Response to Reviewer Comments

August 3, 2021

Author Statement

Dr. Stolzenburg contacted me via email and offered to send additional comments regarding Section 2.3 as written in my response to the original comments. He did so on July 10, 2021. I thank Dr. Stolzenburg for his additional effort (and very appreciated comments) to help me improve the clarity of the manuscript. Since I had not uploaded a revised version of the manuscript prior to receiving these comments, I respond to them here. The revised version of the manuscript that will be submitted to AMT will have taken these comments into account. A new version of section 2.3 is included at the end of this comment. It supersedes the version in AC1 and AC2.

Response to Reviewer #2 (Mark Stolzenburg)

The comments were sent in the form of an annotated pdf document. Below I transcribe the comments in the following form (1) Text passage that the comment refers to, (2) Verbatim annotated comment, (3) response to comment, (4) revisions. If the context is unclear from the text fragment, please see the full text on starting on pg. 13 of AC2 on the discussion site.

Overview

[1] The integrated response downstream of a tandem DMA that is operated at voltages V_1 and V_2 is given by a double integral and the summation of all selected charges. The integrals are over the upstream size distribution and the aerosol conditioner function, which here is the growth factor frequency distribution.

[1] Text

[2]The proper verbiage used with integrals is as follows: integral(f(x)dx) is the integral of f(x) over x. f(x) is called the integrand. The integrals here are actually over the upstream particle size, or some substitute for it, and the grown or downstream particle size, or some substitute for that. The functions you specify represent only part of the integrand in each case given here. Even if you change the "over"s to "of"s, the statement is still very misleading given the many unaccounted for factors missing in the integrands. An integral over "the aerosol conditioner function" makes little sense since it is a function of both upstream and downstream sizes.

[2] Referee

[3] *Thank you.*

[3] *Response*

[4] **The integrated response downstream of a tandem DMA that is operated at voltages V_1 and V_2 requires solving integrals of the upstream particle size distribution over size and the grown particle size distribution over size. The integration must be repeated for each charge state.**

[4] **Revision**

[5]The objective is to find find a design matrix that maps the growth factor frequency distribution to the raw TDMA response function.

[5] Text

[6] Perhaps it would be clearer to say "For the forward calculation, the objective is ...". Correct “find find”.

[6] Referee

[7] *Thank you.*

[7] *Response*

[8] **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.**

[8] **Revision**

[9]The resulting expressions are concise. They are easily identified within actual source code.

[9] Text

[10] For anyone familiar with the specific language used.

[10] Referee

[11]**The resulting expressions are concise. They are easily identified within actual source code when working through the examples provided with the package documentation.**

[11] **Revision**

[12]Size distributions encoded as a *SizeDistribution* composite data type.

[12] Text

[13] This is not a sentence, there is no verb. "Size distribution are encoded .."?

[13] Referee

[14]**Size distributions are represented as a histogram and internally stored in the form of the *SizeDistribution* composite data type.**

[14] **Revision**

[15]Composite data types combine multiple arrays into a single symbol for ease of use, facilitating faster experimental design and analysis. *SizeDistribution* consists of vectors of bin edges, bin midpoints, number concentration, log-normalized spectral density, and logarithmic bin widths.

[15] Text

[16]"+1 mobility diameter bin edges, bin midpoints". Since later many different "size" parameters are introduced and used, it is best to be specific here. As with the DMA transfer function, are corresponding +1 mobilities also part of this data type?

[16] Referee

[17]*I agree with the suggestion. Yes, the mobility grid is also included. In practice arrays of centroid mobilities and mobility bin edges are created, from which the +1 mobility diameter is computed.*

[17] *Response*

[18]**Composite data types combine multiple arrays into a single symbol for ease of use, facilitating faster experimental design and analysis. The size distribution data type *SizeDistribution* includes vectors of the selected mobility bins considered by the DMA, +1 mobility diameter bin edges and +1 mobility diameter bin midpoints computed from the mobility grid, number concentration, log-normalized spectral density, and logarithmic bin widths.** [18] **Revision**

[19](Note that the Petters (2018) used $T. \cdot \mathbb{n}$ is the elementwise scaling. The extra dot has which has been dropped to stay consistent with the current software implementation). [19] Text

[20] [The referee highlighted multiple places of dangling wording introduced during editing and made suggestions for improvements.] [20] Referee

[21] **(Note that Petters (2018) used $T. \cdot \mathbb{n}$ as the elementwise scaling. The extra dot has been dropped to stay consistent with the current software implementation).** [21] **Revision**

[22]Functions are used to reduce expressions. [22] Text

[23] Not all of the following reduce the dimension of the expression. Or are you talking about a reduction or compactness in notation? If so, use a different word to avoid confusion with later usage. [23] Referee

[24]**Functions are used to evaluate expressions.** [24] **Revision**

[25] If $f(X)$ evaluates to a vector, the sum is the sum of the vectors. [25] Text

[26]Should this be $f(x)$ or are you intentionally using X in indicate a vector? [26] Referee

[27] **If $f(x)$ evaluates to a vector, the sum is the sum of the vectors.** [27] **Revision**

[28]The function $map(f, x)$ applies f(x) to each element of vector x and returns a vector of results in the same order. [28] Text

[29]Perhaps this should be (f, X)? [29] Referee

[30] *Based on the above, no change.* [30] *Response*

[31] The function $reduce(f, x)$ applies the bivariate function $f(x, y)$ to each element of x and accumulates the result. [31] Text

[32]From this and what I have read online, my understanding is that f(x,y) uses the result of its previous application and combines that with the next x value. Nearly every online example I can find only uses functions that treat x and y the same, e.g. f is given as simply "+" or "*" such that interchanging x and y has no effect. Since f can be a user-defined function, it need not be symmetric in x and y, e.g. f(x,y)=x2-y. The documentation does not make it clear whether such usage is allowed. However, if it is, the order of arguments in f(x,y) matters, that is, which is the result of the previous operation and which is the new x value. From online examples, I have gotten the impression that x is the previous result and y is the new x value. This makes the limited documentation such as given here quite misleading. Given no additional information, the most natural assumption would be that the new x value is associated with the x in f(x,y) and the previous result is associated with y. But this would be just the opposite of what is needed. At minimum, I would suggest writing this as "f(y,x) where y is the result of the preceding operation". Also according to an online example at jhub.com/julia, note that the for the first operation, x1 is the previous result and x2 is the new x. [32] Referee

[33] *The referee is correct that reduce is not associative. The order of operation matters. The more general version of reduce(f,x) is foldl(f,x) and foldr(f,x), which guarantee left or right associativity. In regular Julia programs, reduce(f,x) = foldl(f,x). However, there is no guarantee made by the language and any applied parallelism could break the expression. Using strictly associative folds is more precise. The change has been made throughout. The definition has been clarified as suggested by the referee.* [33] *Response*

