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**Intercomparison of
six Hygroscopicity-
TDMA
systems**

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Results and recommendations from an intercomparison of six Hygroscopicity-TDMA systems

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Abstract

The performance of six custom-built Hygroscopicity-Tandem Differential Mobility Analyzers (H-TDMA) systems was investigated in the frame of an international calibration and intercomparison workshop held in Leipzig, February 2006. The goal of the workshop was to harmonize H-TDMA measurements and develop recommendations for atmospheric measurements and their data evaluation. The H-TDMA systems were compared in terms of the sizing of dry particles, relative humidity (RH) uncertainty and consistency in determination of number fractions of different hygroscopic particle groups. The experiments were performed in an air-conditioned laboratory using ammonium sulfate particles or an external mixture of ammonium sulfate and soot particles.

The sizing of dry particles of the six H-TDMA systems was within 0.2 to 4.2% of the selected particle diameter depending on investigated size and individual system.

With regard to RH uncertainties, the H-TDMA systems showed deviations up to 4.5% RH from the set point at RH=90% investigating the hygroscopic growth of ammonium sulfate particles and comparing the results with theory.

The evaluation of number fractions investigating an externally mixed aerosol delivered differences up to $\pm 8\%$ in calculated number fraction for one and the same aerosol type.

We analysed the datasets of the different H-TDMAs with one fitting routine to investigate differences caused by the different data evaluation procedures. The results showed that the differences were reduced from $+12/-13\%$ to $+8/-6\%$. We can conclude here that a common data evaluation procedure to determine the number fraction of externally mixed aerosols will improve the comparability of H-TDMA measurements.

We finally recommend, to ensure a good calibration of all flow, temperature and RH sensors in the systems. It is most important to thermally insulate the RH control unit and the second DMA and to monitor those temperatures as accurately as 0.2°C . For a correct determination of external mixtures, it is necessary to take into account size-dependent losses due to the diffusion in the plumbing between the DMAs and in the

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1 Introduction

Aerosol particles play a significant role in terms of their influence on regional and global climate by interacting with solar radiation (Ramanathan et al., 2002) and their ability to form cloud droplets (Kaufmann et al., 2002). Hygroscopic properties of aerosol particles are crucial for the determination of the understanding of the particle's impact on regional and global climate (IPCC, 2007). These properties, measured at subsaturated conditions, are strongly linked to the particles' ability to form cloud droplets. In addition to the particles' contribution to the direct and indirect effect, their presence in the atmosphere reduces the visibility, which is an important issue in highly polluted areas (Cheng et al., 2008). This effect increases with ascending ambient relative humidity in the atmosphere.

The H-TDMA (Hygroscopicity-Tandem Differential Mobility Analyzer) system was firstly introduced by Liu et al. (1978), and in the last three decades, this technique has been established and used to measure size-dependent hygroscopic properties such as growth factors and state of mixing of submicrometer particles with high temporal resolution in various atmospheric environments (e.g. Kandler et al., 2007; Swietlicki et al., 2008). Up to now, H-TDMA systems are not commercially available and all existing systems are, thus, based on custom-built set-ups (Cubison et al., 2005; Hennig et al., 2005; Johnson et al., 2005; Massling et al., 2003; Rader et al., 1986; Weingartner et al., 2002). In general, these systems show substantial differences in technical details, used criteria for data quality and applied data evaluation procedures. Data evaluation procedures may differ in applied data analysis methods and may also account for system-dependent calibrations (Cubison et al., 2005; Gysel et al., 2008; Stolzenburg and McMurry, 1988; Voutilainen et al., 2005).

In this study, six European HTDMA systems were investigated in an international calibration and intercomparison workshop held at the Leibniz Institute for Tropospheric

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Research in Leipzig, Germany, in February 2006. This workshop was the base of, in total, three experimental H-TDMA intercomparisons.

The aim of this experimental study was to investigate the performance of those systems in terms of sizing dry particles as a function of particle diameter, RH (relative humidity) certainty of growth factor determination based on RH (relative humidity) certainty (also over of time), and determination of number fractions of externally mixed aerosol particles with different hygroscopic growth. In addition, a standard data evaluation procedure was applied to all raw data to intercompare the performance of the different H-TDMA systems reliant on the applied data evaluation procedure.

To the best of the author's knowledge, there is only one study stating a comparison between H-TDMA systems (Duplissy et al., 2009). Additionally, to the existing comparison study by Duplissy et al., we investigated the sizing uncertainties as the function of particle dry size, and the comparibility on the determination of number fractions reliant on the use of data evaluation routines. Also, the set of custom-built H-TDMA systems in this study includes four different systems that were not tested before.

On the basis of our investigations, we provide general recommendations to technically harmonize H-TDMA systems and to standardize procedures for data quality. Such recommendations are needed to assure the comparibility of measured hygroscopic properties of submicrometer particles.

