

Thank you very much for the critical review, especially the respond to the mathematical elements. It has been a pleasure answering certain questions. Also thank you for the nice words to the style of writing.

## General comment

We want to point out and specify more precisely our motivation! The aim was a fast and lightweight algorithm but with sufficient precision for atmospheric conditions (with a minimum assumptions). However, this framework is with extended functionality and a larger output (e.g. error propagation). Therefore we've changed the title and try to emphasize this more in the text.

This algorithm is not a competitor to the solution of sets of Fredholm integral equations, due to total different priorities!

## Specific comments

At the beginning we have focused on the three major points. Depending on the case, we will already answer minor issues.

### Nomenclature

*p. 4741, lns 7-8: As the parameter  $n$  is already being used as the number of elementary charges on a particle, a different notation for particle concentration should be used here.*

The concentration is now replaced by  $c$  (consequently  $dc/dZ$ , ...)

*[...] As used here, the only way to make sense of the equations given is to define the electric mobility as the product of the particle mobility and a single elementary charge regardless of the actual number of elementary charges,  $n$ , on the particle. Without this altered definition, Eq. (7) makes no sense. [...]*

You are absolutely right! It is now explicitly pointed out, that  $Z$  has to be considered all the time as the electrical mobility for singly charged particles.

*The authors should keep in mind at all times that  $Z'$  and  $Z$  are two quite distinct parameters. The first is a property of a particle while the second represents the centroid of the DMA transfer function and is only a property of the instrument parameters. [...]*

For convolution, the variable with the " ' " is all the time the variable used for integration. That means, in this special case,  $Z$  is the centroid of the TF, shown in Fig. 1. and also written in the text:

*The transfer function  $h^{dma}$  of particles through a DMA at the position  $Z$ , can be simply described by a triangular function [...]*

Units etc.

*The second issue is the lack of attention to dimensions (units) leading to some incorrect equations.*

*As far as units are concerned, there is a discrepancy in these in the case of the first and second lines of Eq. (10). Both  $f$  and  $f^*$  have units of concentration and cancel each other out. From the definition in Eq. (9), it is seen that  $E$  is dimensionless [...]*

At a certain point you are absolutely right! But, the problem is quite more complex!

The equations are almost correct, with the exception that we have named the triangular function  $h^{dma}$ , which is confusing. There is also no reason why the equation shall be essential wrong, otherwise the good comparison to the other inversion routines would be just coincidence.

Using the definition of a transfer function for convolution, which is preserving the unit, everything is consistent. This implies, that the pure integral over the transfer function is dimensionless! Or in other words, in this special case, the unit of the transfer function should be the inverse of the electrical mobility.

With this in mind, the definition of the triangular DMA transfer and the physic behind is different from that. Calculating the particle concentration [number per volume], with a specific PNSD [number per volume and diameter] with a DMA at Position  $Z$ , downstream, is not a convolution integral, preserving the unit.

So let us split the Problem, calculating the concentration downstream of the DMA. Afterwards let us try to find an expression which fulfil the "style" of the convolution, preserving the unit.

Hopefully, this will clarify also this part:

*p. 4741, lns 7-8: [...] Also, the function of  $C_i$  is probably better understood by noting that  $f^*$  has the units of  $dn/d\ln Z$  which are converted to  $dn/d\log D_p$  for  $f$  using  $C_i$ . This has nothing to do with any approximation by Stratmann et al. (1997).*

The simple expression for calculating the concentration downstream of the DMA (single charged):

$$N(Z) = \int_{-\infty}^{\infty} \frac{dn}{dZ'}(Z') f(Z, Z') dZ' \quad (1)$$

With the Approximation, that the PNSD should be almost constant in the range of the DMA TF.

$$N(Z) \simeq \frac{dn}{dZ}(Z) \int_{Z(1-\beta)}^{Z(1+\beta)} f(Z, Z') dZ' \quad (2)$$

$$N(Z) \simeq \frac{dn}{dZ}(Z) \cdot \alpha\beta Z = \frac{dn}{d\ln Z}(Z) \cdot A(Z) \quad (3)$$