[34]**The function** $\mathrm{foldl}(f, x)$ **applies the bivariate function** $f(a, x)$ **to each element of** x **and accumulates the result, where** a **represents the accumulated value. For the first element in** x**,** a **is the neutral value. For example** $\mathrm{foldl}(-, [1, 2, 3])$ **evaluates the function** $-(a, x)$ **and yields** $1 - 2 - 3 = -4$**. The function** $\mathrm{mapfoldl}(f, g, x)$ **combines** map **and** foldl**.** [34] **Revision**

[35] [35] **Revision**

$$\mathbf{A} = \mathrm{mapfoldl}\{z^s \to \Sigma[k \to T^{\Lambda,\delta}_{size}(k, z^s), m]^T, \mathrm{vcat}, Z_s\} \tag{11}$$

[36]It applies function f to each element in x, and then reduces the result using the bivariate function function $g(x, y)$. [36] Text

[37] $g(y, x)$ or even $g(a, x)$ where a represents the accumulated value so far. See preceding note. [37] Referee

[38] *Thanks for the suggestion.* [38] *Response*

[39] **It applies function f to each element in x such that $y = f(x)$ and then reduces the result using the bivariate function function $g(a, y)$ where a represents the accumulated value. For the first element $f(x)$, a is the neutral value. For example,** $\text{mapfoldl}(\text{sqrt}, -, [4, 16, 64])$**evaluates to** $\text{foldl}(-, [2, 4, 8]) = 2 - 4 - 8 = -10$**.** [39] **Revision**

[40] The function $vcat(x, y)$ concatenates arrays x and y along one dimension. [40] Text

[41] For clarity, shouldn't this be "first dimension" according to online documentation? [41] Referee

[42] **The function $vcat(x, y)$ concatenates arrays x and y along the first dimension in Julia. However, other programming languages may concatenate along a different dimension as definition of horizontal and vertical is arbitrary.** [42] **Revision**

[43] Petters (2018) gives a simple expressions that model transfer through the DMA. [43] Text

[44] Either "a simple expression that models" or "simple expressions that model" with no preceding "a". [44] Referee

[45] **Petters (2018) gives a simple expression that models transfer through the DMA.** [45] **Revision**

[46] [46] Text

$$T^{\Lambda,\delta}_{size}(k, z^s) = \Omega(Z, z^s/k, k). * T_c(k, D_{p,1}). * T_l(Z, k) \tag{10}$$

[47] The given dependence of omega does not make sense to me. Let z be an element of Z. For the mobility passed in a basic transfer calculation, it is the value of z/zs that matters. For the diffusion calculation, it is z/k that matters. zs/k does not get used directly. Though what is shown may not be technically wrong, it obscures the true dependencies. Simply (Z, zs, k) would be better, as in Eq. (15) for DMA 2. [47] Referee

[48] *Note that revisions will be indicated after responding to the next few comment about this equation. The referee is correct that implementation of the function is $\Omega(Z, z^s, k)$. The charge state is needed to compute the diffusion coefficient. However, for the $T^{\Lambda,\delta}_{size}(k, z^s)$* [48] *Response*

function z^s/k is the correct argument to produce the actually transmitted mobility size distribution. One way to think about this is to ask the question: what centroid mobility z^swould I have to set the DMA to transmit particles carrying k charges if they had only a single charge? Eq. (10) therefore represents the mobility size distribution transmitted. In Eq. (15) is $\Omega(Z, z^s, k)$, as all particles have the same mobility. This version allows using the same omega function in both cases.

[49]

$$T^{\Lambda,\delta}_{size}(k, z^s) = \Omega(Z, z^s/k, k). * T_c(k, D_{p,1}). * T_l(Z, k) \quad (10)$$

[49] Text

[50]What is Dp,1? Apparently, it is the true particle mobility (not electrical mobility) diameter, Dp,1=Dp(z,k=1), where z is an element of Z. If it is to be used, has it been defined elsewhere? Perhaps it is part of the definition of the size distribution composite data type. If so, it should be defined there. Tl (penetration efficiency, not loss) also depends on this diameter, Dp,1. Why is it not written that way? Otherwise, simply Tc(Z,k).

[50] Referee

[51]Are the dots necessary here in ".*" or should they be dropped as in Eq. 13?

[51] Referee

[52]In Eq. (10), Z is a vector of mobilities -> "a vector of particle mobilities". As Z is apparently used for both particle mobilities as well as DMA centroid mobilities (see note below), it would be best to be clear to which it applies each time it is used.

[52] Referee

[53] *Loss has been changed to "penetration efficiency". Yes, the dots are still required for the * operator. They were only dropped for the · operator. The interpretation of $D_{p,1} = D_p(z, k = 1)$, where z is an element of Z is correct. Tl was written this way in the original draft and in Petters (2018), but changed during the first round of revisions. The change is reverted here to stay consistent with previous notation. I revised the text as follows to better explain how this equation works. The meaning of Z is also further clarified when discussing the discretization scheme near the end of the document.*

[53] *Response*

[54] **Petters [2018] gives a simple expression that model transfer through the DMA. The function $T^{\Lambda,\delta}_{size}(k, z^s)$ evaluates to a vector representing the fraction of particles carrying k charges that exit DMA$^{\Lambda,\delta}$ as a function of mobility**

$$T^{\Lambda,\delta}_{size}(k, z^s) = \Omega(Z, z^s/k, k). * T_c(k, D_{p,1}). * T_l(D_{p,1}) \quad (10)$$

where z^s is the centroid mobility selected by the DMA (determined by the voltage and DMA geometry), Z is a vector of particle mobilities, Ω is the diffusing DMA transfer function [Stolzenburg and McMurry, 2008], T_c is the charge frequency distribution [Wiedensohler, 1988], and T_l is the diameter-dependent penetration efficiency [Reineking and Porstendörfer, 1986]. The diameter $D_{p,1} = D_p(z, k = 1)$, where z is an element of Z. The function Ω has been updated from Petters (2018). The version in Petters (2018) computed the shape of the transfer function corresponding to singly charged particles and then applied the same shape of

[54] **Revision**

the transfer function and diffusional loss to the multiply charged particles. The functional Ω depends on three arguments $\Omega(Z, z^s, k)$ [Stolzenburg and McMurry, 2008]. The charge state is used to compute the diffusion coefficient and thus account for diffusional losses and broadening of the transfer function for multiply charged particles.

The output of $T^{\Lambda,\delta}_{size}(k, z^s)$ 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 the size of 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 $T_l(D_{p,1})$ are evaluated at the correct diameter. The function $T^{\Lambda,\delta}_{size}(1, z^s)$ evaluates to a vector of the same length as Z. Performing an elementwise sum over all $T^{\Lambda,\delta}_{size}(k, z^s)$ produces the net mobility distribution transmitted by the DMA. Examples for $T^{\Lambda,\delta}_{size}(1, z^s)$, $T^{\Lambda,\delta}_{size}(2, z^s)$, and $T^{\Lambda,\delta}_{size}(3, z^s)$ is shown in Figure 2, right panel in Petters (2018). Note that Eq. (10) can be evaluated using arbitrarily discretized Z vectors.

