Referee Comments: Christopher Oxford

This paper first lays a brief theoretical framework around regularization, SMPS inversion, and TDMA inversion. The theory surrounding SMPS and TDMA inversion is framed within the developed Julia software environment and not within the traditional DMA/TDMA inversion. However, the author does reference the 2018 paper (first version of this software) which does translate the software framing into the traditional SMPS/TDMA framework. Readers new to this subject will likely need to use the 2018 paper to digest the results as mentioned at the end of the introduction. I was able to do so without issue.

During the theoretical explanation, the author documents the differences between the 2018 edition and this new edition. These changes in TDMA inversion are positive and highlight our current understanding of multicharged behavior. As such, this routine represents a full multi-charge inversion in TDMAs as we currently understand it. Additionally, different regularization techniques are now a part of this software package. These two changes are a marked improvement over existing inversion methods which restrict inversion to a single method or neglect multicharged particles.

The paper then proceeds to test different regularization methods on large data sets, which at this point, is of great value to the community and to me. Comparison of different regularization methods is of great interest, and I suspect I will read the final version several times to digest the results of this study. After reading, I have no major issue with the revision. I have a few comments and questions as documented below.

Line 26: I understand "mixing state" to be internal, external, or a combination of the two. The mentioned variables do not fully define my understanding of mixing state. Maybe a sentence or two clarifying this statement is needed.

The paper is revised to be in line with the formal definition by Riemer et al.: "Aerosol mixing state is defined as the distribution of properties across a population of particles within an aerosol."

To predict the impact of aerosol on the Earth system, the distributions of particle size, chemical composition, hygroscopicity, and morphology must be known. The distribution of these properties across a population of particles formally defines the mixing state of the aerosol (Riemer et al., 2019). Accurate measurements of the mixing state are critical for formulating models that link aerosol, cloud, and climate properties.

Line 61: I am not crazy about the use of the word "shape." I may misunderstand the inversion, but many of these routines do not assume a "parameterized function", however, they do assume a shape. (i.e., a series of rectangles or a series of trapezoids (lines))

The paper has been changed to read "the functional form of the mobility growth factor frequency distribution".

Line 111: Is this supposed to begin a new paragraph???

Fixed.

Line 144: I do not understand where the 8 combinations come from. Is it omission or presence of D and B along with two algorithms?

Revised as follows.

There are eight combinations by which to compose methods via Eq. (5), L, Lx_0, Lx_0B, Lx_0D, Lx_0BD, LB, LBD, and LD. Combined with the three most common filter matrices $L_0 = \mathbf{I}$, $\mathbf{L}_1$ = upper bidiagonal(−1, 1) and, $\mathbf{L}_2$ = upper tridiagonal(1, −2, 1) this results in 24 unique methods.

Line 159: I may be a little confused. Does this mean that only the initial guess is bounded?

The solution is bounded. If the initial guess produces results that are outside the bounds, passing this initial guess to the solver will result in an error. Hence the truncation. To be clear the text is revised as.

The net result is an optimized solution that is within the specified upper and lower bounds. The upper and lower bounds are vectors of the same size as x.

Line 169: Is the initial guess also the a-priori estimate? If so, the words “initial guess” seem inappropriate. When I see “initial guess”, I assume these are the beginning values for x. However, equation 5 states that x0 is not an “initial guess.” Is x0 both an initial guess and the estimate?

The text is now changed to read *a-priori* estimate.

Line 276: there appears to be a Zs (or other variable) missing in the sentence.

Fixed

Equation 11: I do not see how this yields an array. Based on the previous example for mapfoldl, I assume that the output is a sum (the variable a) as you have previously defined. Is the sum (or subtraction) only an example and replacement of – with vcat changes the output from a progressing sum to a concatenating array? If so, can we change the writing in the example to say that the sum is an example.

Yes, mapfoldl will just apply the given function to reduce the mapped collection. If the function is +, it is a sum. If it is vcat, it concatenates arrays.

However, other programming languages may concatenate along a different dimension as the definition of horizontal and vertical is arbitrary. Passing vcat to foldl (or mapfoldl) will result in a concatenated array.

Line 295: I think the word "exiting" should be "entering."

Thank you. Revised.

Lines 295 through 303: A gentle reminder to the reader regarding the meaning of the dot (·) between growth factor and the product of T and the inlet size distribution. For a long while, I thought you were converting the size distribution into another form. Only after some extended study did I realize that g0 was applied only to diameter.

Caveat added a little but further below (line 305).

The size distribution is grown by the growth factor g_0, which is achieved by applying the · operator to ....

