Twomey's method is utilized as benchmark of the non-parametric GF-PDF inversion in this study. As demonstrated in the authors' previous work (Zhang et al., 2022), Twomey's method outperforms the Tikhonov algorithm in retrieving the GF-PDF from HFIMS measurements. We note that Tikhonov regularization is described in the "Introduction" as part of the literature review of non-parametric inversion methods, but it is not used as a primary benchmark.

In this study, we proposed Deep-GF-PRM, a deep neural network that maps instrument responses directly to modal parameters. The Deep-GF-PRM model was trained on a large dataset of synthetic instrument responses generated using a wide range of GF-PDFs and noise levels, and three standard ML algorithms — Linear Regression (LR), K-Nearest Neighbor (KNN), and Random Forest (RF) — are trained on the same dataset and serve as benchmark comparisons of Deep-GF-PRM's performance. We also applied the Deep-GF-PRM model to real-world measurements, and compared with nonparametric inversions using Twomey's method.

We note that Fig. 1 (also adapted here as Fig. R6) in the manuscript already presents a schematic comparison of the two core workflows for inverting and parameterizing the GF-PDFs, i.e., the conventional two-step approach (non-parametric inversion + parametric fitting) vs. the proposed Deep-GF-PRM approach (end-to-end mapping).

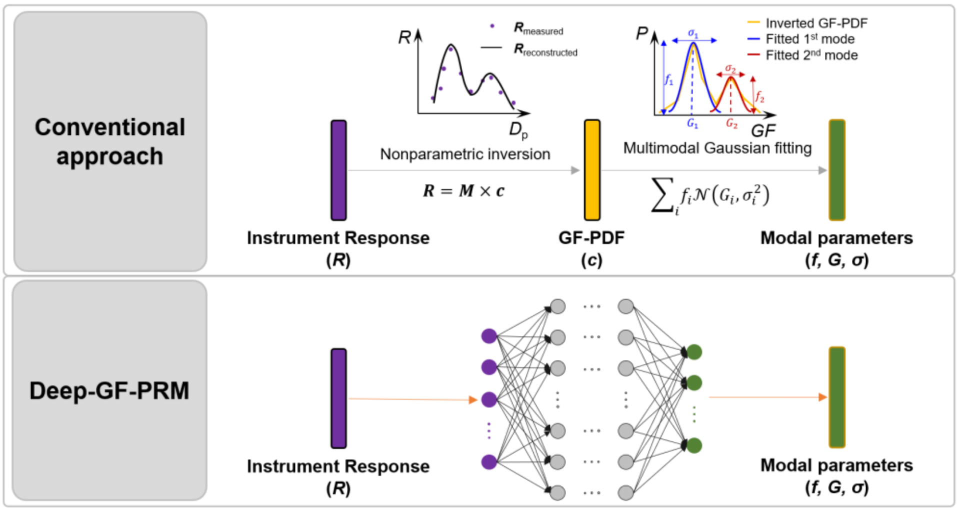

Fig.R1. Scheme of parameterization of aerosol hygroscopic growth factor probability density function (GF-PDF). The conventional approach, i.e., nonparametric inversion of the GF-PDF spectrum from the instrument response followed by a multimodal Gaussian fitting to retrieve the modal parameters. The Deep-GF-PRM approach, i.e., a deep learning model directly maps the instrumental responses to the GF-PDF modal parameters.

[2] Given that the Tikonov, the Gaussian, and the neural network inversion algorithms are compared in this work, and considering that the "ground truth" appears to be the Tikonov-inverted data, the Tikonov result should be included alongside all other models in Figure 4 and in other comparisons as well.

