Dear referee,

first, we would like to thank you for your exceptional review. You are a person with a most outstanding experience and overview of aerosol measurement techniques including their historical development. Therefore, we highly appreciate your suggestions.

As you point out, quite a substantial number of inversion of algorithms have been developed in the past. One class of algorithm, for example, is represented by the Bayesian approach (Ramachandran and Kandlikar, 1996). These algorithms avoid solving the inverse problem by reverting to solve many cases of the forward problem. They can easily handle input data from multiple instruments as long as the instrument response matrix is known. However, these algorithms require a massive computational effort to solve even one single size distribution inversion.

Another class of algorithm uses constrained regularization of the inverse problem (e.g., Wolfenbarger and Seinfeld, 1990). These algorithms allow a free form for the instrument response matrix (non-quadratic), but require a careful choice of constraining parameters and weights of penalty functions.

In fact, we appraise these achievements, and do not challenge these works. We do not think at all that the present paper questions those achievements or makes any of them redundant.

Nevertheless, it has not escaped even you that these algorithms have not been sufficiently implemented in practice. The apparent main reason seems these algorithms have been perceived as too complex (hindering practical implementation) and/or numerically demanding. Most of the described algorithms contain elements of arbitrariness, such as how to smooth data and/or penalize deviations from prescribed size distribution shapes. It is an unfortunate observation that the majority of inversion algorithms that are actually used much in practice (e.g. observation networks), seem to be documented only poorly.

Our own starting point was to provide an algorithm that is not necessarily the most accurate one, but one that fulfills the following requirements:

1. to create a fast algorithm — designed to handle large amounts of data currently produced in measurement networks

2. to create a lightweight algorithm — i.e. one that has a reasonable chance of being implemented by other researchers

3. to implement an algorithm that performs as few numerical steps as possible, particularly avoiding direct data interpolation, and multiple iterations

4. to create an algorithm that assumes as few approximations as possible, thus preserving as much of the original measurement information as possible

These issues have been held high in priority during the development of our algorithm. Meanwhile, it was kept in mind that the uncertainties taken into account were always inferior to the very real uncertainties involved by the measurement procedure. The comparisons of different inversion algorithms shown in Wiedensohler et al. (2012) (including inversion algorithms that use constrained regularization, such as ULUND, UHEL, NILU) suggest that this is well the case.

specific comments:

It should be noted that, while the title identifies OPC measurements, they are not discussed in the paper.

We are aware of this discrepancy and consequently changed the title to the former working title: *A fast and lightweight inversion algorithm considering additional number size distributions outside the detection range.*

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.

This is a harsh statement. As mentioned in our introductory statement, we do not plan to compete with inversion approaches that might yield, from a theoretical point of view, more "accurate" results. However, the requirements for a rigorous treatment of a flexible instrument response (e.g., concerning wide and non-triangular DMA transfer functions) have unarguably made the algorithms complex and unhandy. According to practical experience, we consider the uncertainties associated with the use of different inversion algorithms to fall short of those associated with the real measurement uncertainties (see Figure 2). With this regard, one can certainly see a usefulness for such a development.

First, in the interest of simplicity of matrix inversion, the authors limit themselves to the non-diffusive 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.

This is not true. Particle losses due to diffusions are accounted for by the size-dependency of the transfer function area $A(Z)$ (p. 4739, Eq. 5). There might have been a misunderstanding due to our non-appropriate use of the reference (Stratmann, 1997). We clarified this issue now in the revised text.

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.

See above. Please keep in mind that the the height α and width β, respectively the area A is variable! (see above) Using Fig. 2 as an argument or example seems to be strange, because it is exactly Fig. 2 that suggests that variations in the numerical treatment of the transfer function area do hardly

matter as the lower end of the size distribution range.

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. [...]

See above. Particle losses due to diffusions are accounted for by the size-dependency of the transfer function area $A(Z)$ (p. 4739, Eq. 5).

The second major failing is the inappropriate treatment of additional data. Numerous investigators have previously addressed the question of inversion of aerosol data aris- ing 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 as- suming that the manufacturers of the instruments have produced a perfect inversion algorithm, an assumption that I cannot accept. [...]

In practice, our starting point for additional spectral information is a volume equivalent size distribution.

How this volume size distribution might be derived from concrete instrumental data (APS, OPC, etc.) is an issue that we deliberately leave open to the user of the algorithm. In the case of APS size distributions, one would need to assume an effective particle density (combining shape and gravimetric material density). In the case of the OPC, considerations of refractive index might be necessary, and/or information on how the OPC was calibrated.

In any case, the generation of the volume equivalent size distribution might differ from one to another experimental situation and atmospheric aerosol type. The procedure might depend on the capabilities of the sensors. It would be difficult to say from a general standpoint what would be the best choice for these values.

In any case, we prefer to keep our algorithm general with respect to this issue.

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.

and

[...] 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.

Here, we are trying to understand what your mean by your criticism. You are right in that there is usually a finite diameter interval in which both size

spectrometers overlap.

Our approach, however, keeps the two size distribution branches apart, i.e. there is no statistical merging of the two branches. In our case, the diameter of separation is the uppermost channel of the DMA, i.e. the DMA mobility distribution enters into the calculation as a whole, while the APS/OPC data enter only above this diameter. (Below the diameter of separation, the APS/OPC data are disregarded; the reason is that we perform an inversion of mobility distributions, so that the APS/OPC data are considered as an auxiliary contribution. I think nobody would not hesitate to invert a DMA mobility distribution even without APS/OPC data, would they?)

To make it clearer: We are inverting DMA mobility distributions, so there is every reason to take the full existing DMA mobility distribution into account, i.e. without statistical merging of APS/OPC data. Our inversion algorithm then subtracts contributions of multiply-charged coarse particles above the threshold diameter from the DMA mobility distribution below the threshold diameter.