Line 337: you state that the mobility grid for DMA 2 is Z s,2 . Do you mean this vector is the array of centroid mobilities for DMA2? Line 315 states that it is such. Is this variable supposed to be Z?

The following has been added to line 315 to clarify.

Note that the choice of Z inside Ω is up to the user. Sensible choices are $Z = Z_{s,1}$ or $Z = Z_{s,2}$ the implications of which are further discussed later.

Line 351: is n the number of growth factor bins? This sentence may need rearrangement.

Yes. It is stated a few lines above. "Thus the growth factor probability distribution P_g in Eq. (17) can be discretized into n arbitrary growth factor bins."

Line 360: Equation 18? Do you mean 17?

The black line is from the Matrix form, i.e. Eq. (18). The colored lines are from Eq. (15) as stated in the caption.

Line 380: Our TDMA does not normally geometrically step (although it can). Will that impact the use of this routine?

It will work with any binning. Perhaps the sentence is too myopic because our HTDMA operates like that.

This is due to the evaluation of the humidified size distribution along a geometrically stepped mobility grid~~, which is typical in scanning DMA setups~~.

Line 390: Please choose how to denote the subscript for D. Because there is no way to show the superscript for e, confusion can ensue. The example on Line 151 does not use exponential form. It would be good if one form or the other is used throughout. I spent a good amount of time trying to figure out what "e" was. I had a long laugh when I did figure it out.

That is a good point. I fixed the notation for equation 6 and defined the use there.

The method $L_1D_{1e\text{-}2}$ represents a filter matrix with a first-order derivative operator applied to Eq. (6) with ϵ = 1e-2. Exponential notation is used because subscripts are difficult to superscript.

Line 552: I expected a more direct statement that summarized which inversion method was better for the dataset. From this I assume I should choose L_2B. Is this correct?

Perhaps. The answer for this dataset is yes. However, I don't want to overgeneralize. I could imagine cases where L_2B results in over-smoothing when two modes are close together.

Line 628: Would prefer using the word function instead of shape as the inversion uses rectangles which is also a shape.

Changed to

… functional form of the growth factor frequency distribution

Referee Comments: Mark Stolzenburg

Referee Comments for Manuscript AMT-2021-51 Ver. 3 “Revisiting Matrix-Based Inversion of SMPS and HTDMA Data” Markus D Petters

This manuscript addresses the important issue of automating the processing of SMPS and tandem DMA data. The idea of inverting data with regularization is sound. However, there is still a functional problem with the development of the matrix-based forward model of calculating system response from a known input distribution. Furthermore, some of the descriptions in that section could be much clearer. If these issues can be properly addressed, the resulting software package should prove of great utility.

Major Comments

As before, the main focus of this review is the development of the forward matrix model in the manuscript. The author has generally addressed my previous comments satisfactorily. In particular, the changes to properly account for multiply-charged particles in the second DMA of a tandem DMA setup are greatly appreciated. However, there appears to be a remaining problem in this situation with tracking the true mobility diameter of the particles after growth, D_{wet}, to the same diameter in DMA2.

When changing thermodynamic state, Λ or (T,p), true mobility diameter (*i.e.* physical diameter) of the particles is the only “size” parameter that remains constant regardless of charge state. For multiply-charged particles, apparent mobility diameter does not equal true mobility diameter and it depends on (T,p). Therefore, for multiply-charged particles transitioning from DMA1 conditions to DMA2 conditions, the relationship between apparent and true mobility diameters changes. If apparent mobility diameter is held constant across the transition, then it is necessarily true that mobility diameter is not.

The somewhat garbled explanation of the function of $\Pi_k^{\Lambda,\delta}$ (lines 299-301) seems to indicate that the particle size sent to the convolution matrix, $\mathbf{O}_k$, for transport through DMA2 is the apparent mobility diameter at DMA1 conditions. Though the actual end effect may be small, this is technically incorrect for multiply-charged particles.