2 Instruments and experimental set up

An H-TDMA consists of two Differential Mobility Analyzers (DMA) in series and an aerosol humidifier in between. A schematic is given in Fig. 1. The first DMA selects a narrow particle size fraction from the aerosol (I). This fraction is then humidified in the aerosol conditioner to a well-defined relative humidity (II). The altered, humidified size distribution is then determined by a size spectrometer (III) consisting of a second DMA in combination with a Condensation Particle Counter (CPC).

Overall, custom-built H-TDMA systems show substantial differences in technical de-

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tails. Systems compared within this study varied in the type of DMA, type of CPC, type of flow scheme (separated sheath and excess air or closed loop), DMA 1 drying section, DMA 2 humidifying section, type of RH measurement and principle of RH regulation, principle of temperature stabilization of the humidifying section and of DMA 2. Table 1 gives an overview of the differences in technical details of the operated H-TDMA systems within this comparison. Detailed descriptions of the systems compared within this study can be found in the corresponding publications (Cubison et al., 2005; Haemeri et al., 2000; Hennig et al., 2005; Massling et al., 2003; Villani et al., 2008; Zhou, 2001).

All instruments were placed in an air-conditioned laboratory operated at $T=22^{\circ}\text{C}$ and were connected to a mixing chamber. Four different experiments were carried out. For Experiment 1, 2, and 3, ammonium sulfate particles were generated (concentration of $(\text{NH}_4)_2\text{SO}_4$ in water 0.1 g L^{-1}) using an atomizer (TSI, Model 3076) and were conducted to the mixing chamber. The mixing chamber consisted of a $0.75\text{ m}\times 0.75\text{ m}\times 0.75\text{ m}$ stainless-steel box with a small blower inside to provide a homogeneously mixed aerosol. The particle flow of about 2 L min^{-1} of this atomizer was diluted by dry, particle-free, and oil-free air of approximately 4 L min^{-1} . During Experiment 4, soot particles (Printex 85) were suspended in water and atomized using a second atomizer of the same model. The soot aerosol (2 L min^{-1}) was added into the mixing chamber to provide an externally mixed aerosol together with the ammonium sulfate particles.

All systems measured simultaneously from the mixing chamber and were operated during all experiments by the same parameters in terms of selected dry size and controlled RH. Data were obtained for the dry sizes $D_p=30, 50, 70,$ and 100 nm except for system 1, which could not be operated at $D_p=100\text{ nm}$ because it was designed to investigate ultrafine aerosol particles and, therefore, the size range was limited. The aerosol was dried only by the system's built-in drying sections before entering the first DMA. As listed in Table 2, not all instruments delivered data during all four experiments.

3 Data evaluation

It was found, that the quality criteria used for the different H-TDMA systems and their retrieved data differed. Quality checks with regard to the stability in RH were done for all systems, but flow rates and temperatures were only monitored within some set ups.

Individual data evaluation procedures were used for each H-TDMA system due to the different number and type of corrections applied to the retrieved humidified particle number size distributions from each single system. Differences may occur in terms of the following corrections: CPC efficiency, diffusional particle losses in the DMA, system-dependent individual particle losses in tubings, correction for the DMA sizing of dry particles, and also due to used DMA transfer functions (experimentally derived or ideal).

Here, all individual data evaluation procedures took into account the effect of the DMA transfer function on the retrieved humidified number size distributions. Some of the individual data evaluation procedures used the experimentally derived transfer function of the used DMA and other procedures used the ideal transfer function of the used DMA having the defined shape of a triangle. All individual data evaluation procedures were corrected for the DMA sizing of dry particles. In the following description, those individual data evaluation procedures will be labelled as external data evaluation.

Also, all raw data were post-processed in the same way using a common data evaluation procedure. This data evaluation procedure included the recalculation of the humidified particle number size distribution ($dN/d\log D_p$) without accounting for multiple charging, neglecting DMA diffusion losses and individual CPC efficiencies. However, the ideal transfer function of the used DMA 2 was taken into account for this calculation. In the following, this evaluation will be labelled as internal data evaluation procedure.

In this study, the data were evaluated following both data evaluation procedures. Except for one external routine, all data evaluation procedures (external and internal) had in common the lognormal functions that were fitted to the retrieved humidified particle number size distributions to obtain mean values of single particle modes and

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the number concentration assigned to those using individual programs. For the internal data evaluation procedure one and the same fitting routine was used.

4 Results and recommendations

All results shown in the following sections are based on mean values of the parameter, in question, of the respective experiment for the individual systems. Some systems were only operated for a certain time window during the experiment. In addition, an evaluation with respect to time variability is presented.

4.1 DMA sizing of dry particles

To obtain exact hygroscopic growth factors at a chosen RH using the H-TDMA technique, it is most important that both DMAs select the same particle size during dry conditions when operated at the same size. The exact sizing of a DMA can differ from the positioned value because of several reasons. Some of these reasons are due to an incorrect calculation of the particle mobility where the ambient temperature and pressure in the DMA and the geometry of the DMA is used. Other reasons might be due to an inadequate operation of the system like sheath flow deviations from the set value, the relative humidity in the DMA, or calibrations of the high voltage power supply.