[55]corresponding to singly charged particles and then apply the same shape of the transfer function

[55] Text

[56]"applied"

[56] Referee

[57] **fixed in the revised paragraph above.**

[57] **Revision**

[58] Petters (2018) also gives an expression that evaluates to the convolution matrix for passage through a single DMA.

[58] Text

[59]Summation over k means the information on particle physical diameter of multiply charged particles is
lost. Eq. (11) will not work for the first DMA in a TDMA setup using diffusing transfer functions or other diffusion effects in the second DMA. Something should be included here to indicate that, or that this expression is only for something like an SMPS system. I feel that "single DMA" is just not sufficient. The reference to "apparent +1 mobility diameter" at the end of this topic is useful in making this point but does not really make the point of "only apparent diameter, not physical" and comes far too late to make the point in question.

[59] Referee

[60] *Correct. The information is now explicitly included*

[60] *Response*

[61] **Petters (2018) also gives an expression that evaluates to the convolution matrix for passage through a single DMA that is valid in the context of size distribution measurement system, e.g. SMPS. Since the expression includes a summation over all charges, the information on particle physical diameter of multiply charged particles is lost.**

[61] **Revision**

[62]

$$\mathbf{A} = \text{mapreduce}\{z^s \rightarrow \Sigma[k \rightarrow T_{size}^{\Lambda,\delta}(k, z^s), m]^T, \text{vcat}, Z\} \tag{11}$$

[62] Text

[63] I feel it would be cleaner to first define a convolution matrix for transport through a DMA, with no summation over k, as in Eq. 15, perhaps Okv or better yet Ov(k) where v (actually nu) is the DMA. This could then be used for DMA 2 as well below, using an analogous definition of Tsize. Here, for use as the convolution matrix through a single DMA, Av = sumk(Ov(k)). Otherwise, in the current development of the matrices, the similarity of treatment for DMA 1 and DMA 2 is buried. At the very least, though hardly preferable, Eq. 11 should be rearranged to put the summation as the outermost operation on the right side. This would at least provide a little more symmetry of treatment of the two DMAs.

[63] Referee

[64] *I do not fully disagree with the referee about the potential elegance of making the expressions more symmetric. However, the matrices* **A** *and* **O** *serve two separate purposes. The former is valid for passage through a single DMA that is valid in the context of size distribution measurement system (which is now clarified), the latter is valid for evaluating the response after passage through the tandem DMA, starting with the transmitted distribution. The expressions for these are well-defined. It should be clear by now that the approach is in principle highly expressive. The version here, or the version in Petters (2018) are two examples how to write* **A**. *More ways certainly exist, perhaps even more elegant ways. In the context of this work, I believe that this approach will lead to further distance the expressions from the original work and in the end result in more confusion rather than additional clarity.*

[64] *Response*

[65] ... and Z is a vector of centroid mobilities scanned by the DMA.

[65] Text

[66] Z=Zs (see following note) implies **A** is a square matrix. Do you really want to introduce that restriction at this point?

[66] Referee

[67] Eq. 10 used Z as a vector of particle mobilities. Are these two vectors identical, that is, the DMA centroid mobilities and input particle mobility bin midpoints? If so, that should be explicitly noted to avoid confusion. Better yet would be to change Z here to Zs noting somewhere previously that Z=Zs.

[67] Referee

[68] *Although I have been using the same Z vectors in Eq. (10) and (11) - and thus square matrices - this is not a necessary restriction. This is also further clarified when discussing the discretization scheme.*

[68] *Response*

[69] **... and Z_s is a vector of centroid mobilities scanned by the DMA. The matrix is square if $Z_s = Z$ in Eq. 10. However, this is not a necessary restriction.**

[69] **Revision**

[70]...removing the julia specific splatting...

[70] Text

[71]Julia documentation uses this as capitalized, "Julia". Also, this should be hyphenated as "Julia-specific".

[71] Referee

[72] *Changed capitalization here and throughout the work.*

[72] *Response*

[73]**...removing the Julia-specific splatting...**

[73] **Revision**

[74] To help with parsing the expression, $T^{\Lambda,\delta}_{size}(k, z^s)$ evaluates to a vector of transmission for k charges and set point centroid mobility z^s as a function of the entire mobility grid (e.g. 120 bins discretized between mobility z_1 and z_2).

[74] Text

[75]So Tsize is actually a matrix of size [n x m] where n=120 is the number of particle mobility bins. As the above equations are defined it would be useful to clearly indicate the dimensions of the operands and the result.

[75] Referee

[76]"entire particle mobility grid". Though Z serves a dual purpose, in this context it is particle mobility.

[76] Referee

[77] *$T^{\Lambda,\delta}_{size}(1, z^s)$ evaluates to a vector of length Z; $T^{\Lambda,\delta}_{size}(2, z^s)$ evaluates to a vector of length Z. The text prior to this paragraph is updated to better explain the origin and dimensionality $T^{\Lambda,\delta}_{size}(k, z^s)$.*

[77] *Response*

[78] **See revisions to previous comments.**

[78] **Revision**

[79] Note that by design `n` and `r` are *SizeDistribution* objects, which represented the distribution as a histogram in both spectral density units (dN/dlnD) and concentration per bin units.

[79] Text

[80]The input distribution, (blackboard "n"), is readily defined in terms of the true mobility (k=1) diameter, Dp1. However, information on the original charge state exciting the charger is lost from the response distribution (blackboard "r") leaving

[80] Referee

only electrical mobility, z, as the size parameter. Thus, the response distribution cannot accurately be converted to dN/dlnD. Any such conversion must neglect the true charge distribution.

[81] *That is correct. Nonetheless the mathematical/computational representation is that of a SizeDistribution object. The limitation is now noted.* [81] *Response*

[82] **By design $\mathbb{n}$ and $\mathbb{r}$ are *SizeDistribution* objects, which represent the distribution as a histogram in both spectral density units (dN/dlnD) and concentration per bin units. The latter is the raw response function defined as integrated response downstream of the DMA as a function of upstream voltage (or corresponding z^s or apparent +1 mobility diameter but not true physical diameter for multiply charged particles). Note, however, that the response function is not a true particle size distribution in the scientific sense since information about multiply charged particles is lost. The representation of $\mathbb{r}$ as *SizeDistribution* object is to allow response functions to used in the expression-based framework used here.** [82] **Revision**

[83]The latter is the raw response function defined as integrated response downstream of the DMA as a function of upstream voltage (or corresponding z^s or corresponding apparent +1 mobility diameter). [83] Text

[84]The first "corresponding" is sufficient for both, delete second "corresponding". Perhaps one could add here ", but not true physical diameter for multiply charged particles". Just a thought as to how to make this point clear. [84] Referee

[85] *Thank you for the suggestion.* [85] *Response*

[86] **The latter is the raw response function defined as integrated response downstream of the DMA as a function of upstream voltage (or corresponding z^s or apparent +1 mobility diameter but not true physical diameter for multiply charged particles).** [86] **Revision**

[87] [87] Text

$$\mathbb{M}_k^{\delta_1} = \Pi_k \cdot \left\{ g_0 \cdot \left[T_{size}^{\Lambda,\delta}(k, z^s) * \mathbb{n} \right] \right\} \tag{13}$$

In Eq. (13), $\mathbb{M}_k^{\delta_1}$ evaluates to the apparent +1 mobility distribution particles that exit the DMA$^{\Lambda,\delta}$ at the nominal setpoint-diameter defined by mobility z^s (or z-star) in DMA 1 and particle charge k.

