Atmos. Meas. Tech. Discuss., 6, C2098–C2104, 2013
www.atmos-meas-tech-discuss.net/6/C2098/2013/

Atmospheric Measurement Techniques Discussions

AMTD

6, C2098–C2104, 2013

Interactive Comment

Interactive comment on "A novel inversion algorithm for mobility particle size spectrometers considering non-sphericity and additional aerodynamic/optical number size distributions" *by* S. Pfeifer et al.

Anonymous Referee #4

Received and published: 16 August 2013

[english]article [T1]fontenc [latin9]inputenc [a4paper]geometry verbose,tmargin=2cm,bmargin=2cm,lmargin=2cm,rmargin=2cm

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Review of Pfeifer et al. AMTD-2013-108

This paper describes an effort to develop a general inversion algorithm that incorporates differential mobility analysis, either DMPS or SMPS, including shape effects and the influence of multiple charges, along with other analyzers that employ different measurement physics, e.g., aerodynamic particle sizers, or optical particle counters. It should be noted that, while the title identifies OPC measurements, they are not discussed in the paper. Though the authors claim to present a state-of-the-art inversion method, it fails to meet that standard on several counts. On the basis of these weaknesses, I must conclude that the work is neither new nor useful, and should not be accepted for publication.

First, in the interest of simplicity of matrix inversion, the authors limit themselves to the nondiffusive transfer function for the DMA, i.e., that originally derived by Knutson and Whitby (1975). They suggest that this transfer function is more general, and attribute it to Stolzenburg (1988) who did, indeed, present a much more general transfer function that includes the effects of diffusion on particle transmission. Given present day interest in measurements that extend to very small particles, a nondiffusive transfer function can hardly be considered general. The size distributions used as test cases extend to 5 nm, where diffusional effects are important for most DMAs. The size range shown in Fig. 2 spans four orders of magnitude in mobility. Operation of any DMA at constant flow rates over such a wide range means that diffusion must have played an important role in those measurements. Therefore, the triangular nondiffusive transfer is inappropriate for the problem posed by the authors. Others (Stratmann et al., 1997) have sacrificed the predictive capabilities of the Stolzenburg transfer function (or others derived based on the physics of particle transport within the DMA), represented the diffusional effects by broadening the triangular transfer function. The fundamentally-derived transfer functions allow prediction of variation in performance as particle size or DMA operating parameters change. The use of the triangular transfer function sac-

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rifices that predictive capability, as well as all information about the tails of the transfer function which can be quite important in some measurements.

The second major failing is the inappropriate treatment of additional data. Numerous investigators have previously addressed the question of inversion of aerosol data arising from use of two or more instruments. The authors here assume that the additional data are perfect, i.e., the transfer function is an identity matrix. Perhaps they are assuming that the manufacturers of the instruments have produced a perfect inversion algorithm, an assumption that I cannot accept. Furthermore, the authors assume that, in overlap regions, one measurement is correct, and the other should only be used for corroboration. A viable, multi-instrument inversion must employ the best knowledge of the performance of each instrument, and must also address the differences in measurement physics, and the uncertainties that those differences impart to the data analysis problem.

Even if one assumes that the triangular transfer function is a valid approximation for the DMA, and that all additional measurements are perfect, the paper still suffers a fatal flaw. The authors seek an solution that can be performed by direct matrix inversion, e.g., by a simple Gauss-Jordan algorithm. Even if the transfer function for the instruments were perfectly known, measurement uncertainty introduces noise which, as the authors describe in their discussion of error propagation, can be amplified by direct inversion algorithms. The discussion of error is, as the authors suggest, rarely sought in the form of analytical solutions, but not for the reasons that they state. To obtain those estimates, the authors had to force the system to be fully linear. Nonlinearities in the instrument response functions preclude such solutions. The authors suggest that previous authors have ignored error propagation in the use of an inversion algorithm, but that is incorrect. Many authors have examined how their inversions deal with measurement error, most commonly by performing the inversion with synthetic data to which error has been added, i.e., the Monte Carlo approach that the authors suggest "must be used, further on."

In addition to ignoring the power of statistical methods in the solution of sets of Fredholm integral equations, the emphasis on direct inversion forces unnecessary assumptions on the form of the solution sought. Specifically, the authors constrain the solutions such that the number of sizes considered must equal the number of measurement channels, and suggest incorrectly that this is the optimal approach. They further suggest that the channels should be positioned to match the mobilities of the measurement channels. While this might be possible for measurements with one instrument alone, multiple instruments will necessitate mismatches between input and output particle sizes (or other metrics). It should be noted, however, that they do allow for mismatch through the use of linear interpolation. The authors might consider it instructive to read the paper by Wolfenbarger (cited below) which clearly outlines a rational treatment of the size distribution that is compatible with the multi-instrument inversion problem.