For H-TDMA systems, two DMAs are operated in series and these above-mentioned issues occur in both DMAs and may lead to a shift between the selected particle sizes of both DMAs, resulting in a growth factor during dry conditions. Therefore, the size-dependent dry-growth factors for the individual H-TDMA system under operating conditions were measured.

The dry-growth factors GF_{dry} were derived by the calculation of the ratio of the derived diameter $D_{p\text{DMA}2}$ (dry conditions) of the particle number size distribution measured with DMA2 and CPC 2 and the initial diameter $D_{p\text{DMA}1}$ (dry conditions) set by

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DMA 1 following Eq. (1).

$$GF_{\text{dry}} = \frac{D_{\rho\text{DMA}2}(\text{RH} < 20\%)}{D_{\rho\text{DMA}1}(\text{RH} < 20\%)}. \quad (1)$$

During Experiment 1, H-TDMA systems were operated during dry conditions measuring pure ammonium sulfate particles yielding the dry-growth factor (ideally 1.0) for selected dry sizes of $D_p=30, 50, 70,$ and 100 nm.

In Fig. 2a and b, the results of this experiment are shown presenting the evaluation of both procedures (external and internal data evaluation). For a better comparison, the relative difference in % from the set value is plotted in Fig. 2a and b and the nominal values of GF_{dry} are additionally listed in Table 3.

In general, shifts in sizing between the two DMAs of individual systems were between 0.2% and 4.2%, depending on the investigated particle size. The external and internal data evaluations differed within 2% on an absolute scale and only for some exceptional cases. This result shows that the application of external and internal data evaluation may lead to different results, although these evaluations are based on one and the same raw data. For some set ups, relative shifts in the DMA sizing of dry particles increased or decreased slightly with increasing particle size. For other set ups, the relative shift was nearly constant over size. This implies that the reasons for the observed discrepancies varied depending on employed system. It is assumed that deviations in DMA sheath air flows caused the observed discrepancies, at least for some of the systems. Statements on discrepancies, due to the calibration of the high voltage power supply, cannot be concluded here as these affect the DMA sizing of dry particles in general at smaller diameters than $D_p=30$ nm. Insufficient drying by the system's built-in drying sections will also be excluded as ammonium sulfate particles deliquesce at 35–40% RH and all systems were operated at smaller RH.

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Recommendations

Because the shifts in sizing between the two DMAs of individual systems were up to about 4%, we propose to correct the measured growth factors for individual system shifts based on determined dry-growth factors by the investigation of calibration salt particles as ammonium sulfate particles (recommended as showing no shape factor) during dry conditions. Dry scans should be performed regularly during field studies, at least once a week. We also suggest determining the dry-growth factors for the same particle sizes as selected for the field or laboratory studies. These dry-growth factors (see Eq. 1) should then be applied to all measured data. This recommendation is consistent with suggestions found by Duplissy et al. (2009).

In addition, high voltage power supplies should be calibrated before set up of the instrument especially for small voltages (<100 V). The calibration of the power supplies should be done after a warm up period of at least one hour. An exact calibration of the flow sensors may avoid larger deviations in the DMA sizing. The sizing of individual DMAs should be verified by Polystyrene Latex (PSL) spheres with known particle size in the operating size range before field experiments. Also, if possible, the real experimentally derived DMA transfer function should be used in the data evaluation procedure.

4.2 RH uncertainty

For a well-calibrated H-TDMA system, the largest uncertainty in hygroscopic growth factor results from RH uncertainties in the second DMA operated at humidified conditions. The RH in the system is controlled by an RH control unit, containing an RH sensor in the flow line as an input and a mechanism controlling the water vapour concentration as an output. The RH in the DMA may differ from the set value, because the temperature of the DMA and the temperature measured in the RH control unit may be different as the RH sensor is not a part of the DMA itself. DMAs consist of massive metal bodies and follow only slowly changing room temperature. Even an air-

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conditioned laboratory may show a fluctuation in temperature of about 1 to 2 K over 24 h in time. The hygroscopic growth measurement is conducted in the DMA and a temperature shift leads to incorrect hygroscopic growth calculations. An inadequate calibration of the RH sensor used for the control unit may also lead to similar uncertainties. For the following experiments, ammonium sulfate particles were generated and introduced into the mixing chamber.