Responses: We thank the reviewer for pointing this out. First, we would like to clarify that for the synthetic test dataset, the "ground truth" is the known GF-PDF used to generate the instrument response, not the inverted GF-PDF using non-parametric inversion methods, such as Twomey's method or Tikhonov regularization. As demonstrated in Fig. 3 in the manuscript, we evaluated the performance of Deep-GF-PRM on the independent testing dataset and compared to the ground-truth GF-PDFs. The MSE, Wasserstein distance, and the first moment of the GF-PDF were used to evaluate the accuracy of the model predictions against the ground-truth. The inverted GF-PDFs using Twomey's method were also shown as references. For the field dataset, where no ground truth exists, we therefore directly compare the model predictions with the Twomey inversion results.

Regarding the inclusion of Tikhonov algorithm, following the reviewer's suggestion, we have added a direct comparison between Tikhonov regularization and Twomey's method on the synthetic test dataset, as shown in Fig. R7 below. The optimal regularization parameters were obtained using the generalized cross-validation (GCV) method, which has been proved to be effective by Petters (2021). As shown in Fig. R7, Tikhonov regularization yields systematically inferior performance than Twomey's method, across almost all different metrics, i.e., MSE, reconstruction error (χ^2), smoothness (smaller = better), and Wasserstein distance. The average GF-PDF MSE of Tikhonov solutions is approximately ~2-folds of that of Twomey's. These results are consistent with that concluded in Zhang et al. (2022). Therefore, we selected Twomey's method rather than Tikhonov, as the representative non-parametric benchmark throughout the paper.

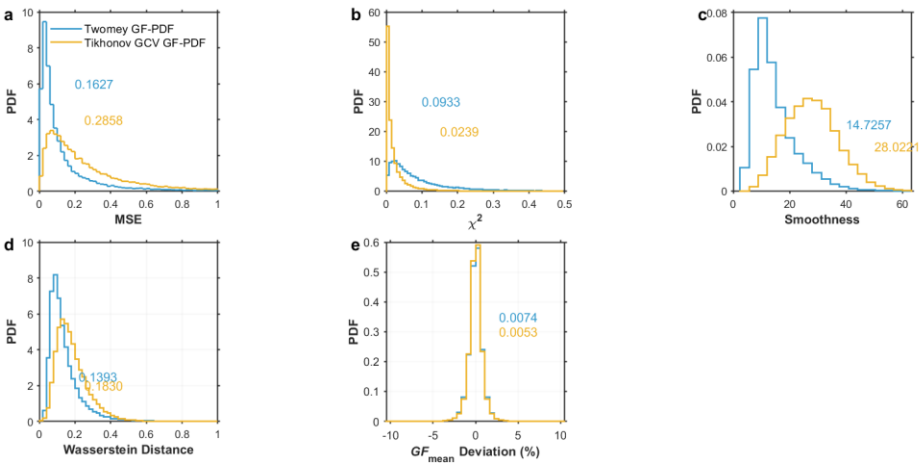

Fig. R7. Comparison of Tikhonov GCV regularization and Twomey iterative regularization for GF-PDF inversion on the synthetic test dataset, across GF-PDF MSE (a), reconstruction error (χ^2, b), smoothness (c), Wasserstein distance (d), and first-moment of GF-PDF (e).

[3] The authors have neglected to quantify the computational power needed to run each model, and they also do not quantify how the complexity of the computation scales with the number of points, for each model discussed. This information is important to assess the feasibility of implementing the model at scale and should be included alongside the model descriptions.

Responses: We thank the reviewer for this important suggestion. We only qualitatively mentioned that Deep-GF-PRM is faster than the conventional approach, without providing quantitative metrics. We conducted a quantitative runtime comparison on the testing dataset containing ~27,000 samples, for both the non-parametric inversion and the Deep-GF-PRM prediction. Note that the testing was conducted on a workstation equipped with an AMD Ryzen 5600 CPU and an NVIDIA GeForce RTX 4060 Ti (16 GB) GPU. The results demonstrate that the Twomey inversion took about ~1 hour CPU time (mode fitting not included), while the Deep-GF-PRM only took 10 seconds CPU time to finish all the predictions for 27,000 samples, and this time can be reduced to ~0.1 seconds if implemented on a GPU.