In equation form, this may be seen as follows. Let the function h represent the forward calculation of mobility from diameter as $z_1/k = h_1(D_m)$ where the calculation is done at DMA1 conditions. Then the inverse calculation is represented as $D_m = h_1^{-1}(z_1/k)$. Using this, the apparent diameter at DMA1 conditions is given as $D_{a1} = h_1^{-1}(z_1) = h_1^{-1}(k \cdot h_1(D_m))$. If D_{a2} is equated to D_{a1}, then the corresponding true mobility diameter in DMA2 is given as $D_{m2} = h_2^{-1}(h_2(D_{a2})/k)$. For $|k|=1$, all these equations simply collapse such that $D_{m2} = D_{a2} = D_{a1} = D_m$. But for $|k|>1$, $D_{m2} \neq D_m$, meaning that the true mobility (physical) diameter is not preserved across the transition. Furthermore, if the output of $\Pi_k^{\Lambda,\delta}$, is apparent mobility diameter at DMA1 conditions, then in order to correctly compute the diffusion effects associated with DMA2, this conversion to apparent mobility diameter would need to be directly reversed (at DMA1

conditions) just to get back to true mobility diameter, a computational waste. As far as I can see, the apparent mobility diameter is of no use in the context of these TDMA equations.

Eliminating that concept entirely from the development would also eliminate the need for such strange terminology as an "apparent +1 mobility diameter". At least to me, that equates to a "false true mobility diameter". The term "mobility diameter" alone is well-defined in terms of the particle dynamic mobility (z/ke where e is the unsigned electronic charge). The addition of "+1" is then, in fact, somewhat redundant. There is no need to retain the "+1" when referring to an "apparent mobility diameter".

Since I have not even looked at the code for this, I admittedly know little of the details of the programming of these equations for the calculation of the forward model. However, with the implementation of the corrections discussed above, it seems likely that there remains no compelling reason to treat the input parameters of DMA1, $\Omega^{\delta,\Lambda}(Z, z^s/k, k)$ (Eq. 10), and DMA2, $\Omega^{\delta,\Lambda}(Z, z^s, k)$ (Eq. 15), differently in these equations. Assuming that the algorithm has no hidden switches for different input structures, these variations in input can be quite confusing and seemingly contradictory.

In addition, there is an interpolation step from the input distribution diameter grid to the DMA1 mobility grid and a very similar interpolation step from the grown particle diameter grid, D wet , to the DMA2 mobility grid. Each of these interpolations takes as input the incoming diameter grid and the outgoing mobility grid with its associated DMA Λ parameter. From this, a relatively sparse matrix is generated to operate on the incoming concentration vector resulting in an outgoing concentration vector interpolated to the new grid. In these equations, the two interpolations could be similarly codified as $\Pi^{\Lambda 1}(Z_{s,1}, D_{in})$ and $\Pi^{\Lambda 2}(Z_{s,2}, d_1^s \cdot G)$ where D in is the input diameter grid, $d_1^s = D_p(z_1^s)$ is the mobility diameter associated with z_1^s , G is the vector of discretized g_0 values, and d_1^s, G is the grid of grown diameter bins.

Notation such as used above can greatly improve the clarity of these equations with significantly less reliance on explanations in the surrounding text. In general, there is much room to improve the consistency and logical arrangement of the notation. For instance, there is no obvious reason why the Z dependence, which makes the result a vector, is omitted from $T_{size}^{\Lambda,\delta}\left(k,\ z^s\right)$ (Eq. 10) and the subscript size seems to add no information at all. The use of a subscript 1 (to indicate singly-charged?) in the expression $D_{p,1} = D_p(z,k=1)$ (line 259) becomes confusing when used in Eq. (15) for DMA2 and especially so in $g = D_{p,1}/D_d$ (line 345) where it represents the diameter associated with an element of $Z_{s,2}$. It would be far better to reserve subscripts 1 and 2 for their most frequent usage in associating the parameter with a particular DMA. To distinguish particular values of diameter from the function that generated them, lowercase 'd 's could be used as above. And though the superscript s of z^s was apparently originally representative of "star" in "z-star" or z*, at this distant removal from that original notation and in this context, it simply looks out of place. Even the author changed it to a subscript when the Z_s parameter was introduced. Given the number of flaws uncovered at this point in the formulation of the forward model in Petters (2018), there would seem to be little value in clinging to its confusing notation.

At least in terms of the forward model, the current work should be seen as superseding the previous work.

Overall response: The manuscript was revised to eliminate the need for $\Pi_k^{\Lambda,\delta}$. The transfer function $\Omega^{\delta,\Lambda}(Z, z^s/k, k)$ in (Eq. 10) and (Eq. 15) are now treated identically. This simplifies the notation and also addresses the concern about using the correct thermodynamic state for DMA 2 in the forward model. **Note that these changes did not affect the results in any meaningful way.**

In response to the comment to eliminate "apparent + 1 mobility diameter". To me, the concept of "apparent + 1 mobility diameter" and "apparent growth factor" are useful and I wish to retain them. I added a section upfront that defines these. This section also explains the utility of the concept as I see it.