4.2.1 RH deviation

To investigate and intercompare the RH uncertainty and the RH stability of the different H-TDMA systems, they were operated at 90% RH selecting initial dry diameters of $D_p = 30, 50, 70$ and 100 nm. An air conditioned laboratory stabilized the room temperature around 22 °C. For comparison, the measured hygroscopic growth factors GF measured at 90% RH were corrected for the individual dry-growth factor derived from Experiment 1. Then, the theoretical relative humidity RH_{theo} based on the experimental hygroscopic growth of the observed ammonium sulfate particles was calculated based on data given by Tang and Munkelwitz (1994). The measured relative humidity $RH_{measured}$ was determined in the RH control unit, hence $RH_{measured}$ is different for every system. The deviation in RH, here ΔRH , was calculated as follows:

$$\Delta RH = RH_{theo} - RH_{measured} \quad (2)$$

Fig. 3a and b show the ΔRH for external and internal data evaluation for the individual H-TDMA systems. In addition, the derived hygroscopic growth factors are listed in Table 3 together with the theoretical growth of ammonium sulfate particles at 90% RH reported by Tang and Munkelwitz (1994).

In general, system 1 shows higher relative uncertainties in growth factor than other systems for both types of evaluations. The reason was found to be technical problems with the temperature control of DMA2. The DMA2 itself was cooled slightly with water circulation, but insufficient insulation of the DMA2 enclosure caused the controlling RH sensor to be roughly 1 °C warmer than the DMA body itself. For system 2, deviations

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in hygroscopic growth factors from theoretical values are smaller than 7% resulting in differences in RH at 90% up to 3.5% RH. The reason for this is unclear. Temperature fluctuations can attribute for some of the deviation, but this does not explain the dry size offset, which is most likely due to problems with the flow control, since voltage calibration is in general only a problem for smaller particles, but not for larger sizes (as e.g. 100 nm dry size). For system 5 and 6 a difference in RH at 90% smaller than 1.5% RH was found. In total, a size dependent error, which is decreasing for system 1 and increasing for system 2 with increasing investigated size was observed. This tendency for system 1 and 2 as well as the calculated absolute deviation in RH for all systems was found independently of the type of evaluation except for system 2 evaluated for $D_p=50$ nm. Discrepancies were most likely due to temperature gradients in the systems and to insufficient calibrations of the RH control units. The high uncertainty for system 1 and 2 was revealed within the test which shows this test as a good tool to investigate the performance of the H-TDMA system during application in the field or laboratory. Similar results were obtained for both types of evaluation having in mind that each type of evaluation procedure included a correction of DMA sizing evaluated by the same data evaluation procedure.

4.2.2 RH stability

Figure 4a and b show the time series of recalculated relative humidity based on the procedure introduced in Sect. 4.2.1. Here measurements of ammonium sulfate of about 12 h obtained at RH=90% (setpoint) and $D_p=50$ nm are illustrated. Unfortunately, this experiment is lacking data from systems 3 and 4 and only a few data points are available for system 5. It can clearly be seen that all RH measurements follow a more or less constant gradient resulting in a slowly decreasing RH over time (about eight hours). A substantial change in RH occurred after a restart of most of the systems when a power failure occurred in the laboratory. External and internal evaluations reflect observations obtained in Sect. 4.2.1 as differences from the RH setpoint are smaller observed for externally evaluated data in comparison to internally evaluated data. However, the

general pattern in determined RH during this experiment can be seen in both evaluations. This pattern in determined RH implies that a parameter changing with time and in terms of hours is responsible for the observations. This drift in determined RH value is likely not due to inaccuracies in RH control unit calibration, but rather more likely a result of temperature instability of the RH control unit or the DMA 2 body itself.

4.2.3 DRH deviation

The accuracy of the RH within the H-TDMA systems was tested in Experiment 3 by determining the deliquescence relative humidity (DRH) of ammonium sulfate. The DRH observed as an offset in hygroscopic growth was expected at 79.97% RH for ammonium sulfate particles (Seinfeld and Pandis, 1998). In the transition zone at relative humidities near the DRH, sometimes non-deliquesced and deliquesced particles were measured simultaneously. This observation can be explained by slightly a non-homogeneous relative humidity in DMA 2 or by multiply charged particles with larger diameters behaving differently because of the Kelvin effect.

During this experiment, the systems were operated at $D_p=70$ nm increasing the RH from 75% to 85% yielding the offset in hygroscopic growth as DRH of ammonium sulfate particles. The following procedure was chosen for the internal data evaluation to obtain the DRH from the measured humidified particle number size distributions: the distributions were processed following the internal data evaluation procedure fitting one lognormal function when finally only one particle mode was observed. The following four parameter exponential equation was fitted to the RH-dependent growth curve.

$$GF = GF_{\max} - \left\{ \frac{GF_{\max} - GF_{\min}}{1 + \exp[(RH - DRH)/S]} \right\} \quad (3)$$

GF_{\max} is the maximum GF and GF_{\min} is the minimum GF in the S-curve in Fig. 6, DRH is the relative humidity in the inflexion point and S presents the slope of the S-curve. The shape of this S-curve in Eq. (3) and the four parameters are displayed in Fig. 5.