We clarified this in the revised manuscript, and it now reads:

"For example, while the conventional approach typically requires hours to perform GF-PDF inversion (mainly inversion matrix derivation) and mode fitting on the 27,000-sample testing dataset, the model predictions can be implemented instantaneously (~0.1 seconds on GPU and ~10 seconds on CPU). This efficiency makes Deep-GF-PRM particularly suitable for processing large-scale and long-term datasets.".

[4] The key question that the authors do not address is: what is the use case for the neural-network based calculation, given that the Tikhonov calculations are faster and apparently more accurate.

Responses: As we stated in Sect. 4 in the revised manuscript, Deep-GF-PRM is applicable to the rapid, parallelized processing of aerosol hygroscopic measurements from instruments such as HTDMA and HFIMS, directly predicting parameterized hygroscopic modal parameters from instrument responses. It has three main advantages: 1) Deep-GF-PRM achieves a good accuracy of recovering GF-PDF spectrum from instrument responses that matches conventional non-parametric inversion; 2) the direct mapping avoids the parametric fitting conventionally required to resolve GF-PDF modes, thereby eliminating associated fitting errors; 3) the parallelized architecture makes Deep-GF-PRM particularly efficient for unified, high-throughput processing of large-scale field datasets.

MINOR COMMENTS:

[5] Lines 38-40: what is the difference between HFIMS and WFIMS? Additional clarification is necessary here.

<u>Responses</u>: We have clarified in the revised manuscript, and it now reads: "A recently developed humidity-controlled fast integrated mobility spectrometer (HFIMS) drastically accelerates aerosol hygroscopic growth measurements by employing a water-based fast integrated mobility spectrometer (WFIMS) that captures the humidified particle size distribution at a time resolution of 1 s (Pinterich et al., 2017). As a result, HFIMS extends the size-resolved hygroscopic growth measurements to multiple RH levels (Pinterich et al., 2017; Zhang et al., 2021). "

[6] Line 159, 167: The number of hidden layers described in these two paragraphs is inconsistent. Why did the authors choose ten hidden layers (line 159)? He et al. (2016) chose 158 hidden layers.

<u>Responses</u>: We have corrected in the revised manuscript, and it now reads: "The Deep-GF-PRM framework, as schematized in Fig. 2, consists of an input layer, a series of hidden layers, and an output layer."

We note that the 152-layer ResNet proposed by He et al. (2016) was designed for high-dimensional image recognition tasks, which require extremely deep networks to capture complex spatial features. Our task is fundamentally different, i.e., a 1D regression problem with a 64-dimensional input and a 9-dimensional output. The model architecture, e.g., the number of hidden layers, number of neurons per layer, were determined through Bayesian hyperparameter optimization.

[7] lines 96-102 and equation 1: The omega's are not defined in the appropriate place. These should be directly below the equation and not in a subsequent paragraph.

<u>Responses</u>: We have moved the physical definitions of Ω_1 and Ω_2 directly below Equation (1), and adjusted the text accordingly.

[8] Lines 107, 110, 118, 119, and 140: c is not defined. This could be concentration or count, and the reader likely knows the context, but omitting the definition is one of many small things that make this paper difficult to parse.

<u>Responses</u>: The symbol *c* denotes the GF-PDF spectrum, i.e., $c_{\mathrm{cond}}(g)$, the probability density of the growth factor g. As elaborated in the manuscript, i.e., Line #106-111, $c_{\mathrm{cond}}\left(D_{\mathrm{p2}}, D_{\mathrm{p1}}\right)$ represents the represents the GF-PDF, i.e., the probability of particles with a dry diameter of D_{p1} growing to a diameter of D_{p2}. The GF-PDF can be normalized as a function of growth factor g (i.e., $D_{\mathrm{p2}}/D_{\mathrm{p1}}$), which satisfies $c_{\mathrm{cond}}(g, D_{\mathrm{p1}})dg = c_{\mathrm{cond}}(D_{\mathrm{p2}}, D_{\mathrm{p1}})dD_{\mathrm{p2}}$. Given the narrow particle size range classified by the 1st DMA, it is assumed that the GF-PDF is the same for all particles classified by the 1st DMA, and therefore $c_{\mathrm{cond}}(g, D_{p1})$ is independent of D_{p1} for the integration in equation (2) and can be simplified as $c_{\mathrm{cond}}(g)$, or $c(g)$.