Though the title suggests that particle geometry will be taken into account, the paper only takes the most trivial approach, stating that a shape factor can be applied. No discussion of the nature of that shape factor is provided, or how one would determine its value and, importantly, its variation with particle size. Perhaps more significant is the lack of discussion of the role of shape in the multi-instrument problem in which different instruments operate on different physical principles and are, therefore, affected by shape in different ways.

There are many more problems with the paper which I discuss more briefly below.

First, though the problem of aerosol data inversion has been studied extensively, the nomenclature used in this manuscript is among the most confusing that I have seen. The particle mobility distribution (named by the authors the electrical particle mobility distribution as though there were electrical particles involved) is denoted $f(Z)$, and the signal obtained in a measurement is denoted$f^*(Z)$. The transfer function is denoted $h(Z - Z')$. Through much of the paper, it is suggested that h is determined by the DMA alone, ignoring all of the other factors that influence the measurement. Equation 8 suggests including in h the contributions of charging probability, and the so-called

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height parameter as a surrogate for efficiencies in transmission through plumbing and CPC counting. This could be stated much more clearly if one were to follow the logic in the many papers on data inversion, only a few of which are cited here.

The charging probability is taken from the Wiedensohler fit to the Hoppel-Frick model predictions which, in turn, was based on the theory of Fuchs. Not to belittle the very important contribution of Wiedensohler which made the Hoppel-Frick simulations useful to the community, but the primary references should be given, particularly since one of the suggested improvements is to reexamine the theory of particle charging, suggesting Fuchs as the starting point even though others have already identified errors and limitations in that early work.

Appendices are given for establishing an equation for linear interpolation, and for the translation of $dN/dlnD$ to $dN/dlogD$. Neither of these would be needed if the sections where those arise were written clearly.

The "enhanced inversion" is a strange amalgamation of the inversion proposed for DMA data and undefined inversion of data from the additional measurements. On p. 4725 it is incorrectly stated that to use measurements from two instruments in the overlap range leads to problems: "If it is (were) assigned to both, it would be overvalued and considered wrongly twice." A statistical analysis of the data would use both sets of measurements to obtain the best picture of the distribution in that regime.

In section 3.4 on suggested improvements, it is noted that the finite width of the transfer function is ignored by the present version of the algorithm. Wolfenbarger and Seinfeld (*J. Aerosol Sci.* **21**: 227, 1990) demonstrated that the true width of the transfer function can be taken into account in formulating the sort of approach presented here, at least when the linear interpolation representation of the size distribution is employed, and did so in a statistical inversion. Unfortunately, that algorithm, though much more clearly defined than the present one, is sufficiently complex that it has seen little use, though Swihart has reported an effort to make it more user friendly (Talukdar and Swihart, *Aerosol Sci. Tech.* **37**, 2003). Nonetheless, the Wolfengbarger paper formulates the data analysis problem with transparency that is lacking here.

Minor points:

The written language needs considerable editing. Included below are a number of points, though I have not attempted to exhaustively edit this paper.

1. p. 4738, l. 8: Delete "vice versa." Possibly rewrite as: It is possible to calculate the real particle size distribution from the measured mobility distribution by deconvolution.
2. p. 4378, l. 16: half-width
3. p. 4379, l. 1: multiply charged particles
4. p. 4379, l. 6: What do you mean by "almost constant?" Steady in time, or slowly varying with Z?
5. p. 4379, l. 10: insert commas after "that" and "particles"
6. p. 4379, l. 12: insert "those" after such as
7. p. 4379, l. 18: non-spherical
8. p. 4380, l. 9: parameters.
9. p. 4380, l. 10: multiple charges, i.e., for a given . . . .
10. p. 4380, l. 12: multiply-charged
11. p. 4742, l. 4: The diffusion losses inside the LDMA and the CPC efficiency need to be incorporated into the total efficiency E. An example of this is given in Appendix C wherein the efficiency is modified for use of a . . . .

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12. p. 4742, l. 12: As previously mentioned, the measured mobility distribution is given in N discrete mobility channels, where . . .

13. p. 4742, l. 14: delete “the equation of”.

14. p. 4642, l. 15: Define Z_i and Z_j.

15. p. 4743, l. 10: Where does the constraint described by the equation beginning “furthermore . . . ” come from?

16. p. 4743, l. 15: Equation 15 clearly indicates that the authors are considering only direct inversion, and are ignoring the statistical nature of the inversion problem.

17. p. 4744, l. 6: Tammet and coworkers have extended differential mobility analysis to 10 microns.

18. p. 4745, footnote: the the

19. p. 4748, l. 1: replace “less” with “sparsely”

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