

**Authors' response to reviewer 1 comments on manuscript titled "Fast time response measurements of particle size distributions in the 3-60nm size range with the Nucleation Mode Aerosols Size Spectrometer", submitted to AMT 24<sup>th</sup> January 2018**

The authors would like to thank the reviewer for their considered and positive evaluations of the manuscript. Our responses are detailed below, with the reviewer comments in normal text and our response in italics.

1. p. 4, lines 3-5. The authors might consider mention of Winkler's DMA train among the fast-time response instruments for measuring nanoparticle size distributions (Pichelstorfer et al. 2018). Due to its higher sensitivity to number concentrations and lower weight and power requirements, the NMASS is more suitable for use on aircraft. However, the DMA train is a new development that has its place and might be mentioned.

*This is an excellent point, and the DMA train has now been considered on P4 lines 6-8. Text now reads: "A "DMA-train" composed of 6 differential mobility analyzers measures only the aerosol fraction that is charged in an ionizer (Stolzenburg et al., 2017), and is neither compact nor optimized for operation at reduced pressures."*

*We chose to reference Stolzenburg 2017, instead of the recommended Pichelstorfer 2018, as Stolzenburg 2017 provides a more in-depth description of the instrument and its capabilities.*

2. p. 8, figure caption. The authors cite the Airmodus PSM (Vanhanen et al., 2011) as an instrument that uses a working fluid other than n-butanol (diethylene glycol) and a two-stage CPC detector. These aspects of the Airmodus instrument are based on the earlier work of Iida et al. (2009), who identified diethylene glycol as a suitable condensing fluid for sub 3 nm particles and pioneered the use of a butanol CPC "booster" as a second stage detector for the small droplets on which diethylene glycol had condensed. The authors should consider citing Iida's contributions as well. Iida et al. (2009) also experimentally studied differences in activation efficiencies for positively and negatively charged sub 3-nm particles. This may be pertinent to your discussion on p. 11.

*Iida et al reference added to Fig 3. Caption. Caption now reads:*

*Fig.1 Kelvin diameter, or critical diameter,  $D^*$  as a function of difference in temperature between saturator and condenser, for n-butanol (used in many commercial CPCs such as the TSI-3776 (Hermann et al., 2007)), diethylene glycol (used in commercial and custom-built two-stage CPCs such as the Airmodus Particle Size Magnifier (Vanhanen et al., 2011; Iida et al., 2009)) and Fluorinert FC-43 (used in the NMASS CPCs). The saturator temperature is 34.8 °C. For a given  $D^*$  the slope of the curve for FC-43 is less than for n-butanol or diethylene glycol. The measured diameter of 50% detection efficiency,  $d_{50}$ , for an NMASS CPC is also shown as a function of temperature, as discussed in section 3.2. The NMASS CPC  $d_{50}$ s are larger than the theoretical Kelvin diameter for Fluorinert because heat and mass transfer within the condenser limits the supersaturation achieved to values less than the theoretical maximum. The range of  $d_{50}$ s shown here, around 40-60nm, are on the steep part of the diameter curve. This limits the largest  $d_{50}$  that can be achieved with the NMASS because, in this region, a small variation in temperature difference causes a large variation in  $d_{50}$ , making the detection efficiency unstable.*

3. p. 15: Although it may be obvious to those who have worked with CPCs, you might point out the reason that the counting efficiency for CPC5 decreases with decreasing size below 7 nm.

*We assume that the referee meant 70 nm, and have added clarification to the caption of Figure 6. Text now includes "At diameters <70 nm, the roll-off in detection efficiency of CPC5 ( $dp_{50}=59.1\pm 6.5$  nm) is already evident."*

4. pages 19 & 21. The paper indicates that the method used in section 4.1 was used to invert both the NMASS and SMPS data. The discussion is mercifully concise, but I

am still curious about several points:

-Are the data from the two NMASS instruments merged prior to inversion, or are the data merged separately and the inverted distributions merged after inversion? It is not entirely obvious to me which approach would be preferable, and the paper provides no insight. Systematic differences between the instruments that could lead to large errors in concentration differences between adjacent channels might argue in favor of separate inversions, but constraining a single inversion with more data points might argue in favor of merging the data before inversion. A sentence or two would suffice.

