

## Reply to the comments of Reviewer #4

This study presents intercomparison of a number of SMPS carried out in instrument workshops. The authors developed technical standards for design and operation of SMPS, and data archive. These technical standards, when applied in future long-term surface-based measurements, will help improve the reliability and comparability of the SMPS dataset, which are important for improved understanding of the impact of aerosols on climate and air quality. The paper is quite comprehensive, and the topic is well suited for the journal of Atmospheric Measurement Techniques.

### Thank you

#### General comments:

One important consideration when inverting SMPS data is the time required for particles to travel from the exit of DMA to the CPC detector. For laminar flow, this travel time is non-uniform and its distribution is often described using two time constants: a fixed delay time and a mixing time (Collins et al., 2002 and Russell et al., 1995). If the distribution of the travel time is not taken into consideration, inverted size distributions may be skewed, especially for faster scans (1-2 minutes). The fixed delay time and the mixing time can be derived by comparing up scan and down scans. (If both up scans and down scans are used in SMPS, one additional data quality check is to compare up and down scans; they should be identical when appropriate fixed delay time and mixing time are employed in data inversion). For some SMPS, such as the commercial SMPS from TSI, the mixing time is not taken into consideration. Therefore, it is important that these SMPS are operated at very slow scanning speed (e.g. > 5min), which should be sufficient for surface-based measurements. I would suggest the authors to include in the manuscript the measurement speeds of SMPS during the study, if and how the correction of travel time distribution is implemented in the data inversions, and whether both up and down scans are used in the SMPS measurements.

**You are complexly right. We will add some sentences emphasizing this problem in the recommendation, although it was not the case during the workshops.**

The intercomparisons show substantial differences among SMPS measurements when particles are smaller than 20 nm or larger than 200 nm. Ideally these differences should be fully resolved with additional experiments. I think it would also strength the paper just including some analyses and discussions to narrow down the potential causes for these discrepancies. If the distribution of the particle travel time between DMA and CPC detector (as described above) is not fully considered in some of the inversion programs, the skew size distribution may contribute to the some of the discrepancy at  $D_p > 200$  nm, especially for fast scans and when only up scans are used.

**We will try to narrow down the reason for the increased uncertainty above 200 nm. First we try to invert all measurements with the same routine to find out if the multiple charge corrections are correct, although they were visible in the comparison of the inversion routines. Again, the delay time is probably not the problem since each up- or down scan was larger than 2 min.**

Figure 8 shows large difference in size distributions measured NILU and IFT REF1 at large particle sizes. Both NILU and IFT REF1 use Hauke-type DMA and TSI, 3010 CNC. The DMAs were also operated at the same flows (1:5). The IFT inversion routine uses “real area of DMA transfer function” whereas NILU uses Stolzenburg (1988) DMA transfer function. Could the differences be reduced if both routines use the same DMA transfer function? This could be easily verified as it does not require additional experiments.

**Yes, we will check this by using the same inversion routine.**

The difference could also be due to different neutralizers used in the two systems (TSI Kr85 vs. Ni63). We occasionally observed differences in measured size distributions when different neutralizers were used in SMPS. This could be easily tested by using identical neutralizers in both systems (but unfortunately would require new experiments).

**Yes, this is true. Unfortunately, we cannot repeat this experiment easily.**

Regarding the discrepancies for particles smaller than 20 nm, one potential cause is the treatment of particle diffusion losses. It is not clear from the manuscript whether the size dependent particle loss in each system was characterized experimentally or was estimated based on tubing length and flow rate. The other potential reason for the differences may be related to the counting efficiency of CPCs.

**Ok, I see. The lengths of the different pipes were measured. We will clarify this in the text.**

It is very surprising to see that the NILU and TNO showed no sensitivity at all for particles less than 15 nm, while Figure 3 shows both NILU and TNO CPCs have detection efficiencies greater than 80% at 10 nm! It is also not clear from Table 2 if CPC counting efficiency was corrected in inversion routine for TNO. It is worth examining the raw data to find out if any particles less than 15 nm were detected at all for the NILU and TNO SMPS. If not, this might suggest major problems/malfunctions of the systems.

