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Interactive comment on "Inversion of droplet aerosol analyzer data for long-term aerosol-cloud interaction measurements" *by* M. I. A. Berghof et al.

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Interactive comment on "Inversion of droplet aerosol analyzer data for long-term aerosol-cloud interaction measurements" by M. I. A. Berghof et al. Anonymous Referee #3 Received and published: 23 January 2014 This well written paper presents a new technique for analysing data from the Droplet Aerosol Analyser (DAA) in a more systematic way than previously employed. It provides a detailed, step-by-step description of the different stages required to process the data, from the mathematics of the data inversion to the losses within the system. The paper also presents examples of the data analysis products from a field campaign. It will be a good reference paper for any

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future projects producing data from the DAA. There are a few minor comments/issues which I would like addressed before full publication in AMT.

A: The authors are grateful to the reviewers for their comments that improved the manuscript.

Minor comments/issues

I feel the introduction is focusing too much on atmospheric processes and why there is a need for a DAA and not enough on why the data analysis products are required or why the inversion has been developed. The DAA already exists and has been described elsewhere. There is no need, in my opinion, to justify it here. For example, page 10271, paragraph 1, describes what a DMPS, OPC and fog monitor can do and the limitation with not being able to relate dry size to droplet size. Similarly with the discussion on the CVI. To my mind, this should be saying 'The DAA can provide a direct relationship between dry size and droplet size, unlike other instrumentation such as DMPS, OPC etc even when used in parallel. It has been shown to etc, etc, but the challenge has been in the processing of the data. '. Similarly with the details of the data from the Po valley and the DAA - DMPS closure study, this demonstrates the functionality of the DAA, which is already established, but not the need or advantage of the inversion, which is the main body (and title) of the manuscript. Do you get better statistics with the inversion? More data coverage? Is it simply more efficient? I think it would be better to cut down the introduction and make it much more focused.

A: The parts on previous results obtained with the DAA were given in order to increase the interest in the DAA. We see, however the logic of this comment. In response we will shorten these parts of the introduction and add material on the new developments of the DAA. We will also use the suggested method to review different cloud measurement methods.

Can the authors provide an estimate of the error associated with their assumption on page 10276 that the influence of multiply charged particles on the largest sizes

can be neglected? I include a graph of some data I collected recently from a DMPS using the same assumption and no impactor, which clearly shows a breakdown in the assumption. This is important because a) it is most noticeable at low concentrations and; b) it is very noticeable when the distribution is bi-modal, such as that produced by cloud processing with a distinct Hoppel dip. Both a and b are highly likely to occur in the DAA as the dry, residual distribution may not be the same as the dry, pre-cloud distribution on which their assumption is based.

A: In order to keep this problem to a minimum, we measure up to large dry sizes where number concentrations are very low in the atmosphere. The largest particle diameter in each second run was 0.55 μm and in the other runs 0.78 μm. First multiple charge correction appears for particle sizes 0.31 μm and 0.41 μm, respectively, i.e. far above the size where the Hoppel minimum usually is found. In the data presented here, concentrations in the highest channel were very low, see the example in Figure 2. Reviewing previous DAA measurements, the number concentration drops by a factor 7 to 100 from the size the largest measured particles to where this particle size appears as doubly charged (Martinsson et al., Atmos. Res. 50, 289-315, 1999; Martinsson et al., Tellus 52, 801-814, 2000), and in the data set used here the drop was between 14 to 27. Assuming similar gradient above the highest channel would thus indicate errors of the order of less than one per mille to approximately 1 percent error in the first corrected channel. Due to the strong gradient in this part of the atmospheric particle size distribution these errors do not propagate to smaller particle sizes.

A: In the manuscript this description will be given in short with a warning that gradients weaker than a factor 10 increase in concentration for a decrease in particle size corresponding to a factor of 2 increase in electrical mobility, can cause error of the order of 1% or larger .

Can the authors state the basis (or errors associated with) the derivation of equations 1 and 2? i.e. the linear relation.

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A: This error is size-dependent. For smaller particles the f(3) is small and hence the error is small. In most cases the 2, 3 and 4 charge particles are on a common gradient, implying that the estimated cannot deviate much from the actual concentration. In worst case scenario we have a narrow peak at the position of triply charged particles which will cause underestimation of the concentration of triply charged particles. A minimum is less sensitive because in that case the concentration of singly charged particles is higher compared to the triply charged particles, which reduces the effect. For the purpose of estimating the error, typical shapes of atmospheric size distributions need to be considered. For that purpose we searched for size distributions with narrow modes and found such a distribution in Hoppel et al., (JGR 101, 26553-26565, 1996; Figure 1). Placing doubly and quadruply charged at the same concentration around the maximum at 0.085 μm radius would cause an underestimation of the true concentration of triply charged particles by 15%. The only important effect of this error, i.e. the effect on the channel for singly charged particles (0.06 μm radius) would be an overestimation by 1.4%. Due to the dominance of singly charged particles for still smaller particles this error will not propagate to smaller sizes. Despite using the worst case scenario on a size distribution with a very distinct Hoppel minimum the error due to the approximation according to equations 1 and 2 is minor.

A: In the manuscript a short version of this explanation will be included with a warning that this approximation can generate errors for distributions with modes more narrow than in Hoppel et al., (JGR 101, 26553-26565, 1996, Fig. 1).

Section 3.3.4 are there losses in the dryer and are these accounted for?

A: Yes. The losses in the dryer are described in Sjogren et al. (AMT 6, 349-357, 2013) and are accounted for in the data evaluation. Note that the particles in that study were non-evaporating. Thus the supermicron particles in Figure 4 of that paper are lost in the outlet of the dryer. They are thus not representative of cloud droplets.

Figure 3a (legend), please change dry particle diameter to dry (residual) particle di-

ameter. Figure 4c and page 10285. I have a slight issue with the definition of the 50% activation as it is potentially mis-leading. This relates back to my earlier comment about cloud processing. The DAA will measure a modified, residual, dry size distribution, which is not the same as the distribution the droplets formed on. Some of the smaller particles will have grown in size and been removed from the sub-100nm mode to the larger accumulation mode when dried. It might only be a small effect, but the data could be skewed to larger sizes. This needs commenting on.

A: We agree. This is a dilemma of all sizing of cloud droplet residuals. This can be addressed by performing measurements outside clouds in comparison with cloud interstitial aerosol measurements. This method however, suffers from problems of precision because of the difficulty to probe the same air mass pre- and in-cloud. We will clarify.

Page 10286 and fig 5c. I am not convinced the analysis is rigorous enough to be so absolute and the emphasis of this section should be changed to focus on the application of the DAA products. For example, the opening sentence could be: 'Another application of the DAA data products is the estimation of the solute concentration within the cloud. This is achieved by etc, etc and assuming etc etc. A more rigorous approach would be to have HTDMA measurements'. Or similar. Then describe how to use the data and interpret figure 5c

A: Thank you. We will use the formulations.

Interactive comment on Atmos. Meas. Tech. Discuss., 6, 10269, 2013.

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