Reviewer Comments for Manuscript AMTD-6-4735-2013 “A novel inversion algorithm for mobility particle size spectrometers considering non-sphericity and additional aerodynamic-/optical number size distributions” S. Pfeifer, W. Birmili, A. Schladitz, T. Müller, A. Nowak, and A. Wiedensohler.

General Comments and Major Issues

This work is essentially an extension of the work of Wiedensohler et al. (2012) directed toward harmonization of techniques to measure aerosol size distributions using mobility particle size spectrometers. As shown in that previous study and reproduced here in Fig. 2, there is notable variance in the results of various methods used to correct for multiple charge effects on the mobility distribution. The object of this work is to propose and clearly document a relatively straight-forward procedure for inversion of the SMPS or DMPS data. Though not specifically stated here, it is understood that this procedure is in essence being proposed as a possible standard for uniform inversion of mobility distributions.

The authors have done an excellent job of accomplishing what they set out to do. The writing is reasonably clear and complete. Nevertheless, there are a few significant issues that need to be addressed. The first of these is the unorthodox definition of particle electric mobility that has been used throughout the work. Because this definition is never explicitly given, it can result in much confusion for any reader. The second issue is the lack of attention to dimensions (units) leading to some incorrect equations. Another problem is that the issue of non-spherical particles and how they may be dealt with in the proposed article is not really covered in sufficient detail.

The commonly accepted definition of particle electric mobility, Z', is the product of the particle mobility and its net charge (see e.g. Fuchs, 1964, Eq. 27.2, page 113). 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. Granted, the altered definition makes it possible to define a one-to-one relationship between particle electric mobility and diameter; nevertheless, the authors should stay with the accepted definition, replacing $Z'_{\text{(altered)}}$ with $Z'_{\text{(common)}}/n$ everywhere. In this way, $Z'_{\text{(common)}}$ will correspond to the normal definition of particle electric mobility. Also, the one-to-one relationship between Z'/n and D_{pve} will be preserved.

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. It is only through the approximation of the DMA transfer function as a Dirac delta function, δ, that the values of these two parameters can be equated. Nevertheless, even then their meanings are quite distinct. Thus, $f^*(Z)$ is the response of the SMPS system at a DMA voltage, V, from which Z is calculated. Conversely, $f^*(Z')$ is the particle electric mobility (Z') distribution, which, in the case of the aforementioned approximation to the DMA transfer function, is numerically equal to the response function, $f^*(Z)$.

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 as is δ. This leaves only dZ' with units of electric mobility on the right side of the first two lines of Eq. (10). The resulting discrepancy in units is "conveniently" lost upon incorrectly integrating the δ function with respect to Z' to obtain a dimensionless 1. The problem here can be seen as originating in Eq. (3) where the DMA transfer function is written as a function of (Z'-Z) rather than the more naturally occurring (Z'/Z - 1) as seen in the following Eq. (4). Using the latter notation means there are no extra Z parameters floating around in the definition of h^{dma} which becomes inherently dimensionless being the function solely of the dimensionless parameter (Z'/Z - 1). The equations should then be adjusted such that the integrand in Eq. (10) becomes $\delta(Z'/Z - 1)\, dZ'/Z$ which in turn properly integrates to a dimensionless 1.

For non-spherical particles, switching to the volume-equivalent particle diameter, D_{pve}, as the size parameter is a reasonable first step. However, what is needed for the DMA transfer theory is a mobility-equivalent particle diameter and for the charging probability a charging-equivalent particle diameter. Using average values associated with randomly oriented particles, this reviewer imagines that both of these latter size parameters would be greater than D_{pve} but not necessarily equal to each other. For each of these there is then associated a different shape factor, neither of which have been indicated in any of the equations (excepting Eq. (A2) in the Appendix). 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}. Though APS data were used in the example of Fig. 3, no details of the employment of the aerodynamic shape factor used are given. As the authors are attempting to provide a clear and complete "recipe" for multiple charge correction and claim to be able to handle non-spherical particles, there should be much better documentation of this part of the procedure.