The DMA selects particles by electrical mobility. The relationship between mobility and mobility diameter is well known and well defined. The relationship is given, for example, in Eq. (2) in Petters (2018). This work also makes use of the "apparent +1 mobility diameter". It is defined as the conversion from mobility to diameter assuming singly charged particles using the mobility grid scanned by either DMA 1 or DMA 2. The apparent +1 mobility diameter represents the natural diameter axis of a DMA response function, i.e. a plot of the raw detector response versus the nominal DMA setpoint diameter. It is an equivalent measure of mobility. The apparent +1 mobility diameter is ambiguous. Larger particles carrying more than one charge may have the same apparent +1 mobility diameter as smaller particles carrying fewer charges. The "apparent growth factor" is defined as the apparent +1 mobility diameter in scanned by DMA 2 divided by the nominal selected dry diameter in DMA.

In response to eliminating the $\Pi_k^{\Lambda,\delta}$ notation/concept: I followed the referee's suggestion but I do want to give a brief explanation. The $\Pi_k^{\Lambda,\delta}$ function was introduced to map from mobility diameter to apparent + 1 mobility diameter BEFORE entering DMA 2. The origin for this was a simple back fold for visualization between mobility and apparent +1 mobility as shown/calculated in Petters (2018). This is strictly correct when no size changes occur between DMA 1 and DMA 2. However, the mapping between mobility and apparent +1 mobility is non linear in the humidified tandem DMA. For this reason $\Pi_k^{\Lambda1,\delta1}$ was introduced. It was applied before entering DMA 2 to maintain conceptual symmetry to the prior work. As pointed out by the referee, the pressure in DMA 2 may be lower. In practice, a few 10s Pa of pressure drop have a negligible effect on $\Pi_k^{\Lambda1,\delta1}$. (In fact even 200 hPa is hardly noticeable). It is possible to apply $\Pi_k^{\Lambda2,\delta2}$ after passage through the DMA to address this concern. However, when doing this there is no utility for visualization. Furthermore, the $\Pi_k^{\Lambda,\delta}$ function can be eliminated while also addressing the referee's concern about the non-symmetrical treatment of the DMA transfer function in the **A** and **O** matrices. For this reason, the $\Pi_k^{\Lambda,\delta}$ function was removed from the treatment. However, there is one **BIG** disadvantage incurred by doing so. The size grid in DMA must be extended to much larger sizes to capture the humidified multiply charged particles. This results in extra computational cost. Furthermore, the revised figures will have a slightly different

binning due to the doubling of the range. The decision for removal of the Π function was motivated by the desire for conceptual clarity and precision over computational speed.

Changes

1. Removal of the PI function and associated text. The key change is that PI is removed and Omega in Eq. (15) is as in Eq. (10). The resulting Figure 1 is essentially unchanged (except for the change in the binning scheme explained below).

Please see changed text after Eq. (13) up to Eq. (16)

2. Bin numbers in all figures were doubled and the range was extended (resulting in approximately the same bin width as before. The reason for this is explained in the text. Due to the new calculation the root mean square error values changed slightly and are updated accordingly.

.. 120 bins between $0.8 < g < 5$. Note that the size grid (or apparent growth factor grid) must be extended to large sizes to capture the growth of multiply-charged particles computed via Eq. (13).

In response to the remaining comments about notation: I appreciate the input and suggestions by the referee. I have implemented most of the referee's suggestions in this and prior revisions. It is my opinion that the added clarifications are sufficient to make the current representation clear enough to follow. The current notation remains close to the implementation in the code and I prefer to keep it that way. Changing the formulation of T, renaming diameters, or introducing alternate constructs to express the interpolation steps will muddy the waters more than clearing it up. Specifically, the software package does retain full backward compatibility with Petters (2018), while also enabling the more precise treatment that is described here. This link will become harder to maintain, the more changes are introduced, and ultimately result in more confusion.

While one aspect of Petters (2018) contained an approximation, i.e. the mapping mobility and apparent +1 mobility diameter for humidified particles in the tandem DMA, it does not mean that it is flawed or superseded in general. The approximation was not that terrible, though it was an unnecessary oversight on my part. However, the remaining points discussed in Petters (2018) fully stand. For example, the excellent agreement between the TSI inversion and inversion using the **A** matrix, the method to compute moments aereas from size selected aerosol using the T function, the application to fit of size-resolved CCN activation spectra while accounting for multiply charged particles, or the application to predict the output size distributions of the dual tandem DMA method. The **A** matrix may be formulated differently in this work, but it is still the **A** matrix. The larger point, a composable computational notation that can express transfer algebraically is also still true. One outcome of this approach is that multiple formulations exist to express the same thing, which is a feature of a language.