*This was indeed not explicitly addressed, so a sentence explaining the choice of a single inversion over 10 channels and why there are no large systematic differences between the instruments that might make this problematic has been added p20 lines 5-6. Text now reads "We use channels from both NMASSes in a single inversion and the calibrations ensure no large systematic differences between the two instruments."*

-Standard SMPS inversion methods (e.g., TSI's AIM software as well as software used by most aerosol scientists) would lead to accurate results for mean size and concentration for aerosols sampled from a DMA, (Figs. 9 & 10). However, the size distributions provided by those methods would be broader than the measured size distributions. This is because the distributions delivered by the DMA are broad relative to the transfer function of the DMA in the SMPS. I believe the modified Twomey technique that was used in this analysis should not suffer from that problem, but too few details are given for me to be certain. Nevertheless, if the DMA in Fig. 9 was operating properly, the size distribution of the sampled aerosol would be exactly equal to the DMA transfer function times the size distribution of the aerosol from the atomiser. It would be interesting to see this theoretical size distribution on Fig. 10 as well. If it agreed well with the blue dashed line, it would provide further support for the validity of your inversion algorithm. (Most aerosol scientists who have worked extensively with data inversion -I am certainly among them- are skeptical about the results.)

*We have addressed these concerns in an extra section in the supplementary material (section C) including figure S3, referenced on page 22 lines 22-23. New section with extra figure is as follows:*

### **C. Verification of custom-built DMA and nano-SMPS performance**

*A custom-built DMA was used to generate the calibration aerosol used for comparison between a nano-SMPS and the NMASS in Fig. 10. The performance of this DMA was verified by atomizing a nearly monodisperse polystyrene latex (PSL) sphere aerosol with a peak diameter of  $152 \pm 5$  nm and a polydispersity of 2.1% (ThermoFisher Scientific Series 3000 nanospheres). A CPC measured the concentration of particles exiting the DMA as the voltage was manually stepped across the peak in the transmission function (Fig. S3). A fitted Gaussian curve gives a peak diameter of 151.3 nm and a full-width at half-max (FWHM) of 17.4 nm (11.4%). The fitted peak diameter agree with the PSL size standards within uncertainties. The FWHM of the distribution is very close to the expected FWHM of 16.9 nm (11.1%) calculated from DMA theory (Knutson and Whitby, 1975) with the 10:1 sheath/aerosol flow ratio used and accounting for the polydispersity of the PSL. Thus the custom-built DMA is working close to theoretically optimal performance.*

*The custom-built DMA was used to produce a size-classified, atomized ammonium sulfate aerosol that was tested by the nano-SMPS and the NMASS (Fig. 10). The SMPS size distribution, inverted using the same Markowsky-*

Twomey algorithm that is also applied to the NMASS data, displays a FWHM of 13.6% and 12.6% for the 20 and 30 nm sizes selected, respectively. Thus the Markowsky-Twomey inversion applied to the nano-SMPS slightly broadens the aerosol generated by the custom-built DMA. This is not unexpected because the inversion applies a smoothing step, as discussed in Section 4.1.