The issue, how the two branches of size distributions are later merged into one final distribution something that we deliberately leave open to the user. As in a related discussion above, the merging of SMPS/DMPS and APS/OPC data can be a sensitive issue with respect to instrumental details, and also the type of atmospheric aerosol. In the case of the APS, one would typically transfer the aerodynamic size distribution into a volume equivalent size distribution by means of an effective density. (This effective density needs to be assumed, or determined independently.) Depending on the situation, different operators might therefore prefer to use different data assimilation schemes.

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.

This is a false presentation. The course of events was that we first fixed the assumptions, on which the algorithm should be based:

1. Variations of the PNSD across the range of the transfer function are negligible. (Based on a relatively narrow DMA transfer function compared to a wide atmospheric PNSD, and the use "first mean value theorem for integrals"!)

2. The PNSD can be interpolated linearly between discrete sampling points. This is certainly valid for SMPS distributions with a dense grid of sampling points.

Next, we sought a solution to this problem. It turned out that this could be achieved by, as you say, a Gauss-Jordan algorithm. It was not enforced to obtain such a simple system of equations, but is a direct result of these two approximations.

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.

As described in the above paragraph, our system was not "forced to be fully linear" but rather turned out to be. As long as you can accept the underlying assumption of the narrow-DMA-transfer function for atmospheric conditions, you might also accept the consequential results for the propagation of error.

Nevertheless, we still improved the discussion of error according to the criticism of another referee (# 2). A solution for correlated adjacent grid points has now been added to the manuscript.

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."

We are aware of this bad wording, and a possible misunderstanding. Our statement did not refer to other inversion methods, but to the confidence intervals that are rarely attached to inverted PNSD in scientific publications in general.

To improve the wording, the sentences have now been reformulated:

An important point [...] (p. 4748 l. 1)
This aspect is necessary [...] (p.4754 l.1)

This method works, and can calculate a confidence interval (with a scientific basis, based on these two approximations) without a significant increase of computational effort.

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.

See our explicit motivation at the beginning!

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).

5

In our understanding, there is a difference between "should" and "must". (We said "should").

We still believe that keeping the number of original measurement channels is an optimum approach. Why? If you change to a different set of measurement channels, you need to decide how you rebin and/or interpolate the mobility distribution. Various interpolation methods have their ups and downs in conjunction with different shapes of size distributions and/or levels of noise. Why change measurement channels if you do not need to? At this point of the discussion I cannot help the impression that you criticize our work even for its natural advantages.

About the treatment of multi-instrumental (DMA vs. APS/OPC) data, which are treated in a certain hierarchial sense in this work, see above.

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.

We would like to say that our approach could be equally considered the most general rather than the most "trivial". As we know, particle shape factor in a real aerosol is everything else than trivial, and might differ strongly between experimental situations and atmospheric aerosol type. As a matter of fact, we refrained from providing much background information (or guidelines) because this issue is so non-trivial.

In practice, an operator needs to choose a shape factor depending on his experiment and aerosol type.

Nevertheless — in order to improve the text, we now guide the reader with the following transformation formula:

FORMEL

as well as a new reference (Hinds, 1999) where some shape factors are listed in tabular form.

We would like to emphasize that any further and meticulous discussion of aerodynamic shape factors, and its related characterization methods are definitely out of the scope of this paper.

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.

We welcome this sentence and that you pointed this out! We are aware of the big relevance of this issue. While for a SMPS-APS combination it is not too complicated, coupling the influence of morphology for SMPS and OPS is much harder. But this issue is much more complex, larger than a multiple charge inversion and it will go beyond the scope!

By the way: The first author of this article (S.P.) is currently working on the issue of non-spherical particles and their effect on optical light scattering within the framework of his PhD thesis. So, we are certainly aware of the topic

you addressed.

[...] 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 height parameter as a surrogate for efficiencies in transmission through plumbing and CPC counting [...]

"...as though there were electrical particles involved": Sorry, this seems to be a misunderstanding. What are the particles being deflected inside the DMA if not electrical? (They carry a charge, so they are "electrical", of course.) The term "electrical" stems, probably, from the term "electrical mobility", and became "electrical particle mobility distribution" in conjunction with "particle mobility distribution". It is actually our belief that this term is the only really correct term describing this parameter. Otherwise, particle mobility distribution could refer to a purely mechanical mobility distribution, i.e. where multiple charges do not occur!

About the formula symbols of parameters: It is almost impossible to find two literature sources with the same nomenclature! (We decided not to list the results of our survey here.)

We ourselves more or less follow the nomenclature established by Stratmann et al. (1997): The nomenclature for f and f^* is from mathematical basics of convolution. But of cause we've added also the parameter α, β and A.

[...] suggesting Fuchs as the starting point even though others have already identified errors and limitations in that early work.

In this paper we won't analyse or challenge the validity of theory for charging probability. Our algorithm is, in principle, flexible for the implementation of any charging probability formulations to come.

We fell back to the Fuchs theory for this version of the algorithm because it is one of the few formulations that explicitly consider the influence of important environmental parameters, such as temperature and air pressure on the charging probability.

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.

In our opinion, the article is already overstuffed with formulas. A reader who is skilled in interpolation or transformation of density functions, can easily skip these.

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,

We know the article of "Talukdar and Swihart" and thank for the cross-reference "Wolfenbarger and Seinfeld". In accordance to our Motivation, this isn't our aim, neither a increase of resulting grid points nor a reduction of noise! We've added/cited these and similar papers as examples, rather as a demarcation to our Intentions and resulting algorithm.

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