[88]Π_k: Given the appending of superscripts to this function in the following text, it [88] Referee

should appear the same here with (capital lambda)1

[89] $T^{\Lambda,\delta}_{size}(k, z^s)$: The superscripts should have subscript 1 corresponding to the subscript 1 of the left side of the equation. [89] Referee

[90] You say that this is for DMA 1 but there should be an index/subscript of 1 on the superscripts of the first "DMA" in this sentence and of Tsize in the above equation to indicate this. Otherwise, there is no clear relationship between (delta)1 on the left side of the equation and (delta) (no subscript) on the right side. And (capital lambda)1 should also have a subscript of 1. [90] Referee

[91] DMA 1 and 2 which possibly have different geometries, flow rates, and grids, e.g. Λ_1, Λ_2 and δ_1, δ_2. [91] Text

[92] You introduce this notation here but then fail to properly apply it in Eq. (13) and following. [92] Referee

[93] *Thank you for pointing this out. Superscripts have been added in the appropriate places.* [93] *Response*

[94] $\Pi^{\Lambda,\delta}_k$ is the projection of particles having physical diameter D and carrying k charges onto the apparent +1 mobility grid. [94] Text

[95] This function needs to interpolate the diameters of the grown particles onto the +1 mobility diameter grid corresponding to Z. From your description, it sounds like multiply charged particles are mis-sized to smaller diameters. If this function is used with storing the apparent diameter, rather than true diameter, in the distribution, then there would have to be a later function just to undo that before use in Eq. 14. [95] Referee

[96] This function involves both calculation of apparent mobility but also interpolation of the grown bin sizes from Z back onto Z. This interpolation step is important as it affects the propagation of the random error of the input distribution. As such, it should be explicit noted here. In general, any interpolation of a noisy distribution or decedents there of will tend to reduce the overall noise level, but not, I believe, in a very predictable way. [96] Referee

[97] $\Pi^{\Lambda,\delta}_k$ *calculates the apparent +1 diameter of a particle that carries multiple charges. The implementation is quite simple. Start with a physical size e.g. 100 nm. Next compute the mobility of that particle given it's charge state. Next, reinterpret that mobility as if the particle were to carry only a single charge. Finally divide this by the initial diameter to get the projection. Obviously* $\Pi^{\Lambda,\delta}_1 = 1$. *Less obviously* $\Pi^{\Lambda,\delta}_2 < 1$. $\Pi^{\Lambda,\delta}_k$ *does not do any interpolation. In the expression,* $\left\{ g_0 \cdot \left[T^{\Lambda,\delta}_{size}(k, z^s) * \mathbb{n} \right] \right\}$ *evaluates to the grown transmitted size distribution.* $\Pi_k \cdot \left\{ g_0 \cdot \left[T^{\Lambda,\delta}_{size}(k, z^s) * \mathbb{n} \right] \right\}$ *evaluates to the apparent size* [97] *Response*

distribution. The · operator is what shifts the sizes. If a misfit occurs during the shift (which invariably happens), the result in interpolated onto the original size grid of $\mathtt{n}$*. The role of interpolation is now discussed together with the discretization scheme (see further below).*

[98] Also, this function does NOT depend on the DMA configuration (capital lambda)1. It does depend on (delta)1, including the subscript 1. [98] Referee

[99] *It does depend on the DMA configuration, which includes temperature and pressure, because the conversion from mobility to diameter, and hence the projection is temperature and pressure dependent.* [99] *Response*

[100] ...humidifier, $T_{size}^{\Lambda,\delta}(k, z^s)$ is as in Eq. (10), and $\mathtt{n}$ is the mobility size distribution upstream of DMA 1.
To help parse Eq. (13), the product $T_{size}^{\Lambda,\delta}(k, z^s) * \mathtt{n}$ evaluates to... [100] Text

[101] Superscripts should have subscripts 1. [101] Referee

[102] *Fixed* [102] *Response*

[103] Equation (13) differs from that in Petters [2018] where it was assumed that particles of all charges grow by the same amount. This is incorrect. Particles carrying more than a single charge alias at a smaller particle size [Gysel et al., 2009, Shen et al., 2021]. The effect is due to the size dependence of the slip-flow correction factor and captured through the function $\Pi_k^{\Lambda,\delta}$. Equation (13) assumes that g_0 applies to all particle sizes. [103] Text

[104] This does not make sense unless go is a function of particle size, or of zs and k. If this is the case, it has not been made clear. Otherwise, it seems that both here and in Petters it is assumed that particles of all charges grow by the same amount, go, a constant. [104] Referee

[105] *Yes, here and in Petters it is assumed that particles of all charges grow by the same amount, go, a constant. It is said explicitly in " Equation (13) assumes that* g_0 *applies to all particle sizes." The initial wording was rather poor and the text is revised.* [105] *Response*

[106] **Equation (13) differs from that in Petters [2018] where it was assumed that the apparent growth factor for particles carrying multiple charges is the same as for single charged particles. This is incorrect. Particles carrying more than a single charge alias at a smaller particle size [Gysel et al., 2009, Shen et al., 2021]. The effect is due to the size dependence of the slip-flow correction factor and captured** [106] **Revision**

through the function $\Pi_k^{\Lambda,\delta}$. Equation (13) assumes that g_0 applies to all particle sizes.

[107]

$$\mathbf{O}_k = \text{mapreduce}\{z^s \rightarrow [\Omega^{\Lambda_2,\delta_2}(Z, z^s, k). * T_l^{\Lambda_2,\delta_2}(Z, k)]^T, \text{vcat}, Z\} \qquad (15)$$

[107] Text

[108]zs has been used as the centroid mobility for both DMAs. These need to be distinguished as two different parameters. Use subscripts "1" and "2" on these as appropriate to avoid confusion, e.g. here and Eq. 13. There are others in the text as well.

[108] Referee

[109]Is the range of the DMA 2 scan really the same as the range of the input distribution when the former uses 4 times fewer bins? The same Z is used for both.

[109] Referee

[110] *I clarified Z as Zs (same as your earlier comment). Subscript has been added as suggested. The range in Z is arbitrary. Clarification about how the various Z and Zs relate to the discretization has been added to the text (see further below).*

[110] *Response*

[111]**Please see revised section.**

[111] **Revision**

[112] Equations (14) and (15) modified from those in Petters (2018)

[112] Text

[113]"are modified" or "have been modified"

[113] Referee

[114] **Equations (14) and (15) have been modified from those in Petters (2018)**

[114] **Revision**

[115] ... matrix corresponding to singly charged particles and then apply the same matrix ...

[115] Text

[116]Referee suggested: “applied”

[116] Referee

[117] **... matrix corresponding to singly charged particles and then applied the same matrix ...**

[117] **Revision**

[118] If the aerosol is externally mixed, the humidified distribution function is given by ...