Almost all of the excellent suggestions made by the referee in the "Minor Comments and Corrections" were accepted as proposed. Exceptions are explained.

Minor Comments and Corrections

Line 31: "For ~~examples~~ example, …". No matter how many examples are proffered in the following text, standard usage is "For example".

Corrected.

Lines 87-88: "Capital bold-roman letters denote matrices (**A**), lower case roman letters denote vectors (x) and lowercase italic symbols denote scalars (*n*)." However, this list is far from exhaustive. For instance, uppercase italic *T*s are used as vectors in the later development of the forward model.

Revised as follows.

Section 2.1 and 2.2 uses the following linear algebra notation. Capital bold-roman letters denote matrices (**A**) , lowercase roman letters denote vectors (x) and lowercase italic symbols denote scalars (*n*). $\mathbf{A}^T$ denotes the matrix transpose, and $\mathbf{A}^+ = (\mathbf{A}^T\mathbf{A})^{-1}\mathbf{A}^T$ is the matrix pseudo-inverse. Section 2.3 uses additional notation described there.

Line 88 or line 94: Include the definition of the pseudo inverse, $\mathbf{A}^+ = (\mathbf{A}^T\mathbf{A})^{-1}\mathbf{A}^T$, as this is perhaps less well known to many. It also helpful to have that available to compare to the form in Eq. (3).

Added.

Lines 100-101: "Common choices are the first and or second derivative operator defined as the upper bidiagonal(-1; 1) and the ~~upper~~ tridiagonal(1;-2; 1) matrix, respectively." This same reversal is also found in line 145. There is no such thing as an "upper tridiagonal"; the matrix is symmetrically populated about the main diagonal.

Upper has been inserted for bidiagonal. The tridiagonal is defined as the upper tridiagonal matrix as in Huckle and Sedlacek (2012), which is also accordance with the definition given by others, e.g. Eq. 7 in Stout and Kalivas (2006), J. Chemometrics 2006; 20: 22–33, DOI: 10.1002/cem.975.

Line 106: "… the derivative of the right hand side of Eq. (2) with respect to x, …"

Corrected.

Lines 108-109: "The L-curve method involves a plot of $\log\left\|Ax_\lambda - b\right\|_2^2$ vs. $\log\left\|Lx_\lambda - x_0\right\|_2^2$. "Note that x has been changed to x_λ in these expressions."

Corrected.

Line 109: "The optimal occurs at the corner of the resulting L-shaped curve, …"

Corrected.

Lines 111 and following: The description up this point has been very helpful allowing for at least some intuitive grasp of what is going on in these equations and their optimization. Then the GCV estimator is introduced (Eq. 4) with absolutely no physical context for what it means. I realize that the full definition for this is somewhat involved and beyond the scope of this paper, but is there any sort of simplified view of what this parameter physically represents? At least for me, the form of the equation provides no clue at all. Perhaps that is in part due to my ignorance of the standard form and what that physically represents.

The red colored segments have been inserted.

The optimal regularization parameter can be obtained using a variety of techniques, including the L-curve method (Hansen 2000) and generalized cross-validation (GCV, Golub et al., 1979). Both methods use metrics that penalize solutions with large variance (amplified noise) or large bias.

The L-curve method involves …

The generalized cross-validation estimator presents a mathematical shortcut to compute the leave-one out cross-validation estimate, which removes one point from the data, creates a model, computes the error between the model and data point not included in the data, and then averages the result over all permutations. It is given by ...

Line 113: $\mathbf{A}^T$ should be $\mathbf{A}^T$ (no italics on the superscript), two places.

Corrected.

Lines 150-151: Doesn't $\mathbf{D}_{\hat{\mathbf{x}}}^{-1} = diag\left(\left|\hat{x}_1\right|, \ldots, \left|\hat{x}_n\right|\right)$ imply $\left(\mathbf{D}_{\hat{\mathbf{x}}}\right)_{ii} = \left|\hat{x}_i\right|$? But then that together with $\left(\mathbf{D}_{\hat{\mathbf{x}}}\right)_{ii} = \varepsilon$ for all $\left|\hat{x}_i\right| < \varepsilon$ creates a contradiction if the set $\left|\hat{x}_i\right| < \varepsilon$ is not empty.

Apologies to the poor typesetting, above which is due to limited support of mathtype in google docs. The sentence has been revised :

Elements that satisfy $|\hat{x}_i| < \varepsilon$ are set equal to ε, where $0 < \varepsilon << 1$.