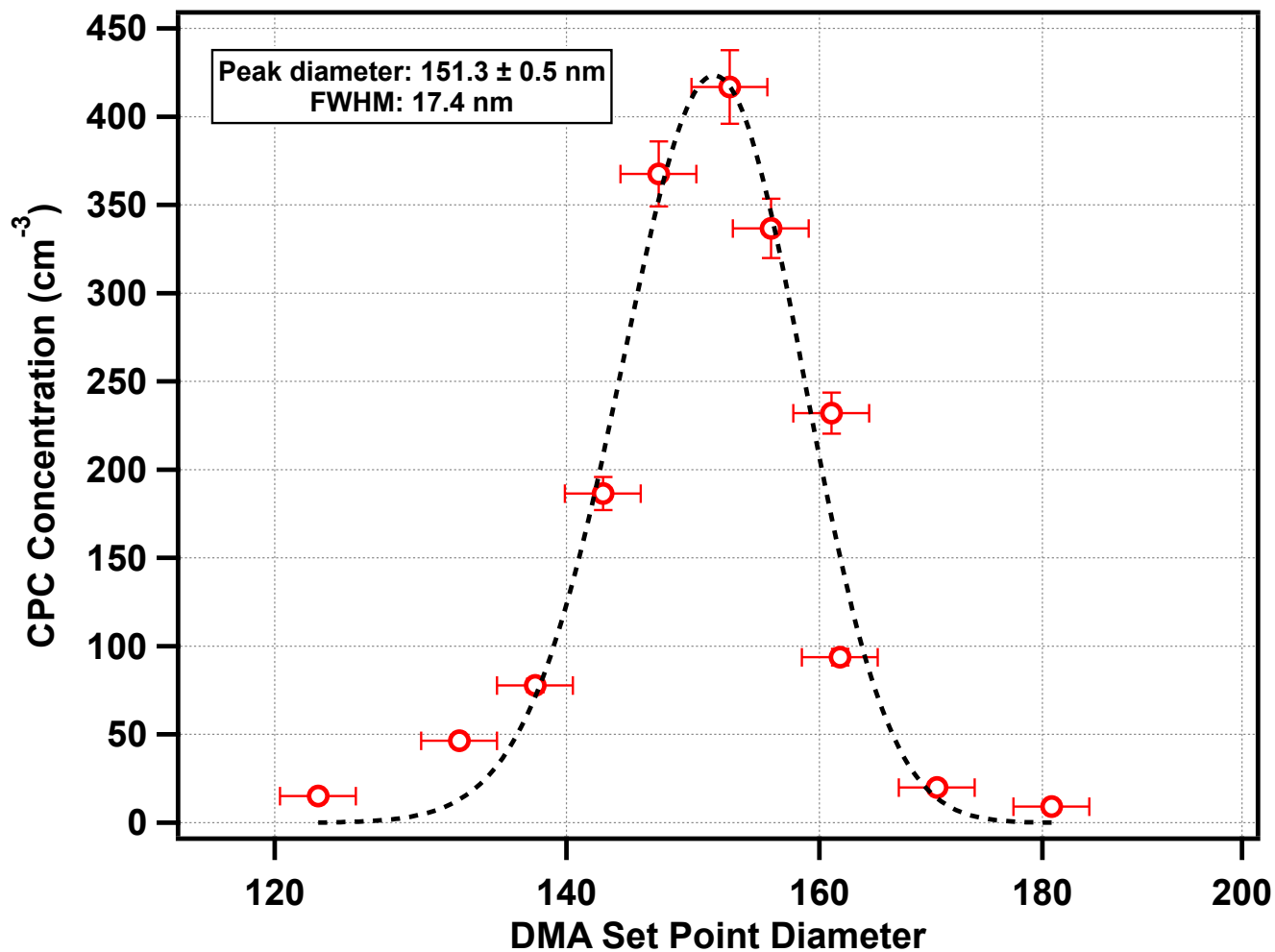


Figure S3. Concentration of particles produced by atomizing nearly monodisperse particles, classifying them in a custom-built DMA, and counting them with a CPC.

Minor Editorial Changes:

p. 7 line 14: missing “).” following Hanson et al., 2002) - *addressed*

p. 11, line 9: and mostly limited to the smallest... - *addressed*

p. 13 line 14: Should this be Fig. 5, not Fig. 6? - *Yes, addressed*

p. 16, caption to Fig7: “smallest cut-off sizes by.” ??? - *should have read “atomizing ammonium sulphate or dioctyl sebacate” – this has been added*  
p. 27, line 9: “for all but one channel” - *addressed*

## References:

Hermann, M., Wehner, B., Bischof, O., Han, H. S., Krinke, T., Liu, W., Zerrath, A., and Wiedensohler, A.: Particle counting efficiencies of new TSI condensation particle counters, *J. Aerosol Sci.*, 38, 674-682, 2007.

Iida, K., Stolzenburg, M. R., and McMurry, P. H.: Effect of Working Fluid on Sub-2 nm Particle Detection with a Laminar Flow Ultrafine Condensation Particle Counter, *Aerosol Sci. Technol.*, 43, 81-96, 10.1080/02786820802488194, 2009.

E.O. Knutson, E. O., and Whitby, K. T.: Aerosol classification by electric mobility: apparatus, theory, and applications, *J. Aerosol Sci.*, 6, 443-451, doi:10.1016/0021-8502(75)90060-9, 1975.

Stolzenburg, M. R., and McMurry, P. H.: An Ultrafine Aerosol Condensation Nucleus Counter, *Aerosol Sci. Technol.*, 14, 48-65, Doi 10.1080/02786829108959470, 1991.

Vanhanen, J., Mikkilä, J., Lehtipalo, K., Sipilä, M., Manninen, H. E., Siivola, E., Petaja, T., and Kulmala, M.: Particle Size Magnifier for Nano-CN Detection, *Aerosol Sci. Technol.*, 45, 533-542, 10.1080/02786826.2010.547889, 2011.