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To determine the parameters GF_{\max} , GF_{\min} , DRH, and S reasonable starting parameters were chosen. Afterwards, the difference of the square from the measured GF and calculated GF was calculated and using the residium of the sum of squares, the best parameters were obtained. The DRH values of ammonium sulfate particles derived during Experiment 3 are listed in Table 3. For a better illustration, the difference in DRH, ΔDRH , was calculated as follows:

$$\Delta\text{DRH} = \text{DRH}_{\text{theo}} (\text{here} = 79.97\%) - \text{DRH}_{\text{measured}} \quad (4)$$

For the external data evaluation, the $\text{DRH}_{\text{measured}}$ was obtained by different individual routines. Figure 6a and b show the differences in DRH, labeled ΔDRH , calculated for the individual systems based on both, external and internal data evaluation.

Based on individual set ups of the H-TDMA systems and problems listed in Sect. 4.2.1, the $\text{DRH}_{\text{measured}}$ was observed at different values hinting that the individual systems were not operating exactly at the RH set point. However, calculations for both types of evaluation resulted in similar values. Also, a consistent underestimation in DRH was observed for all systems, which could also be explained by contamination of the salt solution (Russell et al., 2002). The maximum ΔDRH was about 5% for system 1 and system 4 and the minimum was about 1% for system 2, 5 and system 6. Three out of five systems were close together and determined the theoretical DRH of ammonium sulfate particles within less than 1.3% RH. The high ΔDRH determined for system 1 and system 4 can be explained by technical problems stated above and this deviation was observed also during Experiment 2 (see Sect. 4.2.1).

Recommendations

We strongly recommend to thermally insulate the RH control unit as well as the DMA 2 body itself. An exact temperature measurement as close as possible to the inner housing of the DMA 2 (sensors could be attached to or better built in the wall of the outer DMA housing) and an exact measurement at the RH control unit provide the

possibility of calculating the RH inside DMA 2. Deviations between the RH set point and the RH inside DMA 2 based on temperature differences can, thus, be considered and corrected.

All relative humidity sensors within H-TDMA systems should be calibrated by means of a dew point sensor or saturated salt solutions on regular intervals. We propose to doing this calibration once a year and at least once before the instrument is set up for a field or laboratory study.

In addition, the automated generation of salt particles with known theoretical growth, in combination with their hygroscopic growth measurement, can be used to obtain the real RH inside DMA 2 on regular intervals. This procedure can be supported by DRH measurements using different salt solutions, e.g. sodium chloride, ammonium sulfate and potassium chloride to cover the RH range from 74 up to 86%. The DRH measurement gives a clear measurement as to whether the system is operating at the set point of RH or not, when assured that the RH in the aerosol line is not above the RH in the sheath air line. The combination of these procedures will assure the data quality of H-TDMA systems in terms of RH uncertainty and temperature instability causing most probably the highest uncertainties in hygroscopic growth factor determination. A supplementary measurement of humidograms of e.g. ammonium sulfate particles assures the correct functioning of the H-TDMA system in its operational RH range. Similar recommendations are reported by Duplissy et al. (2009), who investigated the performance of several different H-TDMA systems when determining ammonium sulfate humidograms.

4.3 Determination of number fractions

The number fraction of individual hygroscopic growth groups is an additional important parameter provided by H-TDMA measurements, as in most cases external particle mixtures are present in the atmosphere (Swietlicki et al., 2008). The H-TDMA system is the only instrument which provides an online method to determine the hygroscopic mixing state. Thus, within our study, we performed an experiment to intercompare the H-

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TDMA instruments with respect to their accuracy of determination of number fractions in case of external mixtures. This experiment focused on the ability of the different instruments to distinguish qualitatively between different hygroscopic growth groups and to quantitatively determine the number fractions of these groups for intercomparison.

5 4.3.1 Qualitative separation of different hygroscopic growth groups at varying RH

Within Experiment 4, an externally mixed aerosol was generated by mixing two particle groups with different hygroscopic properties. Here, ammonium sulfate and soot particles were produced using two TSI-type atomizers.

10 The H-TDMA systems were operated at initial dry diameters of $D_p=50$ nm at humidified conditions of (1) RH=75% and (2) RH=85%. As a general result, all H-TDMA instruments only observed one single particle group at (1) RH=75%. Growth factors were close to 1 indicating that ammonium sulfate particles were not in solution yet as theoretically expected. After a change in the conditioning set point at (2) RH=85%,
15 a less hygroscopic particle mode was measured by all H-TDMA instruments assuming a mixture of soot and ammonium sulfate as chemical composition. This particle mode was accompanied by a second more hygroscopic particle fraction with growth factors close to those of ammonium sulfate. It is expected that coagulation processes caused the loss of pure soot and pure ammonium sulfate particles in the mixing chamber.
20 Thus, only chemically mixed particles were observed. In any case, all systems observed the deliquescence of ammonium sulfate being at least one compound of the observed particles.