[8a] Figure 5: Please define c_i and state its units

Responses: c_{i} denotes the GF-PDF value at the *i*-th growth factor g, i.e., the probability density of particles having a growth factor within that bin. It is dimensionless in g space and has a unit of $(\Delta g)^{-1}$, since the GF-PDF integrates to unity, i.e., $\int c_{\mathrm{cond}}(g)dg = 1$. We have added this definition to the Fig. 5 caption in the revised manuscript.

[9] The statement on line 125 is also this is stated above (it is redundant)

Responses: We have removed redundant information in Line #125.

[10] Lines 123-131: This whole paragraph is redundant

Responses: We have removed redundant information in this paragraph, and it now reads:

"Due to the overlapping of the instrument kernel function $\mathbf{M}$, inversion of the GF-PDF from the HTDMA or HFIMS measurements is an ill-posed problem (Gysel et al., 2009). Regularizations are often applied to nonparametric inversions, which can greatly reduce the error in inverted GF-PDF by eliminating noise amplification. Zhang et al. (2022) compared Tikhonov regularization and Twomey's iterative regularization for GF-PDF retrieval from HFIMS measurements and demonstrated that Twomey's method better balances the smoothness of the inverted GF-PDF with fidelity in reproducing the instrument response. As a result, we adopt Twomey's method as the representative nonparametric inversion method throughout this work.".

[11] Line 259: Please define Adam optimizer and revise the paragraph for a larger audience.

Responses: We have expanded the description of the Adam optimizer in the revised manuscript, and it now reads:

“The Adam optimizer (Adaptive Moment Estimation), an adaptive learning rate algorithm that combines momentum with root-mean-square propagation, was adopted with an initial learning rate of 1×10^{-4} and a batch size of 512.”.

[12] Lines 373-374: The measurements were not available and the “ground truth” here is the Tikhonov – can this be further clarified and stated up front?

Responses: We have clarified this at the beginning of Section 3.4, and it reads:

“Since the true GF-PDFs are not available from real-world measurements, the GF-PDFs retrieved from Deep-GF-PRM predictions are compared against those inverted using Twomey’s nonparametric method.”

[13] Lines 396-402: Please revise paragraph for clarity. Specifically, it seems as though a new calculation is being introduced in the results section. The calculation of the HTDMA GF-PDF (observed) is so simple it may be misleading to refer to it as something that is derived. It is D_i/Ddry for each bin i.

Responses: We thank the reviewer for pointing this out. First, we would like to clarify why a simplified approach was adopted for the HTDMA field data. The MOSAiC HTDMA dataset available from the ARM data repository provides the measured humidified particle size distributions ($\mathrm{d}N/\mathrm{dlog}D_\mathrm{p}$) rather than the raw instrument response R, which is required for the kernel-based nonparametric inversion described in Eq. (4). We therefore adopted the following simplified procedure to derive the HTDMA measured GF-PDF: (1) the measured humidified particle size spectrum $\mathrm{d}N/\mathrm{dlog}D_\mathrm{p}$ was converted to particle number concentration dN in each size bin; (2) the humidified diameter D_p was mapped to growth factor $g = D_\mathrm{p}/D_\mathrm{dry}$ for each bin, and the corresponding $\mathrm{d}N$ was reassigned to the GF domain to yield $\mathrm{d}N/\mathrm{d}g$; (3) the resulting $\mathrm{d}N/\mathrm{d}g$ was scaled to $(\mathrm{d}N/\mathrm{d}g)/N$ as the number probability density function of GF, i.e., GF-PDF.