[118] Text

[119]"function exiting DMA 2". Otherwise, it sounds like the distribution entering

[119] Referee

DMA 2, right after humidification.

[120] **If the aerosol is externally mixed, the humidified distribution function exiting DMA 2 is given by ...** [120] **Revision**

[121] ... where P_g is the growth factor probability density function ... [121] Text

[122]This still doesn't say anything about the growth factor being a function of dry diameter. So is Pg the same for all dry particle sizes, including those of multiply charged particles? [122] Referee

[123] *Yes, it is. There is explicit discussion later in the text on how to potentially relax this assumption by using 2D inversions.* [123] *Response*

The following few comments are given without direct response. They all relate to the discretization of the grid and the underlying interpolation scheme. In response (see below) the paragraph was revised to clarify. The individual comments referenced here have been taken in to account.

[124] For purposes of the forward model, the mobility grid for DMA 1 is discretized at a resolution of i bins. Transmission through DMA is computed for a specified z_s [124] Text

[125]This is not clear - "DMA 2", "DMA1" or "both DMAs"? From the following text, it would appear that what is needed and being described is the "transmission through DMA1 and subsequent growth". That is, up to the point of entering DMA 2. Note, simply changing "DMA" to "DMA1" does not work as DMA 1 transmission does not depend on go. [125] Referee

[126] If the input size distribution does not match the mobility grid the grids are merged through interpolation. [126] Text

[127]Presumably, the input size distribution bins are interpolated onto diameter bins corresponding to the Z bins. Saying they are "merged" is ambiguous as to which is interpolated onto the other. [127] Referee

[128] 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. [128] Text

[129]It seems to me the DMA 2 mobility grid must be dynamically set according to how much growth there is. This would be good to note here. Wouldn't it be simpler [129] Referee

to just have the DMA 2 grid be a subset of the DMA 1 grid? However, if the setups of the two DMAs are such that they do not have the same non-diffusing resolution, Qa/Qsh, then perhaps it would be better to use grids of different resolutions. As noted before, interpolations tend to smooth the data, thereby confounding the error analysis.

[130] *First, in this framework, the DMA 2 mobility grid is arbitrary and fixed. Interpolation is used throughout. The effect is factored into the framework through the size distribution operators, specifically the · operator. Potential smoothing effects are factored into the framework through the numerical tests.*

*The reason for using interpolation throughout is to ensure generality of the approach. For example, the size distribution used in the forward model may come from a separate SMPS system (or even a model) that comes with binning that is not necessarily known ahead of time. The way we configured our TDMA is to set the voltage/size in DMA 1 denoted as Dd and then perform an SMPS scan over the range, for example, 0.7 * Dd to 2.5 * Dd over 60 s. The flow ratios in DMA 1 and DMA can differ, though we usually keep both 5:1. The bins are constructed as a geometrically stepped mobility grid between the lower and upper range. The only information about DMA is that of the nominal diameter.*

There might clever ways to select the bins in DMA 2 to be a subset of DMA 1, but this does not quite obviate the need for interpolation in the forward model, unless one also matches the allowable growth factor to the discrete bin values. Even then, the growth for particles that carry multiple charges the corresponding mobility will not match that of the discrete binning. The paragraph on discretization includes now text explaining where interpolation is necessary, and where it can be avoided.

[130] *Response*

[131] Reasonable choices are $i = 120$, $j = n = 30$.

[131] Text

[132]The transitions between mobility and diameter with their different natural binning make it difficult to minimize the problems of unpredictable smoothing by interpolations. It seems little can be done about that unless you are willing to set up a universal scale throughout based on either mobility or mobility diameter. Then bin midpoints could be translated to the other parameter and from there to all other scales. Each scale should either match the universal resolution and e midpoints or use an integer multiple of the resolution (e.g. each midpoint in one scale matches every third midpoint in another scale). If this approach were used, it would seem best to use a universal scale with uniform increments in either ln(dp) or ln(z) as appropriate. I believe that if the universal scale matched that of Pg and with uniform increments in ln(g) or ln(dp), then grown particles from one bin would land exactly into another bin with no interpolation or fractional bin calculation required.

[132] Referee

[133] *Please see response to previous comment and text below. Interpolation is deeply interwoven in into this framework. Since the binning along all dimensions is arbitrary, it is possible to setup a universal (or near universal) grid in which interpolation is minimized, which is*

[133] *Response*

now mentioned. The paragraph describing the discretization is revised as follows.

[134] **Revision**

[134] **For purposes of the forward model, the mobility grid for DMA 1 is discretized at a resolution of i bins by specifying the Z vector in Eq. (10). If the Z vector does not match that of the aerosol size distribution $\mathbb{n}$, the size distribution bins are interpolated onto the diameter bins corresponding to the Z bins. Transmission through DMA 1 is computed for a specified z^s (the dry mobility) and g_0 (the growth factor) via Eq. (13). The resulting $\mathbb{M}_k^{\delta_1}$ lie on the same Z grid with i bins. Any mismatches between the apparent growth factor and the underlying Z grid are resolved via interpolation implicit in the $\cdot$ operator. ($f \cdot \mathbb{n}$ is the uniform scaling of the diameter field of the size distribution by factor f. If the resulting diameters are off the original diameter grid, the result in interpolated onto the grid defined within $\mathbb{n}$)**

The mobility grid for DMA 2 is represented by the vector $Z_{s,2}$ in Eq. (15) and discretized at a resolution of j bins over a custom mobility range. If the vector Z inside the square bracket of Eq. (15), $[\Omega^{\Lambda_2,\delta_2}(Z, z^s, k) . * T_l^{\Lambda_2,\delta_2}(D_{p,1})]$ equals that of DMA 1, the matrix is non square. The product $\mathbf{O}_k * \mathbb{M}_k^{\delta_1}$ will map the i bins from DMA 1 to the j bins in DMA 2. Alternatively, if the Z vector inside the square bracket of Eq. (15) is taken to be equal to $Z_{s,2}$, the matrices $\mathbf{O}_k$ are square. 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 $\mathbf{O}_k * \mathbb{M}_k^{\delta_1}$. The advantage of interpolation is that the the matrices $\mathbf{O}_k$ are smaller.

The forward model, defined by Eq. (14) can be evaluated for arbitrary g_0 values. Thus the growth factor probability distribution P_g in Eq. (17) can be discretized into n arbitrary growth factor bins. A natural choice is to accept growth factor values that coincide with the mobility grid of DMA 2, i.e. the bins align with $g = D_{p,1}/D_d$, where D_d is the nominal diameter selected by DMA 1 and $D_{p,1} = D_p(z, k = 1)$ and z is an element of $Z_{s,2}$. However, this is not required for evaluating Eq. (17). Equation (17) is cast into matrix form such that the humidified mobility distribution function is given by

$$\mathbb{m}_t^{\delta_2} = \mathbf{B} P_g + \epsilon \tag{18}$$

where the matrix B is understood to be computed for a specific input aerosol size distribution, and ϵ is a vector that denotes the random error that may be superimposed as a result of measurement uncertainties. 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 choice of i, j, n, the ranges of mobility grids for DMA 1, DMA 2, and the range of P_g is only constrained by computing resources and a physically reasonable representation of the problem domain. Reasonable choices are $i = 120$, $j = n = 30$. The size of B is $j \times n$. Uncertainties in the size distribution propagate into B. The main influence of the error will be the relative fraction of +1, +2, and +3 charged particles. Assuming a random error of $\pm$20% in concentration, the overall effect on $\mathbb{m}_t^{\delta_2}$ is expected to be small.