Line 179: "RegularizationTools.jl also provides an abstract generic interface …"

Corrected.

Line 196: "… the raw response function defined as the integrated response …"

Corrected.

Line 201: "… requires ~~solving~~ evaluating integrals …"

Corrected.

Lines 203-206: "For the forward calculation, the objective is to find a design matrix that maps the growth factor frequency distribution to the raw TDMA response function. ~~The objective is to find a design matrix that maps the growth factor frequency distribution to the raw TDMA response function.~~"

Corrected.

Line 208: "… provide a domain ~~specific~~ independent language …" This change would seem to make more sense in this context.

Domain specific language is used as a language that is specific to a domain, in this case to express the transformations occurring in the domain of differential mobility analyzer systems. No change was made.

Line 223: "… +1 (singly charged) mobility diameter bin edges …" If you wish to continue to use the somewhat superfluous "+1" notation, it should be explained such as this on first use.

Done.

Line 226: "For ~~examples~~ example, …"

Corrected.

Line 227: "…f * $\mathbb{m}$ is the uniform scaling of the concentration fields by factor f." Here and in the subsequent example of $f \cdot \mathbb{m}$, f is treated as a simple scalar. But in the very next paragraph f and g are used as functions. Rather than tempt confusion, use some other letter such as a for scalar multiplication, e.g. a * $\mathbb{m}$.

Corrected.

Line 233: "… and sums the results."

Corrected.

Lines 234 and following: The description of the foldl() function is clearer than that of the previous splatting function. However, you have stated the generic rule for the first element while giving examples directly from Julia documentation which use a different rule. The generic rule is that for the first element, a is the neutral value. In the Julia implementation of foldl, the first element acts as a for the processing of the second element. For the example foldl(–; [1; 2; 3]), the neutral value is 0 and the operation is – (a, x) = a – x. The generic implementation of this would be 0 – 1 – 2 – 3 = – 6 while the Julia implementation is 1 – 2 – 3 = – 4. The way foldl is used in the development of the forward model would appear to conform to the Julia implementation. So the description of the first element rule should be changed.

The implementation is not Julia specific. Folds without specifying an initial value are used in other languages as well, e.g. Haskell's foldl1 which behaves identical to Julia foldl without initial value. The "neutral value" means that -(a,x) = x. Perhaps the identity element is a better description. In any case, the change below makes it explicit and has been added to the manuscript.

If no initial value is provided, as is the case in this manuscript, foldl applies the function to the first two elements of the list to compute the first a.

Lines 265-266: "… which corresponds to the centroid mobility setting for the DMA to transmit particles ~~with the size of particles~~ with k charges under the assumption that they carry only a single charge."

Corrected.

Even with this change, I find it very difficult to understand what is going on in this section. The only way I can make sense of this is that the Z vector at this point contains values of z/k, not simply z. If this is the case, it needs to be stated as so somewhere. Also, if this is the case, then I

believe the DMA transfer function dependence in Eq. (10) given as Ω (Z, z^s /k, k) should actually be Ω(Z, z^s /k, 1), assuming Ω(Z, z^s , k) is the "normal" arrangement of inputs.

This is not the case. It is just a mathematical trick to coax the generic function Ω(z, z^s , k) to map in a manner such that $D_{p,1}$ becomes equal to mobility diameter. It is what enables the use of $D_{p,1}$ in the subsequent terms. Below is a slightly revised explanation.

The functional Ω depends on three arguments Ω(Z, z*, k) and implicitly on the DMA configuration Λ (i.e., Eq. 13 in Stolzenburg and McMurry 2008). The output is a vector along the mobility grid Z. The maximum transmission occurs at $Z/z^* = 1$. The last argument denotes the number of charges carried by the particle. It is used to compute the mobility diameter from z* and in turn the diffusion coefficient which is required to account for diffusional broadening of the transfer function. The output of $T_{size}^{\Lambda,\delta}\left(k,\ z^s\right)$ is the transmission of particles through the DMA in terms of the true particle mobility diameter. This is achieved by passing z^s/k as argument to Ω, which corresponds to the centroid mobility setting for the DMA to transmit particles with k charges under the assumption that they carry only a single charge. The net result is that $D_{p,1} = D_p(z,k=1)$, where z is an element of Z, becomes equal to the true mobility diameter axis. As a consequence the charge fraction $T_c(k, D_{p,1})$ and penetration efficiency $T_i(D_{p,1})$ are evaluated at the correct diameter.