The number fractions NF_i of the observed hygroscopic growth groups can be calculated dividing the mode number concentration by the total number concentration of all

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modes following Eq. (5).

$$NF_i = \frac{N_i}{\sum_{i=1}^n N_i} \quad (5)$$

The number concentration of each individual hygroscopic growth group can be derived as a result of the lognormal fit to the measured humidified size distribution ($dN/d\log D_p$) after applying the external/internal data evaluation procedure to the data.

Figure 7 shows the consistency of the derived number fractions (mean values) during the operation of the H-TDMA systems at RH=85%. For the external data evaluation differences up to $\pm 8\%$ ($\pm 7\%$ internal data evaluation) in calculated number fraction were observed. Also, the external and internal data evaluations showed, at most, relative differences of $\pm 9\%$ in determining less or more hygroscopic fractions for individual systems. It has to be stated, that these results are based on mean values. The differences in observed values show that even if qualitatively the instruments could distinguish between different particle groups, the quantitative values for the determined number fractions differed depending on the instrument as well as on the data evaluation procedure used.

4.3.2 Quantitative determination of number fractions

Within Experiment 5, the same set-up as for Experiment 4 was chosen (see Sect. 4.3.1). The H-TDMA systems were operated at initial dry sizes of $D_p=30, 50, 70, 100$ nm at 90% RH.

In general, for all systems, an external particle mixture yielding two dominant different hygroscopic growth groups was observed as expected from Experiment 4. Because of unknown reasons, a nearly hydrophobic particle group was also observed sometimes and only for some of the investigated particle diameters for individual instruments. This might be due to a slightly varying concentration of the individual particle groups produced by the atomizers and, thus, resulting in varying coagulation probabilities. It is

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also possible that this mode was produced by residues dissolved in the water itself. This group only occurred with extremely small number fractions. The humidified number size distributions measured in the second DMA was divided into one less hygroscopic particle group ($GF=1.2-1.5$) (probably because of reasons dependent on the set-up as stated in Sect. 4.3.1) and one more hygroscopic particle group ($GF>1.5$) with hygroscopic growth factors close to those of pure ammonium sulfate.

In Fig. 8a and b, the time series of externally and internally determined number fractions derived from Experiment 5 are presented for all H-TDMA systems and for an initial particle size of $D_p=50$ nm. Figure 8a and b show the relative difference of determined number fractions to the mean number fraction calculated from all the instruments for the current evaluation routine. This illustration was chosen because it cannot be excluded and that a change in partitioning between the less hygroscopic and more hygroscopic particle fractions happened during the experiment. Results of this experiment are additionally summarized in Table 3. The internally evaluated data show a much smaller variation here over time for all instruments. Absolute deviations from the mean value are approximately $+12/-13\%$ for externally evaluated data including high fluctuations over time, while for internally evaluated data the differences are only up to $+8/-6\%$ with smooth fluctuations. We assume that the small differences for internally evaluated data are due to the standardized method for the number fraction calculation. Above all, inadequate fitting routines can lead to uncertainties in the determination of number concentrations in lognormal size distributions.

Recommendations

We strongly recommend recalculating the obtained H-TDMA raw data using the individual, experimentally derived transfer function of DMA 2. At least, a recalculation of the $dN/d\log D_p$ distribution using the ideal transfer function has to be applied for a further correct determination of the number of particles assigned to a single hygroscopic growth group. Since TDMA measurements do not provide information about the num-

ber of larger particle sizes as those selected in DMA 1, a multiple charge correction is not possible based on TDMA data only. A data evaluation procedure should be able to correct for individual CPC efficiency functions, DMA diffusion losses, and system-dependent particle losses. As these parameters are size-dependent, they may have significant influence on the obtained humidified particle number size distributions in the second DMA and, therewith, on calculated number fractions of individual hygroscopic particle groups.

5 Summary

Six HTDMA systems were compared within a workshop in terms of sizing as function of particle diameter, RH uncertainty, and determination of number fractions of externally mixed particles.

Sizing uncertainties as function of particle diameter were found in the range of 0.2% to 4.2% compared to the selected size depending on the individual instrument. Here, we recommended investigating the sizing uncertainty of the H-TDMA before field and laboratory studies. The growth factors (measured during dry conditions) can then be used in the data processing to correct the measured hygroscopic growth factors.

A comparison in RH uncertainty and RH stability showed that a poor temperature stabilization of the RH control unit and of the DMA 2 (operated at humidified conditions) can lead to substantial uncertainties in determining hygroscopic growth factors. Except for one system, which had problems in the technical set-up, the difference between the measured value in the RH control unit (sometimes slightly differing from the set value of RH=90%) and the calculated RH based on growth factors of ammonium sulfate was at most 3.5% in RH. The DRH value of ammonium sulfate particles was underestimated by up to 4.7% in RH, including a comparison of five out of the six investigated H-TDMA instruments. Here, we recommend strongly in performing a sufficient calibration of the RH sensors before field and laboratory studies. Furthermore, it is recommended to do continuous and precise measurements of temperature gradients

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between the RH control unit and DMA 2. In addition, we propose the recalibration of RH by the measurement of salt solutions with known hygroscopic growth as a routine on regular intervals during operation. Similar recommendations are reported by Duplissy et al. (2009) stating that temperature instabilities within the instrument are the most important parameter affecting the certainty of hygroscopic growth factors.