This approach accounts for the measured particle concentration in each size bin and properly normalizes the output as a probability density function, rather than just relabeling the size axis. We note that this simplified approach neglects the finite width of the DMA transfer functions and is therefore an approximation to the full kernel inversion. However, under the 10:1 sheath-to-aerosol flow ratio used in the MOSAiC HTDMA measurements, the first DMA transfer function is sufficiently narrow that this approximation is reasonable for the purposes of this study.

We have revised the paragraph in Sect. 2.5 to describe these steps explicitly, and it now reads:

“However, the ARM data repository provides the measured humidified particle size distributions ($dN/d\log D_p$) rather than the raw instrument response R required for nonparametric inversion (Eq. 4). We therefore derived the GF-PDF using a simplified approach: (1) the humidified particle size distributions $dN/d\log D_p$ was converted to particle number concentration dN in each size bin; (2) the humidified diameter D_p was mapped to growth factor GF = D_p/D_{dry}, and dN was assigned to the GF domain to obtain dN/dGF; (3) dN/dGF was normalized by the total particle concentration to yield the number probability density function of GF, i.e., $(dN/dGF)/N$. This approach neglects the finite width of the DMA transfer functions and is therefore an approximation of the GF-PDF inversion. However, the 10:1 sheath-to-aerosol flow ratio ensures that the first DMA transfer function is sufficiently narrow that this approximation is adequate. We refer to it as the “measured GF-PDF” to distinguish it from the inverted GF-PDF from the HFIMS measurement. ”

[14] Line 475: The authors provide inadequate citation to the source of the HTDMA data. They should provide an inline citation to ARM and include mention of prior works if possible.

Responses: We thank the reviewer for this suggestion. We have added an inline citation to the ARM User Facility in “Acknowledgements” in the revised manuscript, and it now reads:

“HTDMA data (http://dx.doi.org/10.5439/1095581) used in this paper were obtained from the Atmospheric Radiation Measurement (ARM) User Facility, a US Department of Energy (DOE) Office of Science User Facility.”

TECHNICAL CORRECTIONS

typo in top title of figure 5

Responses: We have corrected it in the revised manuscript.

102: "simplified as:"

Responses: Revised.

413: "noise" not "noises"

Responses: Corrected and checked throughout the revised manuscript.

412: "stronger" not "strong"

<u>Responses</u>: Corrected.

400-401: GF-PDF distribution is redundant, PDF is a distribution function

<u>Responses</u>: Corrected.

Reference

Petters, M. D.: Revisiting matrix-based inversion of scanning mobility particle sizer (SMPS) and humidified tandem differential mobility analyzer (HTDMA) data, Atmos. Meas. Tech., 14, 7909-7928, 10.5194/amt-14-7909-2021, 2021.

Pinterich, T., Spielman, S. R., Hering, S., and Wang, J.: A water-based fast integrated mobility spectrometer (WFIMS) with enhanced dynamic size range, Aerosol Science and Technology, 51, 1212-1222, 10.1080/02786826.2017.1338664, 2017.

Zhang, J., Spielman, S., Wang, Y., Zheng, G., Gong, X., Hering, S., and Wang, J.: Rapid measurement of RH-dependent aerosol hygroscopic growth using a humidity-controlled fast integrated mobility spectrometer (HFIMS), Atmos. Meas. Tech., 14, 5625-5635, 10.5194/amt-14-5625-2021, 2021.

Zhang, J., Wang, Y., Spielman, S., Hering, S., and Wang, J.: Regularized inversion of aerosol hygroscopic growth factor probability density function: application to humidity-controlled fast integrated mobility spectrometer measurements, Atmospheric Measurement Techniques, 15, 2579-2590, 10.5194/amt-15-2579-2022, 2022.