But this implies:

$$N(Z) = \frac{dn}{dlnZ}(Z) \cdot A(Z) = \int_{-\infty}^{\infty} \frac{dn}{dlnZ'}(Z')A(Z')\delta(Z - Z')dZ' \quad (4)$$

So we obtain:

$$h^{dma} = A(Z')\delta(Z - Z')dZ' \quad (5)$$

The units of  $N$  (or  $f^*$  in the paper) and  $dn/dlnZ$  (or  $f$  in the paper) are  $1/cm^3$ . This leads to the result that the founded transfer function has the inverse unit of  $Z$ .

Furthermore this is necessary, because:

$$\int_{-\infty}^{\infty} \delta(Z)dZ == 1 \quad (6)$$

summary:

- while calculating the concentration downstream of the DMA on  $Z$  scaled axis has nothing to do with convolution, preserving the units, on the  $ln$ -scaled axis it is possible to find such expression
- it was absolutely wrong to suggest this, by naming the defined triangular function  $h^{dma}$  (Eq. 4)
- one have to keep that the transfer functions, respectively the dirac delta function, have the inverse unit of  $Z$ .
- all in all, it isn't an error, noticeable in a wrong result

So we have renamed the defined triangular function  $g(Z, Z')$ , consequently also the figure. The rest, we think, would just confuse the reader.

also suitable at this point:

***The total transfer function is not a convolution but merely a product of the DMA transfer function (Eq. 5) and the charging probability (Eq. 6a,b). Eq. (7) already shows this product except for the factor  $A(Z/n)$ .***

No, it is convolution! Otherwise, it was a waste of time, because now you can play with the transfer function, e.g. adding additional physics like the activation in a CCNC (see Appendix)!

$$f^*(z) = \int \left( \int f(x)h^{dma}(y-x)dx \right) h^{cha}(z-y)dy \quad (7)$$

$$f^*(z) = \int (C_i(y) \cdot A(y) \cdot f(y)) h^{cha}(z-y)dy \quad (8)$$

$$f^*(z) = \sum C_i(z/n) \cdot A(z/n) \cdot p(\dots z/n \dots) f(z/n) \quad (9)$$

But we admit, it is quite tricky, because you have to be careful with the associativity, due to non scalar factors  $A, C_i, \dots$

This is also physical plausible, because diffusional effects, CPC counting efficiency, etc. are a function of the "true" diameter not the eq. electrical mobility diameter (in case of multiple charges).

In the article, we don't want to go to much into detail.

## aerodynamic shape factor

*p. 4754, Eq. (A2): The factor  $\chi(D_{pve})$  in the denominator on the right side is apparently the shape factor. This needs to be defined or explained.*

We are aware, that there is a lack of information. We have added a few sentences as explanation, as well as the used transformation formulas and a reference for characteristic values (see also the reply to Referee 1).

*For the enhanced inversion using APS or OPC data, these would be given in terms of aerodynamic-equivalent or optically-equivalent particle diameters, respectively, with their own distinct associated shape factors to relate them back to  $D_{pve}$ .*

This is right! A reference diameter (e.g.  $D_{pve}$ ) is necessary, especially for the enhanced mode (overlap range). While this is easy for S/DMPS and APS, such approach is complex for S/DMPS and OPS.

But this issue, to convert consistently different equivalent diameters due to the morphology, is much more difficult than this multiple charge inversion. It would go beyond the scope!

## miscellaneous

*page 4738, lines 9-13: In essence, this passage reads as 'We won't try to solve this for the most accurate solution, so we are left with the most direct solution. [...]*

Thanks for deducing exactly our motivation! (see general comment)

Well, this is a little bit a philosophical question. We don't have to discuss that when the number of sampling points after inversion is smaller than the raw input, there is a lost of information (in size resolution).