Line 268-270: "The function $T_{size}^{\Lambda,\delta}\left(1,\ z^s\right)$ evaluates to a vector of the same length as Z. Performing and elementwise sum over $T_{size}^{\Lambda,\delta}\left(k,\ z^s\right)$ produces the net mobility distribution transmitted by the DMA." Perhaps it is clear enough from context, but noting that the summation is over all k could be helpful. More importantly, the result of the summation is only the net mobility distribution if $\sum_k T_{size}^{\Lambda,\delta}\left(k,\ z^s\right)$ is multiplied by the input distribution, $\mathbb{m}$. Otherwise, the bare summation is simply the net transmission probability function.

Revised as follows.

Performing an elementwise sum over all $T_{size}^{\Lambda,\delta}\left(k,\ z^s\right)$ (where the sum is over all charges k) produces the net transmission probability function. Multiplication of the transmission probability function with the input distribution results in the mobility distribution transmitted by the DMA. Examples for $T_{size}^{\Lambda,\delta}\left(1,\ z^s\right) * \mathbb{m}$, $T_{size}^{\Lambda,\delta}\left(2,\ z^s\right) * \mathbb{m}$, and $T_{size}^{\Lambda,\delta}\left(3,\ z^s\right) * \mathbb{m}$ are shown in Figure 2, right panel in Petters (2018).

Line 270: “Examples for … ~~is~~ are shown in Figure 2, …”

Corrected.

Lines 276-277: “… and Z_s is a vector of centroid mobilities scanned by the DMA.” The vector Z_s is missing from this description.

Corrected.

Line 277: “The matrix is square if $Z_s = Z$ in Eq. 10.” I think you mean Eq. 11.

Corrected.

As noted earlier, as this would be a lot clearer if the explicit Z dependence were actually shown in $T_{size}^{\Lambda,\delta}\left(k,\ z^s\right)$ as $T_{size}^{\Lambda,\delta}\left(Z,\ z^s, k\right)$.

As discussed above I prefer to keep it the way it is. Once reason is that $T_{size}^{\Lambda,\delta}\left(k,\ z^s\right)$ function generally queries the Z grid defined by the DMA (it doesn't have to). Thus, in code the Z vector becomes δ.Z and Dp,1 vector δ.Dp. Revising it as proposed would confuse the issue.

Lines 289-290: “The latter is the raw response function defined as integrated response downstream of the DMA …” The latter refers to $\mathbb{r}$ in Eq. 12. $\mathbb{r}$ is similar to a size distribution, it is NOT integrated over all sizes as would be the case for the response of a CPC detector.

Reworded as follows.

The latter is the raw response function, where each element corresponds to the integrated response downstream of DMA 1 for a set upstream voltage (or corresponding z^s or apparent +1 mobility diameter but not true physical diameter for multiply charged particles).

Line 295: “The mobility distribution ~~exiting~~ entering DMA 2 …”

Corrected.

Line 297: “… Eq. (13) … evaluates to … that exit the humidity conditioner ~~DMA1~~ …”

Corrected.

Lines 298-299: “Subscripts are used to differentiate DMA 1 and 2 which possibly have different geometries, flow rates, and grids, …” This list should include thermodynamic states, that is, different temperature and/or pressure. DMA2 is definitely at a lower pressure than DMA1 and this is reflected in the relationship between mobility and mobility diameter.

Added as suggested.

..,which possibly have different geometries, flow rates, thermodynamic state, and mobility grids, ...

Line 311: “The total humidified mobility distribution $m_t^{\delta 2}$ exiting DMA 2 is given by (Eq. 14) …” The total humified mobility distribution exiting DMA2 would be that observed at fixed $z_{s,2}$ without integrating over particle size. $m_t^{\delta 2}$ is the total integrated concentration versus $z_{s,2}$. It is not a true particle distribution.

The “apparent +1 caveat was stated directly after the equation. It is now moved upfront.

The total humidified apparent +1 mobility diameter distribution $m_t^{\delta 2}$ exiting DMA 2 is given by:

Line 322: “… the humidified distribution function exiting DMA 2 …” Same as previous comment. This might be considered to be a pseudo-distribution if what is meant by that is explained up front.

If the aerosol is externally mixed, the humidified apparent +1 mobility diameter distribution function exiting DMA 2

Line 324: In reference to Eq. 16, “…the diameters in $\mathbb{M}_t^{\delta 1}$ are normalized by D_{dry} .” This does not seem correct. The diameters, or at least the associated mobilities, cannot be altered until after passage through DMA2. I can’t figure out where in Eq. 16 you could safely normalize diameters.

