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Atmospheric Measurement Techniques Discussions

Interactive comment on “Development of an H-TDMA for long-term unattended measurement of the hygroscopic properties of atmospheric aerosol particles” *by* E. Nilsson et al.

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1) “However, higher than that.” Quantification of the overall error would be valuable.

Answer:

Quatification of the overall error is what is measured with the ammonium sulphate scans, and displayed in figure 5 and table 1. We feel that the discussion on the temperature measurements in combination with the measured error should answer this question, as an error propagation calculation would need too many assumptions to make it useful. To clarify this, the following passage on page 7 has been changed from: “However, considering uncertainties in flows, voltages, charging efficiency, purity

of the reference solution etc. the deviations from theory can be expected to be higher than that, and this can be seen in figure 5, where a few outliers are visible, most likely due to one or several of the reasons previously mentioned.”

to

“Considering uncertainties in flows, voltages, charging efficiency, purity of the reference solution etc. the deviations from theory can be expected to be higher than that, and this can be seen in figure 5, where a few outliers are visible, due to one or several of the reasons previously mentioned. However, as most scans are within ± 1 % in RH compared to the expected values, it can be concluded that the measurement uncertainty for these salt scans are dominated by the limited precision in the temperature measurements, as previously discussed”

2) More engineering data regarding the flow rates, residence times etc. would be of use and in line with the tenor of the paper.

Answer:

2.1 Flow rates. The following passage has been added on page 4 and 5:

“The sheath flow rates in the system were set to 10 l/min for DMA1 and 6 l/min for DMA2. The aerosol flow through the system was set to 1 l/min, hence giving flow ratios of 10:1 and 6:1 respectively. A higher sheath to aerosol flow will give a higher size resolution of the system, but with the drawback of lower counting statistics and a narrower sizing interval, as higher flows need higher voltages for the selection of a particular electrical mobility, and the maximum voltage that can be used for a DMA is limited. In this work, a flow ratio of 6:1 was set in the second DMA to avoid electrical discharges in the humid air and to still be able to measure a GF of 2.2 for the largest dry size selected (265 nm)”

2.2 Residence times: The following has been added on page 6:

However, this also means that the residence time after the aerosol enters the second box has to be long enough so that the aerosol reaches the new temperature. The residence time between DMA1 and DMA2 has been approximated to 3 seconds, whereof 0.2 seconds is between the humidifier and the wall between box 1 and box 2. A longer residence time would lessen the risk of the particles not reaching equilibrium before entering DMA2, but would on the other hand increase the particle losses, a parameter which can be of importance when conducting background measurements.

3) What is the effect of doublets in the results? Is this estimated?

Answer:

The following passage has been added on page 6:

“It is assumed that the particles entering DMA2 carries only one charge. Doubly charged particles will have a slightly larger GF, as they are larger than the singly charged ones, and consequently less influenced by the Kelvin effect. It is possible to correct for this, as long as size distribution measurements of the entire aerosol is simultaneously conducted, but in previous work this effect has not been taken into account, as H-TDMA measurements are usually conducted at sizes where the probability of double charging is relatively small (<400 nm). The effect can be significant if measurements are conducted at dry sizes below the median value of a narrow size distribution, as sometimes is the case in laboratory studies, but under normal atmospheric circumstances the effect is probably small (Swietlicki et al. 2008).“

4) What is the effect of the scanning inversion on the uncertainty or error cf. eg. Collins' analysis?

Answer:

We are not sure what analysis is referred to here. The effect that scanning of the DMA2 voltage has on the raw data is discussed on page 4, which was added after the technical comments:

“However, too fast scanning will lead to problems in the inversion of the data, since

there is a smearing effect in the CPC, caused by the mixing of particles inside the particle counter and this effect will be an increasing problem as scanning speed increases. This is not an issue for stepping systems, since the particles entering the CPC is not varying with time during the measurement.“

There are a few different ways of working with the raw data, e.g. TDMA-fit from Stoltzenburg and McMurry (1986), or TDMAinv (Gysel et al. 2009). In Santarpia, Li, and Collins (2004) the authors fitted log-normal functions to the raw data to obtain the growth factors. Although this does not give the GF-probability density function, the method itself it is not dependent on if the system is scanning or stepping (more than the smearing effect previously discussed). We ask for a clarification of the question, if further analysis is wanted.