Note that interpolation is widely used in this framework. Interpolation may affect how errors propagate through the model. Interpolation in Eq. (13) is unavoidable. However, interpolation can be minimized by working with non-square $\mathbf{O}_k$ and matching the grid of P_g to that of DMA 2. Informal tests working with different binning schemes suggests that the influence of interpolation choices on the final result is smaller than typical experimental errors.

[135]

$$\mathbb{m}_t^{\delta_2} = \mathbf{A}_2 P_g + \epsilon \tag{18}$$

where the subscript 2 specifies transmission through DMA 2, the matrix $\mathbf{A}_2$ is understood to be computed for a specific input aerosol size distribution

[135] Text

[136]The definition of this **A** is not analogous to that in Eq. 11; this one subsumes the input distribution Mk(delta)1. To avoid confusion, use a different letter.

[136] Referee

[137] *Done - see above.*

[137] *Response*

[138] is expected to small.

[138] Text

[139] *Done - see above.*

[139] *Response*

Revised Section 2.3

Design Matrices For Differential Mobility Analyzers

Differential mobility analyzers consist of two electrodes held at a constant- or time-varying electric potential. Cylindrical [Knutson and Whitby, 1975] and radial [Zhang et al., 1995, Russell et al., 1996] electrode geometries are the most common. Charged particles in a flow between the electrodes are deflected to an exit slit and measured by a suitable detector, usually a condensation particle counter. The fraction of particles carrying k charges is described by a statistical distribution that is created by the charge conditioner used upstream of the DMA. The functions governing the transfer through bipolar charge conditioners, single DMAs, and tandem DMAs are well understood [Knutson and Whitby, 1975, Rader and McMurry, 1986, Reineking and Porstendörfer, 1986, Wang and Flagan, 1990, Stolzenburg and McMurry, 2008, Jiang et al., 2014].

The traditional mathematical formulation of transfer through the DMA is summarized in Stolzenburg and McMurry [2008] and references therein. Briefly, the integrated response downstream of the DMA operated at voltage V_1 is given by a single integral that includes a summation over all selected charges. The size distribution is measured by varying voltage V_1, which produces the raw response function defined as integrated response downstream of the DMA as a function of upstream voltage. The size distribution is found by inversion. The basic mathematical problem associated with inverting the response function to find the size distribution is summarized by Kandlikar and Ramachandran [1999]. The integral is discretized by quadrature to find the design matrix that maps the size distribution to the response function. L_2 regularization is one of several methods to reconstruct the size distribution from the response function [Voutilainen et al., 2001, Kandlikar and Ramachandran, 1999].

The integrated response downstream of a tandem DMA that is operated at voltages V_1 and V_2 requires solving integrals of the upstream particle size distribution over size and the grown particle size distribution over size. The integration must be repeated for each charge state. Scanning over a range of voltages V_2 results in the raw TDMA response function. 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.

Petters [2018] introduced a computational approach to model transfer through the DMA. The main idea of the approach is to provide a domain specific language comprising a set of simple building blocks that can be used to algebraically express the response functions intuitively through a form of pseudo code. The main advantage of this approach is that the expressions simultaneously encode the theory governing the transfer through the DMA and the algorithmic solution to compute the response function. The resulting expressions are concise. They are easily identified within actual source code when working through the examples provided with the package documentation. This makes the code easily modifiable by non-experts to change existing terms or add new convolution terms without the need to develop algorithms.

A disadvantage of the computational approach over the traditional mathematical approach is that computation lacks standardization of notation. This can blur the line between general pseudo code and language specific syntax. Some of the applied computing concepts may be less widely known when compared to standard mathematical approaches. Nevertheless, the author believes that the advantages of the computational approach outweigh the drawbacks. Therefore, this work

builds upon the expressions reported in Petters [2018]. Updates and clarifications to the earlier work are noted where appropriate.

The computational language includes a standardized representation of aerosol size distributions, operators to construct expressions, and functions to evaluate the expressions. Size distributions are represented as a histogram and internally stored in the form of the *SizeDistribution* composite data type. Composite data types combine multiple arrays into a single symbol for ease of use, thus facilitating faster experimental design and analysis. The size distribution data type *SizeDistribution* includes vectors of the selected mobility bins considered by the DMA, +1 mobility diameter bin edges and +1 mobility diameter bin midpoints computed from the mobility grid, number concentration, log-normalized spectral density, and logarithmic bin widths. *SizeDistributions* are denoted in blackboard bold font (e.g., $\mathbb{n}$, $\mathbb{r}$, etc.). *SizeDistributions* are the building block of composable algebraic expressions through operators that evaluate to transformed *SizeDistributions*. For examples, $\mathbb{n}_1 + \mathbb{n}_2$ is the superposition of two size distributions and $f * \mathbb{n}$ is the uniform scaling of the concentration fields by factor f, $\mathbf{A} * \mathbb{n}$ is matrix multiplication of $\mathbf{A}$ and concentration fields of the size distribution, and $f \cdot \mathbb{n}$ is the uniform scaling of the diameter field of the size distribution by factor f, and $T \cdot \mathbb{n}$ is the elementwise scaling of the diameter field by factor T. (Note that Petters (2018) used $T. \cdot \mathbb{n}$ as the elementwise scaling. The extra dot has been dropped to stay consistent with the current software implementation).

Generic functions are used to evaluate expressions. The function $\sum(f, m)$ evaluates the function $f(x)$ for $x = [1, \ldots, m]$ and sums the result. If $f(x)$ evaluates to a vector, the sum is the sum of the vectors. The function map(f, x) applies $f(x)$ to each element of vector x and returns a vector of results in the same order. The function foldl(f, x) applies the bivariate function $f(a, x)$ to each element of x and accumulates the result, where a represents the accumulated value. For the first element in x, a is the neutral value. For example foldl$(-, [1, 2, 3])$ evaluates the function $-(a, x)$ and yields $1 - 2 - 3 = -4$. The function mapfoldl(f, g, x) combines map and foldl. It applies function f to each element in x such that $y = f(x)$ and then reduces the result using the bivariate function function $g(a, y)$ where a represents the accumulated value. For the first element $f(x)$, a is the neutral value. For example, mapfoldl(sqrt, $-$, $[4, 16, 64]$)evaluates to foldl$(-, [2, 4, 8]) = 2 - 4 - 8 = -10$. The function $vcat(x, y)$ concatenates arrays x and y along the first dimension in Julia. However, other programming languages may concatenate along a different dimension as definition of horizontal and vertical is arbitrary. Anonymous functions are used as arguments to reducing functions. Anonymous functions are denoted as $x \rightarrow expression$, where x is the argument consumed in the evaluation of the $expression$. These functions are generic and represent widely used computing concepts. They are implemented in most modern programming languages.