Thank you. This is indeed a poor description on my part. (And the referee is of course correct). In fact the code evaluates

$$\sum_{k=1}^{m}\left(\mathbf{O}_k * \mathbb{M}_k^{\delta 1}\right)$$

which results in m apparent + 1 mobility diameter distributions after passage through DMA 2. The resulting sizes are then normalized by D_{dry}. The text has been updated accordingly.

Line 327: "… the DMA ~~setup~~ setups Λ_1, Λ_2 , δ_1, δ_2 …"

Corrected.

Lines 332-333: "Transmission through DMA 1 is computed for a specified z^s (the dry mobility) and g_0 (the growth factor) via Eq. (13)." The humidity conditioner as represented by g_0 is NOT part of DMA1.

Corrected.

Line 339: "… the matrix is non square." This is only true if j≠i. Perhaps j is typically much smaller than i, but you haven't said anything about that.

This was a "typo" (the sentence was meant to read):

If the vector *Z* inside the square bracket of Eq. (15) equals that of DMA 1, the matrix is ~~non~~ square.

Line 342: "The advantage of interpolation is that the the matrices $\mathbf{O}_k$ are smaller."

Corrected.

Here again it is implied that j is significantly smaller than i but you have not explained why that is.

Corrected.

..the matrices $\mathbf{O}_k$ are square and of dimension $j \times j$. In that case, the transmitted and grown distribution from DMA 1 (i bins along the mobility axis of DMA 1) is interpolated onto the mobility grid of DMA 2 prior to evaluating $\mathbf{O}_k$ * $M_k^{\delta 1}$ The advantage of this approach is that for $j < i$ the matrices $\mathbf{O}_k$ are smaller and subsequent calculations are faster.

Line 347: "… the humidified mobility pseudo-distribution function …" or some such modification.

Corrected.

Equation (17) is cast into matrix form such that the humidified apparent +1 mobility diameter distribution function is given by

Lines 350-351: "If the grids for P_g and that of DMA 2 do not align, interpolation is used to map the P_g grid onto the DMA 2 grid." The P_g grid would refer to the discretization of g_0 , or G, using the notation from above. What is being interpolated onto the DMA2 grid is $d_1^s \cdot G$.

Changed as follows.

If the grids used to represent the growth factor distribution and that of DMA 2 do not align, interpolation is used to map the growth factor bins from the growth factor distribution onto those corresponding to the DMA 2 grid.

Lines 351-352: "The choice of i, j, n, the ranges of mobility grids for DMA 1, DMA 2, and the range of P_g , is only constrained …" Insert comma after P_g . The range of P_g is simply [0,1]; it is the range of g_0 that is needed here or perhaps "the range of the growth grid for P_g ".

Changed as suggested.

The choice of i , j , n, the ranges of mobility grids for DMA 1, DMA 2, and the range of the growth grid for P_g , is only constrained by computing resources and a physically reasonable representation of the problem domain.

Line 359: A new paragraph should start with "Figure 1 …".

Corrected.

Line 390: Concerning bounds for P_g , if $P_g = dF/dg$ from Figure 1 caption and as would be appropriate in Eq.16, then there is no upper bound for P_g. For if one introduced a perfectly monodisperse as the input aerosol, P_g would range from zero to infinity. However, in discretizing the integral of Eq. 16 it is possible to assign the bin width factor, Δg, to the dF/dg vector to get a ΔP_g vector. This would be properly bounded by [0,1]. This should be explained more clearly or the notation in the equations appropriately modified to show this.

The regularization/fitting is performed in frequency space and the data are subsequently converted to $P_g = dF/dg$. The reasons for this are (1) the ability to specify $B_{[0,1]}$ and that the error metrics are better behaved in the frequency domain. This was hinted at in the caption of Figure 3, but should have been made more explicit in the text.

Here $B_{[0,1]}$ is shorthand for setting all lower bounds equal to zero and all upper bounds equal to one. The a-priori estimate x_0 is taken to be the normalized apparent growth factor distribution derived from the measured response function, where the normalization ensures that the sum over all bins is unity. Note that the inversion is performed treating the growth factor distribution in units of frequency instead of frequency density. This choice enables the upper bound constraint of unity. Since the true noise-free input growth factor frequency distribution is known, the fidelity of the inversion can be evaluated by

computing the root mean square error between the noise-free solution and the regularized solution. Evaluating the root mean square error in frequency rather than frequency density space results in more comparable values when contrasting narrow and broad probability distribution functions.

Lines 396-397: “Visual evaluation … ~~suggest~~ suggests …”

Corrected.

The balance of the paper was not reviewed.