5) A statistical summary and table of the data in figure 5 would be valuable including the Kelvin effect.

Answer:

After the technical comments, Table 1 was added. It states the average GF and water activity values and their standard deviations for the salt scans. We have used water activity instead of RH on purpose, since in this way, the Kelvin effect does not have to be treated separately. Smaller dry sizes will have a lower water activity for the same dry size.

6) The case for the "smeared" deliquescence RH needs to be explained more thoroughly. Reference the paper by Lynn Russell, Scripps Institution for Oceanography and coau- thor. Sorry I don't have the citation at hand, ca. 2002.

Answer:

After the technical comments, the discussion on the smeared deliquescence was expanded and now reads as follows (page 7):

“The deliquescence RH (DRH) of the salt is somewhat smeared in the DRH measurement (Figure 2). This is not expected for a pure solution, but the effect has been seen before, especially for small particles (e.g. Hämeri and Väkevä 2000) and can possible be attributed to a contamination of the salt solution. This would be consistent with theory, as it has been shown that a mixture of ammonium sulphate and an organic acid can smear the deliquescence point (Russell and Ming 2002). It is also possible that it is a consequence of temperature gradients in the DMA, deliquescing the particles somewhere inside the DMA. However, given the relatively low RH at which this phenomenon occurs ($\sim$77%) it is unlikely that this alone is the reason, especially considering that the measured temperature gradient of the DMA is less than 0.1 K (see figure 3). Another possibility would be that the particles deliquesce near the walls of the Gore-Tex® tube, though this effect would show a bimodal distribution in the second DMA, and this was not the case."

If we have misinterpreted the paper by Russell or used the wrong paper, please let us know and we will correct this.

7) The support for small longitudinal temperature gradients is shown in figure 3. What are radial gradients?

Answer:

The radial gradients are not measured, since this would require drilling holes at different depths in the DMA, which we did not dare do. We assume that the surface temperature is the same as the inside temperature on the DMA, and this assumption is supported by the fact that the salt measurements are on the right RH value. A radial temperature gradient of 0.2 K would shift these results $\sim$1% in RH. This is commented on pages 4 and 5 as follows:

"The RH in DMA2 is calculated by the combination of the dew point temperature, Tdew, given by a General Eastern hygro M1 dew point meter in the DMA2 loop and the average value of three Pt100-elements taped to the outer wall of DMA2 at different heights (bottom, middle and upper part of DMA2). This way, the longitudinal temperature gra-

dient of the DMA is measured. However, there is no measurement of a possible radial temperature gradient in the DMA body. The temperature in the DMA is assumed to be the same as on the DMA surface."

The following sentence has been added on page 7:

"These results also support the assumption that the radial temperature gradients are negligible, since a radial temperature gradient of 0.2 K would give a shift of $\sim$ 1% in RH."

8) Figure 2. What is the dashed line? What diameter of ammonium sulfate particles was used?

Answer:

These questions are answered in the figure text: "Humidogram of ammonium sulphate for 100 nm particles. The solid and the dashed lines represents electrodynamic balance data from Tang and Munkelwitz (1994). The solid line describes the deliquescence water uptake at increasing RH and the dashed line shows the hysteresis effect when the aerosol has been pre-humidified to an RH above the deliquescence point, and thereafter dried to a given RH."

9) Figure 3 Be consistent with Pt. It is hard to see the details of the Pt100 sensors. Expand the scale or average to relevant time scale.

Answer:

Unfortunately we are not sure if we understand what is meant by "be consistent with Pt". we have found was misprint which said pt instead of Pt. If there is anything else, we ask the referee to clarify. We have also tried to make Figure 3 easier to read.

Best regards on behalf of the authors, Erik Nilsson

References

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