It is a little bit tricky (difficult to show), if using the approximation of the narrow DMA transfer function. It would enforce that the EPMD can be interpolate, which is not a prerequisite for this algorithm! But increasing the number of grid points by interpolation won't increase the information content!

We want to point out, this statement is just valid using this approximation!

In our opinion, if you want to have higher Resolution PNSD, you have to measure it. Everything else seems to be misleading. We give this opinion, also for any other measurements!

Perhaps, a natural smoothing spline interpolation for the EPMD would be a good compromise, especially for reducing the noise. But the fact is the same, any kind of interpolation won't increase the information content!

However, the performance of this algorithm is based on this direct solution, whereby the information content is preserved!

*p. 4738, ln 14: The acronym DMA has not been defined.*

We have added the definition of the DMA.

*p. 4738, lns 15-17: Is there an unstated assumption of equal aerosol flows here? Even with that, the representation of the DMA transfer function as a generic triangle [...]*

We have added the citation Birmilie et al (1997). The height, width and following the area are not constant over the whole range, due to diffusional broadening!

*The two-region definition of the DMA transfer function in this region is superfluous. [...] Also, the signs are wrong within the second absolute value. [...]*

We didn't change the two-region definition. But you are right, we have corrected the typing error

*pp. 4739-4740: Eq. (6a) should come directly after the first sentence of section 2.2. Line 9 of page 4740 should then immediately follow Eq. (6a).*

We have rearranged this section. (see Referee 1)

*The acronyms LDMA and CPC have not been defined.*

It should have been DMA! And we have added the definition of the CPC.

*p. 4742, Eq. (12): Include the range of  $Z_i$  given in the following line at the top of the next page in the same line as the equation as is done for later equations.*

The range is now in the same line!

*p. 4745, Eq. (18): Note that  $Z$  decreases as particle size increases. Thus, the last condition in each line on  $Z_i/n$  should have the sign reversed. That is,  $Z_i/n \geq Z_mN$  for the first line and  $Z_i/n \leq Z_mN$  for the second line. This also means that all of the differences in mobilities in the numerators and denominators of the ratios in Eqs. (12, 13, 18, 20 and 21) are negative. Since the sign changes in both the numerator and denominator of each ratio cancel each other, these equations are nevertheless technically correct as written. However, in the ranges of applicability for these same equations it is*

*unconventional, if not actually incorrect, to right them as (Zhigh , Zlow).*

and

*p. 4749, Eq. (33): The inequality/comparison operators should be reversed.*

That's right. It is a general mistake, a rudiment of a solution directly with volume equivalent diameter (skipping  $Z$ ), but which was to confusing for the reader. We have changed the range by  $(Z_j, Z_{j-1})$  etc. (lower index as the maximum value, etc.) and also reversed the signs.

*pp. 4745-4746, Eqs. (18 and 21): The multiple conditions placed on each line would probably be more readable if the "and" symbol were offset [...]*

We have enlarged the offset.

*p. 4746, Eq. (21): For the ratio of mobility differences in the first line on the right side, there is a missing superscript "a" on the second Z in the numerator and that same superscript should replace the incorrect one on the second Z in the denominator. [...]*

The typing errors are corrected!

*p. 4751, lns 16-18: Previous field studies corroborating this statement about realistic values must actually be cited here.*

We have noticed a considerable typing error: the density has to be 2.8 and the shape 1.1 for this specific plot. Furthermore, we have added also the citation of Schladitz et al (2009) once more.

*p. 4751, lns 23-27 and Fig. 4a: Any uncertainties or variations in the instrument aerosol flow are not likely to be uncorrelated from one channel to the next in the measured size distribution. [...]*

Absolutely right! So we have removed this example! It is now just named fictive error.

But i think this fact is too good to reject, just because there is no strict physical example. However, except for the enhanced inversion, uncertainties or variations in the instrument aerosol flow for different devices can be assumed as uncorrelated.