**Afterwards, we found out that the DMA voltage for these systems were wrong. We will explain this in the text**

Specific comments:

Page 5524, Line 3, Please change “a wide application” to “a wide range of applications”.

**OK**

Page 5524, Line 10, Please add “s” after “distribution”.

**OK**

Page 5524, Line 18, Please change “analyser” to “analyzer”.

**OK**

Page 5528, Line 7, “This work was : : : Evaluation Program)”. It may be more appropriate to move this sentence to Acknowledgements.

**Good idea.**

Page 5531, Line 21 -23, To ensure aerosols size distribution are under “dry conditions”, the RH needs to be even lower than 40%. Some studies suggest the efflorescence RH of ammonium sulfate is \_37% or lower.

**This is true, but I would rather like to keep this 40% RH. Pure ammonium sulfate is rather seldom in the atmosphere. I will write a sentence that one has to be careful for some atmospheric aerosols**

Page 5531, Line 25-27, “Generally, a dry: : : aerosol particles”. Wet aerosol can also reach steady state charging distribution in a bipolar charger. The key is to ensure no changes in

particle size after steady state charge distribution is reached, such that it can be conveniently accounted for in inversion routine. Please rephrase this sentence.

**OK**

Page 5535; Line 5-6, What are the uncertainties? Not enough significant digits in the data format? Please clarify.

**We will check this**

Page 5540, Line 13-14, Were particle diffusion losses characterized experimentally, or estimated using flow rate and tubing length? What are the “unconsidered additional losses”?

**We will clarify this**

Page 5540, Line 14, What are DMA classifying voltages for 20 nm particles?

**We will check this**

Page 5540, Line 15-16, How long were the size distributions (shown in Figure 5 and 8) averaged for? The (averaged?) size distributions appear to be quite smooth, suggesting counting statistics was sufficient. Please provide the uncertainties for each size bin (due to counting statistics) using raw counts detected. This will help narrow down the potential causes of the discrepancies.

**Yes, the averaging time was long enough. We can add the uncertainty bar to the reference systems**

Page 5541, Line 13, please change “beside” to “except”.

**OK**

Page 5543, section 4.4.2, for neutralizers other than TSI Kr 85, how were the particle diffusion losses calculated?

**We assumed no big difference.**

Page 5545; Line 16-17, what are the automated data quality checks? Are they checks of instrument status parameters?

**We will check this**

Page 5545, Line 25-26, how are the data uncertainties propagated? How are the uncertainties in particle size and concentration derived? I think these are the important features of the new data format, and should be detailed.

**Ok, we will clarify this**

Page 5563, Table 2, row “IFT”, column “calculation”, what does “using real area of the DMA transfer function” mean? Does this mean using experimentally determined transfer function?

**Yes, it is experimentally determined. We will mention this in the text.**

Page 5565, Table 2, row “ULLUND”, column “calculation”, what is the ideal width? Width of non-diffusing DMA transfer function?

**We will check this.**

Page 5566, Table 2, For PKU and TNO, is CPC counting efficiency corrected in inversion routines?

**We will check this for PKU. It's not included in the TNO system.**

Page 5568, Table 3, Only three systems are described, how about other systems used in the 3rd inter-comparison workshop?

**We will check this**

Page 5581, Figure 3, TNO CPC 3010, operated at DT=17 degree, showed similar high counting efficiency as other CPCs operated at DT=25 degree. Was there any special modification of this CPC?

**It's an in-built CPC. It might be different to a TSI 3010**

Reference:

Russell, L. M., R. C. Flagan, and J. H. Seinfeld (1995), Asymmetric Instrument Response Resulting from Mixing Effects in Accelerated DMA-CPC Measurements, *Aerosol Sci. Technol.*, 23, 491-509. Collins, D. R., R. C. Flagan, and J. H. Seinfeld (2002), Improved inversion of scanning DMA data, *Aerosol Sci. Technol.*, 36, 1-9.

**We will add this reference**