Other Notable Problems

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.' There is no mention of the possibility of alternative approaches that may fall somewhere in between these two. For instance, though the mobility distribution is measured at N points, one might consider solving for the size distribution at $P > N$ points with the addition of some sort of smoothing constraint that involves minimization of a measure of coarseness of the size distribution. This technique has been around for some time and the reason for rejecting this merits a brief explanation here.

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

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 for all cases within that constraint is an approximation and a citation to a reference that documents this is needed here.

p. 4739, Eq. (4): The two-region definition of the DMA transfer function in this region is superfluous. The quantity in the large parentheses in the expression for the non-zero region naturally goes to zero beyond that region. Thus, there is no need for the region limitation or the second line of this equation. Also, the signs are wrong within the second absolute value. The quantity term within the large parentheses should be $|(Z'/Z) - (1 - \beta)|$.

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).

p. 4741, lns 5-6: The total transfer function is not a convolution but merely a product of the DMA transfer function (Eq. 5) and the <u>charging probability (Eq. 6a,b)</u>. Eq. (7) already shows this product except for the factor $A(Z/n)$.

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. Also, the function of C_i is probably better understood by noting that f^* has the units of dn/dln$Z$ which are converted to dn/dlogD_p for f using C_i . This has nothing to do with any approximation by Stratmann et al. (1997).

p. 4742, ln 4: The acronyms LDMA and CPC have not been defined.

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.

p. 4744, ln 7: This limitation on the size range could use a reference or, lacking that, more explanation.

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, $Z_i/n >= Z^\mathrm{m}_N$ for the first line and $Z_i/n < Z^\mathrm{m}_N$ 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 (Z_{high} , Z_{low}).

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 from the conditions on either side by more space (even more than the space surrounding the “element of” symbol) or the simple inequality conditions (<, >) were put in parentheses. Perhaps both changes would be even better.

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. As in Eq. (18), the inequalities must be reversed (“>” goes to “<”) in the second condition of each of the first three lines on the right side.

p. 4747, ln 4: “… has ~~now~~ no effect …”

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

p. 4750, lns 22,24: EMPD should be EPMD.

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

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. Uncertainties are most likely to take the form of a consistent bias due to calibration issues or variations associated with temperature changes and other parameters that vary over much longer time scales than the time width of a measurement channel. Thus, any variations or uncertainties of measured concentration due to uncertainties in the instrument aerosol flow are likely to have a significant degree of correlation from one measurement channel to the next such that the resulting error/uncertainty in the calculated size distribution cannot be found by simply employing $\mathbf{L}^{var}$ as defined in Eq. (33).

p. 4752, lns 21-26 and following: Typical scan data from an SMPS is quite dense in diameter space compared to any non-linear feature likely to be found in ambient aerosol distributions. 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. In addition, the typical width of a channel is likely on the order of or less than the width of the DMA transfer function. The latter has already had the effect of smoothing the measured size distribution somewhat, so again higher order interpolation would only be fitting channel-to-channel noise. 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. Without proper constraints, such an inversion might actually enhance the noise in the measured data. Also note that for most SMPS setups, diffusional broadening of the DMA transfer function does not become significant until the particle diameter is well below the range of significant multiple charge correction. The two effects are essentially mutually exclusive.

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

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*.

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 $a_5(0) = -0.0105$.

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.

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) or that concentration divided by β to give $dN_{\text{chg}}/d\ln Z$ where N_{chg} is the concentration of charged particles only. Though this indeed leads eventually to the true PNSD, without the initial single charge correction, it is far from it at this point and it does not really add anything to the point being made by the plot. The first part of the inversion process (single charge only) going up the point of the multiple charge correction is well-established within the aerosol community and not being challenged here. Instead of the "raw" curve, it would be more instructive to plot the result of that first process, $dN/d\log D_{\text{pve}}$ 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.

p. 4764, Fig. 4a caption, ln 3: "… influenced by <u>multiple</u> charge <u>correction</u> from …"

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