DMA geometry, dimensions, and configuration are abstracted into composite types Λ (configuration comprising flow rates, power supply polarity, and thermodynamic state) and δ (DMA domain defined by a mobility/size grid). Each DMA is fully described by a pair Λ,δ. Subscripts and superscripts are used to distinguish between different configurations in chained DMA setups, e.g. δ_1 and δ_2 denoting the first and second DMA, respectively. Application of size distribution expressions to transfer functions constructs a concise model of the transmitted DMA mobility distribution, denoted as the DMA response function. Implementation of the language is distributed through a freely-available and independently documented package *DifferentialMobilityAnalyzers.jl*, written in the Julia language. Expressions in the text are provided in general mathematical form for readability.

Petters [2018] gives a simple expression that model transfer through the DMA. The function $T_{size}^{\Lambda,\delta}(k, z^s)$ evaluates to a vector representing the fraction of particles carrying k charges that exit $\text{DMA}^{\Lambda,\delta}$ as a function of mobility

$$T_{size}^{\Lambda,\delta}(k, z^s) = \Omega(Z, z^s/k, k). * T_c(k, D_{p,1}). * T_l(D_{p,1}) \tag{10}$$

where z^s is the centroid mobility selected by the DMA (determined by the voltage and DMA geometry), Z is a vector of particle mobilities, Ω is the diffusing DMA transfer function [Stolzenburg and McMurry, 2008], T_c is the charge frequency distribution [Wiedensohler, 1988], and T_l is the diameter-dependent penetration efficiency [Reineking and Porstendörfer, 1986]. The diameter $D_{p,1} = D_p(z, k = 1)$, where z is an element of Z. The function Ω has been updated from Petters (2018). The version in Petters (2018) computed the shape of the transfer function for the mobility diameter corresponding to singly charged particles and then applied the same shape of the transfer function and diffusional loss to the multiply charged particles. The functional Ω depends on three arguments $\Omega(Z, z^s, k)$ [Stolzenburg and McMurry, 2008]. The charge state is used to compute the diffusion coefficient and thus account for diffusional losses and broadening of the transfer function for multiply charged particles.

The output of $T_{size}^{\Lambda,\delta}(k, z^s)$ 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 the size of 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 $T_l(D_{p,1})$ are evaluated at the correct diameter. The function $T_{size}^{\Lambda,\delta}(1, z^s)$ evaluates to a vector of the same length as Z. Performing an elementwise sum over all $T_{size}^{\Lambda,\delta}(k, z^s)$ produces the net mobility distribution transmitted by the DMA. Examples for $T_{size}^{\Lambda,\delta}(1, z^s)$, $T_{size}^{\Lambda,\delta}(2, z^s)$, and $T_{size}^{\Lambda,\delta}(3, z^s)$ is shown in Figure 2, right panel in Petters (2018). Note that Eq. (10) can be evaluated using arbitrarily discretized Z vectors.

Petters [2018] also gives an expression that evaluates to the convolution matrix for passage through a single DMA that is valid in the context of size distribution measurement system, e.g. SMPS. Since the expression includes a summation over all charges, the information on particle physical diameter of multiply charged particles is lost.

$$\mathbf{A} = \text{mapfoldl}\{z^s \to \Sigma[k \to T_{size}^{\Lambda,\delta}(k, z^s), m]^T, \text{vcat}, Z_s\} \tag{11}$$

where, m is the upper number of multiply charged particles, ^{T} is the transpose operator, and is a vector of centroid mobilities scanned by the DMA. The matrix is square if $Z_s = Z$ in Eq. 10. However, this is not a necessary restriction. Eq. (11) evaluates to the same as Eq. (8) in Petters (2018), but the notation is revised to be more general by removing the Julia-specific splatting construct and replacing it with more widely used generic functions.

To help with parsing the expression, $T_{size}^{\Lambda,\delta}(k, z^s)$ evaluates to a vector of transmission for k charges and set point centroid mobility z^s as a function of the entire mobility grid (e.g. 120 bins discretized between mobility z_1 and z_2). The function $\Sigma[k \to T_{size}^{\Lambda,\delta}(k, z^s), m]$ superimposes the vectors for all charges. Mapping $z^s \to \Sigma[k \to T_{size}^{\Lambda,\delta}(k, z^s), m]$ over the centroid mobility grid Z_s produces an array of vectors, each corresponding to the transmission for a single size bin. Transposing the vectors and reducing the collection through concatenation produces the design matrix that links the mobility

size distribution to the response function, i.e.

$$\mathbb{r} = \mathbf{A}\mathbb{n} + \epsilon \tag{12}$$

where $\mathbb{r}$ is the response distribution, $\mathbb{n}$ is the true mobility size distribution, and ϵ is a vector denoting the random error that may be superimposed as a result of measurement uncertainties. By design $\mathbb{n}$ and $\mathbb{r}$ are *SizeDistribution* objects, which represent the distribution as a histogram in both spectral density units (dN/dlnD) and concentration per bin units. The latter is the raw response function defined as integrated response downstream of the DMA as a function of upstream voltage (or corresponding z^s or apparent +1 mobility diameter but not true physical diameter for multiply charged particles). Note, however, that the response function is not a true particle size distribution in the scientific sense since information about multiply charged particles is lost. The representation of $\mathbb{r}$ as *SizeDistribution* object is to allow response functions to used in the expression-based framework used here.

The mobility distribution exiting DMA 2 in the humidified tandem DMA is evaluated using the expression

$$\mathbb{M}_k^{\delta_1} = \Pi_k^{\Lambda_1,\delta_1} \cdot \left\{ g_0 \cdot \left[T_{size}^{\Lambda_1,\delta_1}(k, z^s) * \mathbb{n} \right] \right\} \tag{13}$$

In Eq. (13), $\mathbb{M}_k^{\delta_1}$ evaluates to the apparent +1 mobility distribution particles that exit the DMA$^{\Lambda_1,\delta_1}$ at the nominal setpoint-diameter defined by mobility z^s (or z-star) in DMA 1 and particle charge k. Subscripts are used to differentiate DMA 1 and 2 which possibly have different geometries, flow rates, and grids, e.g. Λ_1, Λ_2 and δ_1, δ_2. $\Pi_k^{\Lambda,\delta}$ is the projection of particles having physical diameter D and carrying k charges onto the apparent +1 mobility grid. It is a function that converts each diameter/charge pair to mobility and interprets the result as apparent +1 mobility diameter. $g_0 = D_{wet}/D_{dry}$ is the true diameter growth factor, D_{dry} is the selected diameter by DMA 1, D_{wet} is the diameter after the humidifier, $T_{size}^{\Lambda_1,\delta_1}(k, z^s)$ is as in Eq. (10), and $\mathbb{n}$ is the mobility size distribution upstream of DMA 1.