Furthermore we have written (3.3.):  
*The idea [...] is expandable [...] in case of correlated sampling points.*

Here the equation for correlated data:

$$\text{Var}(f_i) = \sum_{j,k=1}^N \text{Cov}(l_{ij}f_j^*, l_{ik}f_k^*) \quad (10)$$

$$\text{Var}(f_i) = \sum_j^N l_{ij} \sum_k^N l_{ik} \text{Cov}(f_j^*, f_k^*) \quad (11)$$

For uncorrelated data  $\text{Cov}(f_j^*, f_k^*) = 0 \forall j \neq k$  this leads to:

$$\text{Var}(f_i) = \sum_j^N l_{ij}^2 \text{Var}(f_j^*) \quad (12)$$

which can be rewritten as a matrix operation (see article):

$$\text{Var}(\mathbf{f}) = \mathbb{L}^{var} \cdot \text{Var}(\mathbf{f}^*) \quad (13)$$

We have added the equation for correlated data!

*[...] This alone would argue against using any spline interpolation of greater order than linear as the higher orders would most likely be fitting noise rather than any true feature of the actual size distribution. [...] Given these conditions, it is also very unlikely that trying to incorporate the true finite width of the transfer function into the inversion routine would improve the accuracy of the final inverted distribution. [...]*

You are right! in a certain way this was already mentioned, but perhaps badly formulated/bad wording  
*it must be noted that these methods have their own disadvantages, because they need proper initial and boundary conditions, and might also lead to artificial overshooting values in some places.*

For both improvements, it would increase the noise, while, of course, a higher interpolation seems to be unnecessary for high size resolution, and are in no proportion.

Also a natural smoothing spline for EPMD is towards the unwanted minimization problem.

Any improvement on accuracy would directly enforce the need of some proper constraints. We have added this! It is now explicitly mentioned, also as a demarcation and limitation to statistical approaches.

***p. 4756, Eq. (C2): In the text at line 5, page 4742, it is indicated that the CPC efficiency would be combined with the total efficiency E. In this equation they have been written separately. There can be a number of other efficiencies to be considered such as penetration efficiency of the plumbing. These should all be combined into E.***

We've added the common CPC efficiency explicitly in (Eq. 9) As you already mentioned, in the Appendix, we wanted give an "unusual" example, in case using a CCNC, the efficiency can be easily extended by the size depended

activation fraction.

*p. 4759, Table 1: There were two errors in the table of coefficients for Wiedensohler’s (1988) fit in the original publication. Those have not been corrected here. The correct values are  $a_4(+1) = -0.1553$  and  $a_5(0) = -0.0105$ .*

Corrected! (see Wiedensohler, 2012)

*p. 4762, Fig. 2: The very thin lines of the plotted curves make it very difficult to distinguish colors. But perhaps that is not as important as simply noting the range of variation.*

This was the intention!

*pp. 4763-4765, Figs. 3, 4a, 4b: Each of these plots has a “raw” PNSD curve plotted on the right vertical axis with a scale of approximately two orders of magnitude lower than the scale for the true or inverted PNSD plotted on the left vertical axis. These raw curves must either be the original data in units of plain concentration (not per size increment) [...]*

That’s right! These are plain concentrations, now additionally pointed out!

*Instead of the “raw” curve, it would be more instructive to plot the result of that first process,  $dN/d\log D_{pve}$  without multiple charge correction of any type, to compare to the final results including multiple charge. In this way, the magnitude of the multiple charge correction is clearly illustrated.*

We don’t understand the aim or intention, plotting such intermediate step/result?

*p. 4766, Fig. 5a: Is the DMPS channel number linearly related to  $\log D_{pve}$ ? If so, it would be helpful to include a second  $x$  or  $y$  axis showing  $D_{pve}$  values.*

A previous version was with  $\log D_{pve}$  instead of channel number. But it was too confusing, for a visualization of matrix entries.

- Replacing or deleting minor words according to the referee is not explicitly listed.