The manuscript reports on a new algorithm for solving the problem of particle mobility distribution to particle size distribution conversion handled by mobility particle size spectrometers (MPS). A major novelty of the algorithm is that it solves the problem for the particle size distributions simultaneously measured by a MPS, commonly limited to the submicron particle size range, and an instrument suitable to measure the particle size distribution of larger micron sized particles like the aerosol particle sizer (APS). The algorithm accounts for multiple charging of the larger micron sized particles, improving the accuracy of the measured particle size distribution in the upper limit of the particle size of the MPS; with respect to algorithms implemented in current MPS which does not account for the presence in the aerosol of particles outside the particle size range covered by the MPS.

The mathematical approaches adopted to solve the matrix inversion problem such as the preservation of the original size bins of the particle mobility distribution measured by the MPS and the linearization of the particle size distribution between size sampling points are well founded, as they allow reducing computational time and assessing error propagation which are essential issues for atmospheric aerosol measurements, when large volume of data and unavoidable uncertainties in the data need to be handled.

More fundamental issues like the election of a “volume equivalent diameter” D_{pve} for the entire particle size range covered by the MPS (based on mobility equivalent diameter) and the APS (based on the aerodynamic particle diameter) are, however, poorly described in the paper. Also classical theories on the bipolar charging of spherical particles are used and extended to non-spherical particles simply by introducing D_{pve} in the formula of the probability charging for spherical particles, without further discussion and without citing previous works in the field.

The paper is relevant to the scientific community on atmospheric aerosol measurements and deserves to be published in the AMT. Further corrections and comments to the manuscript are provided in the Review Report below.

Review Report

P 4736

Abstract

"The algorithm is able to calculate the propagation of measurement errors, such as hose based on counting statistics, into ~~on~~ the final particle number size distribution."

P 4740

"while the transfer function (see Fig. 1) is given by ~~Stolzenburg (1988)~~ Knutson and Whitby (1975)"

Notice that the triangular transfer function of the ideal DMA shown in Fig. 1 is originally due to Knutson and Whitby (1975). Stolzenburg extended this work to account for the broadening of the transfer function of the DMA for diffusive nanoparticles; for these the transfer function is a bell-shaped curve given by a much more complex formula than Eq. (4).

2.2 Charging probability and transfer function of multiple charged particles

The first paragraph of this section is confusing. I suggest rewriting (e.g. as below) and extending it. Also, I recommend citing the reference by Gunn as well as works on the charging of non-spherical particles like the ones given below and/or others.

"Since we intend to consider non spherical particles in the algorithm, we employ the volume equivalent particle diameter D_{pve} as the size parameter. This approach needs to be viewed critically, because the orientated average geometrical cross section, which is the much more important size parameter, would increase for non-spherical particles.

To calculate the probability of multiple charged particles in ~~a~~ bipolar charge equilibrium, we use the analytical approximation formulae given by Wiedensholer (1988)~~. Wiedensholer´s approximation~~, which is valid for singly and doubly charged particles smaller than 1 µm

Equation (6a)

For larger particles or ~~higher~~ highly charged particles we use the ~~Gunn-distribution~~ charge probability distribution by Gunn (1956)

Equation (6b) "

The significance of the first paragraph above is not clear: What does *"orientated average geometrical cross section"* actually mean? Why is it *"the much more important size parameter"*? Why *"would it increase for non-spherical particles"*?

Particle mobility classification

If the particle property relevant to the measurement instrument (MPS) is the electrical mobility, the equivalent diameter of interest should be the mobility equivalent diameter. The election of the volume equivalent diameter needs to be further explained.

Bipolar charging of non-spherical particles

The authors do not justify the approach of dense spheres of the same volume (D_{pve}) for the bipolar charging of non-spherical particles. Actually there are a number of papers both analytical and experimental on this topic (Ku et al., J. Electrostatics 69(6):641-647, 2011; Filippov, J. Aerosol Sci. 25(4):611-615, 1994; Rogak and Flagan, 23(7):693-710, 1992). As an example, in the paper by Rogak and Flagan, they conclude that *"the bipolar diffusion charging of agglomerate particles was found to be very similar to that of dense spheres of the same mobility, suggesting that DMA inversion procedures developed for spheres may be used for agglomerates as well"*, which supports the use of the mobility equivalent diameter.

Also the authors should explain which types of "non-spherical" particles are relevant to atmospheric aerosol measurements: agglomerates, dense particles of large aspect ratios, others?

P 4744

"So we found ~~the~~ a system of equations for the multiply charge inversion with the entries of matrix A"

P 4750

"According to the Bienaymé formula (please, provide a reference) it is valid:"

3 Results

"5. The algorithm can handle all procedures using a constant or size-dependent aerodynamic shape factor."

Define the aerodynamic shape factor: is it the parameter $\chi(D_{Pve})$ in Eq. (A2) in Appendix A? How is it determined in practice? Which are typical values of this parameter for atmospheric aerosols?

Examples of the application of the algorithm to cases in which the aerodynamic shape factor varies in time are not provided in the paper. In this case: are the discrete mobility sampling points Z_i time dependent?

3.2 Inversion of a wide size distribution combining SMPS and APS data

"In Fig. 3, we illustrate the benefits of a multiple charge inversion combining information from multiple sizing instruments,"

Actually Fig. 3 is confusing:

a) The volume equivalent diameter D_{pve} is represented in the horizontal x-axis. However, the caption of this figure states *"Dashed black line: raw electrical particle mobility distribution (EPMD)"*; the raw data of MPS are given as function of the particle mobility Z (not particle diameter);

b) There are two vertical y-axes both on the left and right side and of the figure: which curves corresponds to which axis? Does the right-hand side y-axis correspond to the curves labeled *"raw input"* and *"APS"* and the left-hand side y-axis to the curves labeled *"conventional inversion"* and *"enhanced inversion"*. Please, clarify this in the text or in the figure.

P 4752

3.4 Suggested improvements and extensions

In this section, the authors mention a list of issues to be undertaken in order to improve the predictions of the algorithm. It will be useful to know the opinion of the authors about the priority that should be given to any of these issues, according to its impact on the accuracy of the final particle size distribution as well as on the calculation time.

Higher order interpolation schemes and more accurate analytical formulas of the transfer function may be computationally quite demanding as compared, for example, to the use of a uniform theory for particle charging in the entire particle size range; the impact on the final particle size distribution of any of these issues is either unknown or not assessed by the authors in the paper?.

P 4756

References

Gunn, R.: The ratio of the positive and negative light ion conductivities within a neutral aerosol space J. Colloid. Sci., 11, 691-696, 1956.