To help parse Eq. (13), the product $T_{size}^{\Lambda,\delta}(k, z^s) * \mathbb{n}$ evaluates to the transmitted mobility distributions of particles carrying k charges at the set-point mobility z^s in DMA 1. The size distribution is grown by the growth factor g_0. The resulting size distribution is shifted to the apparent +1 mobility diameter using $\Pi_k^{\Lambda,\delta}$. Equation (13) differs from that in Petters [2018] where it was assumed that particles that the apparent growth factor for particles carrying multiple charges is the same as for single charged particles. This is incorrect. Particles carrying more than a single charge alias at a smaller particle size [Gysel et al., 2009, Shen et al., 2021]. The effect is due to the size dependence of the slip-flow correction factor and captured through the function $\Pi_k^{\Lambda,\delta}$. Equation (13) assumes that g_0 applies to all particle sizes.

The total humidified mobility distribution $\mathbb{m}_t^{\delta_2}$ exiting DMA 2 is given by

$$\mathbb{m}_t^{\delta_2} = \sum_{k=1}^{m} \left(\mathbf{O}_k * \mathbb{M}_k^{\delta_1} \right) \tag{14}$$

where, m is upper number of charges on the multiply charged particles and

$$\mathbf{O}_k = \text{mapfoldl}\{z^s \rightarrow [\Omega^{\Lambda_2,\delta_2}(Z, z^s, k). * T_l^{\Lambda_2,\delta_2}(D_{p,1})]^T, \text{vcat}, Z_{s,2}\} \tag{15}$$

is the convolution matrix for transport through DMA 2 and particles carrying k charges. In Eq. (15), $Z_{s,2}$ is a vector of centroid mobilities scanned by DMA 2. Equations (14) and (15) have been modified from those in Petters (2018) in the following manner. The convolution matrix $\mathbf{O}_k$ is computed individually for each charge. The version in Petters (2018) computed the matrix corresponding to singly charged particles and then applied the same matrix to multiply charged particles. Since $\mathbf{O}_k$ is now charge resolved, it is moved into the summation in Eq. (14). Computation of $\mathbf{O}_k$ through Eq. (15) has been revised to be more general by removing a Julia-language specific construct. $\mathbf{O}_1$ computed by Eq. (15) produces the same matrix as in Petters (2018).

If the aerosol is externally mixed, the humidified distribution function exiting DMA 2 is given by

$$\mathbb{m}_t^{\delta_2} = \int_0^\infty P_g * \left[\sum_{k=1}^{m} \left(\mathbf{O}_k * \mathbb{M}_k^{\delta_1} \right) \right] dg_0 \tag{16}$$

where P_g is the growth factor probability density function and the diameters in $\mathbb{M}_k^{\delta_1}$ are normalized by D_{dry}. $\mathbb{m}_t^{\delta_2}$ in Eq. (16) is the forward model through the tandem DMA. Using the notation in section 2.2,

$$F(\mathrm{x}, \mathrm{c}) = \int_0^\infty P_g * \left[\sum_{k=1}^{m} \left(\mathbf{O}_k * \mathbb{M}_k^{\delta_1} \right) \right] dg_0 \tag{17}$$

where x is the true P_g and the vector c of constraining parameters comprises the DMA setup $\Lambda_1, \Lambda_2, \delta_1, \delta_2$ and upstream size distribution $\mathbb{n}$. Computer code that creates a forward model for tandem DMAs has been added to the *DifferentialMobiltyAnalyzers.jl* package and is annotated in the documentation of the package.

For purposes of the forward model, the mobility grid for DMA 1 is discretized at a resolution of i bins by specifying the Z vector in Eq. (10). If the Z vector does not match that of the aerosol size distribution $\mathbb{n}$, the size distribution bins are interpolated onto the diameter bins corresponding to the Z bins. Transmission through DMA 1 is computed for a specified z^s (the dry mobility) and g_0 (the growth factor) via Eq. (13). The resulting $\mathbb{M}_k^{\delta_1}$ lie on the same Z grid with i bins. Any mismatches between the apparent growth factor and the underlying Z grid are resolved via interpolation implicit in the $\cdot$ operator. ($f \cdot \mathbb{n}$ is the uniform scaling of the diameter field of the size distribution by factor f. If the resulting diameters are off the original diameter grid, the result in interpolated onto the grid defined within $\mathbb{n}$).

The mobility grid for DMA 2 is represented by the vector $Z_{s,2}$ in Eq. (15) and discretized at a resolution of j bins over a custom mobility range. If the vector Z inside the square bracket of Eq. (15), $[\Omega^{\Lambda_2,\delta_2}(Z, z^s, k). * T_l^{\Lambda_2,\delta_2}(D_{p,1})]$ equals that of DMA 1, the matrix is non square. The product $\mathbf{O}_k * \mathbb{M}_k^{\delta_1}$ will map the i bins from DMA 1 to the j bins in DMA 2. Alternatively, if the Z vector inside the square bracket of Eq. (15) is taken to be equal to $Z_{s,2}$, the matrices $\mathbf{O}_k$ are square. 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 $\mathbf{O}_k * \mathbb{M}_k^{\delta_1}$. The advantage of interpolation is that the the matrices $\mathbf{O}_k$ are smaller.

The forward model, defined by Eq. (14) can be evaluated for arbitrary g_0 values. Thus the growth factor probability distribution P_g in Eq. (17) can be discretized into n arbitrary growth factor bins. A natural choice is to accept growth factor values that coincide with the mobility grid of DMA 2, i.e. the bins align with $g = D_{p,1}/D_d$, where D_d is the nominal diameter selected by DMA 1 and $D_{p,1} = D_p(z, k = 1)$ and z is an element of $Z_{s,2}$. However, this is not required for evaluating Eq.

(17). Equation (17) is cast into matrix form such that the humidified mobility distribution function is given by

$$\mathbb{m}_t^{\delta_2} = \mathbf{B} P_g + \epsilon \tag{18}$$

where the matrix $\mathbf{B}$ is understood to be computed for a specific input aerosol size distribution, and ϵ is a vector that denotes the random error that may be superimposed as a result of measurement uncertainties. 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 choice of i, j, n, the ranges of mobility grids for DMA 1, DMA 2, and the range of P_g is only constrained by computing resources and a physically reasonable representation of the problem domain. Reasonable choices are $i = 120$, $j = n = 30$. The size of $\mathbf{B}$ is $j \times n$. Uncertainties in the size distribution propagate into $\mathbf{B}$. The main influence of the error will be the relative fraction of +1, +2, and +3 charged particles. Assuming a random error of $\pm$20% in concentration, the overall effect on $\mathbb{m}_t^{\delta_2}$ is expected to be small.

Note that interpolation is widely used in this framework. Interpolation may affect how errors propagate through the model. Interpolation in Eq. (13) is unavoidable. However, interpolation can be minimized by working with non-square $\mathbf{O}_k$ and matching the grid of P_g to that of DMA 2. Informal tests working with different binning schemes suggests that the influence of interpolation choices on the final result is smaller than typical experimental errors.

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