A comparison of number fractions of a mixture of ammonium sulfate and soot particles using different evaluation routines showed significant discrepancies. Number fractions varied about +12/−13% (external data evaluation) and +8/−6% (internal data evaluation) from a mean value.

The fluctuation in determining number fractions by some H-TDMA systems was quite high, which can only partly be explained by a probable change of partitioning between less and more hygroscopic particle fractions during this experiment. Furthermore, we recommend a more accurate evaluation in real particle number concentration assigned to individual hygroscopic growth groups. This can be achieved by taking into account losses and efficiency functions of the individual parts of the system, as well as using a robust data fitting routine. In order to avoid the application of individual data fitting routines, we suggest using the TDMA_{inv} algorithm provided by Gysel et al. (2009). As a large advantage, this algorithm provides the possibility in obtaining number fractions for selected growth factor ranges. This procedure avoids any uncertainties in the determination of number fractions based on different fitting routines, but rather calculates further the integral in a defined size range of the humidified size distribution treating different humidified size distributions equally.

In general, the differences between externally evaluated and internally evaluated data were very small for measurements of particle sizing and RH uncertainty with some single exceptions. However, substantial differences were observed for determined number fractions in case of external mixtures assuming that inadequate fitting routines were the reason for this finding.

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Acknowledgements. The H-TDMA calibration and intercomparison workshop held at the WMO-GAW World Calibration Centre for Aerosol Physics, Leipzig, Germany, was supported by the EU-projects ACCENT (GOCE-CT-2004-505337), EUSAAR (RII3-CT-2006-14 026140) and by the German Federal Environment Agency (FKZ: 351 01 046: GAW World Calibration Centre).

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Table 1. Differences in technical details of operated H-TDMA systems during the comparison.

Nr.	Technical detail	Differences in technical details
1	DMA	Vienna type (11, 28 cm), TSI type
2	CPC	Different models of TSI butanol based CPCs
3	Sheath air DMA 1/DMA 2	Closed loop set up, Sheath and excess air separated
4	Sheath air DMA 1 drying	Silica gel dryers, Perma pure driers
5	Humidification section	Perma pure humidifiers (gas to gas and water to gas humidification) – mixing of wet and dry air flows, Gore tex tubing with temperature control for humidification.
6	T stabilization of humidification section	None, Water bath, isolated box
7	RH measurement	Different models of capacitive sensors (Vaisala, Rotronic), Self-built sensors, Dew point mirrors
8	T stabilization DMA 2	No isolation, Armaflex isolation, Water isolation, Water bath, Temperature controlled water bath

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Table 2. List of instruments operated during the different experiments.

Experiment Nr.	Aerosol	Objective	Sys. 1	Sys. 2	Sys. 3	Sys. 4	Sys. 5	Sys. 6
1	AS	DMA sizing	+	+	+	+	+	+
2	AS	RH uncertainty and stability	+	+	–	–	+	+
3	AS	DRH deviation	+	+	–	+	+	+
4	AS + soot	Differentiation between two hygroscopic growth modes	+	+	–	+	+	+
5	AS + soot	Determination of number fractions	+	+	–	–	+	+

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Table 3. Mean dry-growth factors (DMA sizing of dry particles) (Exp. 1), hygroscopic growth factors (Exp. 2), deliquescence relative humidities (Exp. 3) derived for ammonium sulfate particles based on internal and external data evaluation procedures and more hygroscopic number fractions derived for the externally mixed aerosol (Exp. 5).

	AS theor. value	Sys. 1 ext. eval.	Sys. 1 int. eval.	Sys. 2 ext. eval.	Sys. 2 int. eval.	Sys. 3 ext. eval.	Sys. 3 int. eval.	Sys. 4 ext. eval.	Sys. 4 int. eval.	Sys. 5 ext. eval.	Sys. 5 int. eval.	Sys. 6 ext. eval.	Sys. 6 int. eval.
Experiment 1: DMA sizing													
$D_p=30$ nm	1.00	1.02	1.00	1.00	0.99	0.99	0.97	1.02	1.02	0.99	0.99	1.02	1.02
$D_p=50$ nm	1.00	1.01	1.00	0.98	0.97	1.01	1.00	1.01	1.02	0.99	0.99	0.98	0.98
$D_p=70$ nm	1.00	1.02	1.01	0.97	0.98	1.02	1.02	1.02	1.02	1.00	1.00	0.97	0.97
$D_p=100$ nm	1.00	–	–	0.96	0.96	1.03	1.03	1.01	1.02	1.00	1.00	0.97	0.97
Experiment 2: RH uncertainty													
$D_p=30$ nm	1.61	1.78	1.79	1.63	1.61	–	–	–	–	1.64	1.64	–	–
			+/-	+/-	+/-	+/-				+/-	+/-	+/-	
		0.04	0.04	0.04	0.02					0.05	0.05		
$D_p=50$ nm	1.66	1.82	1.82	1.62	1.57					1.69	1.69	1.69	1.69
			+/-	+/-	+/-					+/-	+/-	+/-	+/-
		0.04	0.05	0.04	0.02	–	–	–	–	0.07	0.07	0.00	0.00
$D_p=70$ nm	1.68	1.84	1.84	1.58	1.58	–	–	–	–	1.70	1.70	1.67	1.67
			+/-	+/-	+/-					+/-	+/-	+/-	+/-
		0.05	0.05	0.04	0.01					0.07	0.07	0.00	0.00
$D_p=100$ nm	1.70	–	–	1.57	1.56	–	–	–	–	1.72	1.72	1.67	1.67
				+/-	+/-					+/-	+/-	+/-	+/-
				0.02	0.01					0.08	0.08	0.00	0.00
Experiment 3: DRH deviation													
DRH, $D_p=70$ nm	79.97	75.11	74.76	79.14	78.82	–	–	75.54	75.43	78.73	78.73	78.86	78.86
Experiment 5: number fractions 50 nm (90%)													
NF (more hygroscopic mode)		0.49	0.48	0.41	0.50	–	–	0.47	0.49	0.49	0.49	0.43	0.43
		+/-	+/-	+/-	+/-					+/-	+/-	+/-	+/-
		0.02	0.02	0.02	0.02					0.02	0.02	0.02	0.02

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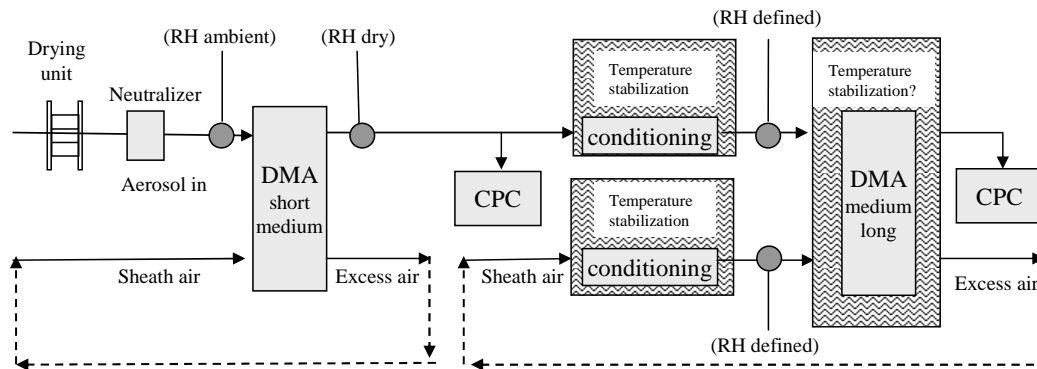


Fig. 1. Schematic picture of the custom built H-TDMA systems used within this study. Systems showed varying technical details. The dotted lines show the possibility for a closed loop system realized by some of the used set ups. The sheath air of the second DMA can also be used for humidification of the aerosol sample air, which is not shown in the sketch, but realized for some of the used set ups.

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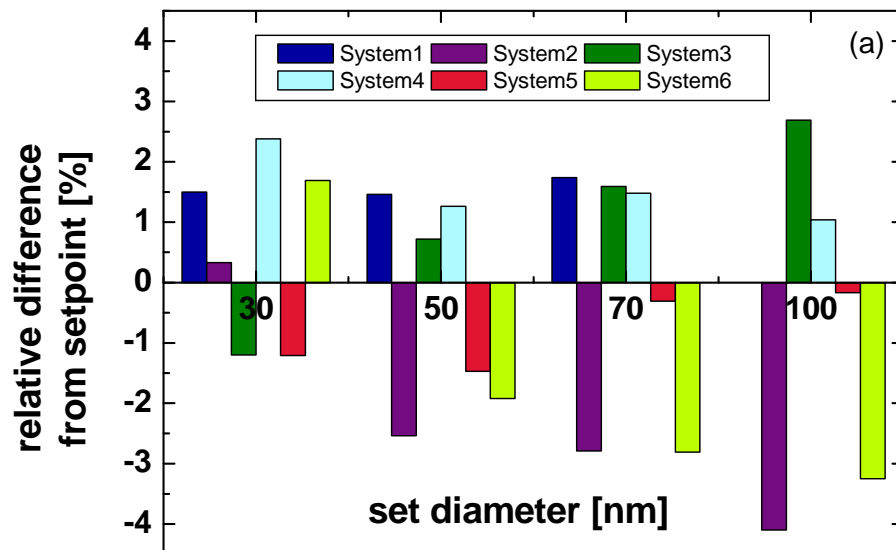


Fig. 2a. (a) Externally and (b) internally evaluated growth factor difference for all systems at dry conditions.

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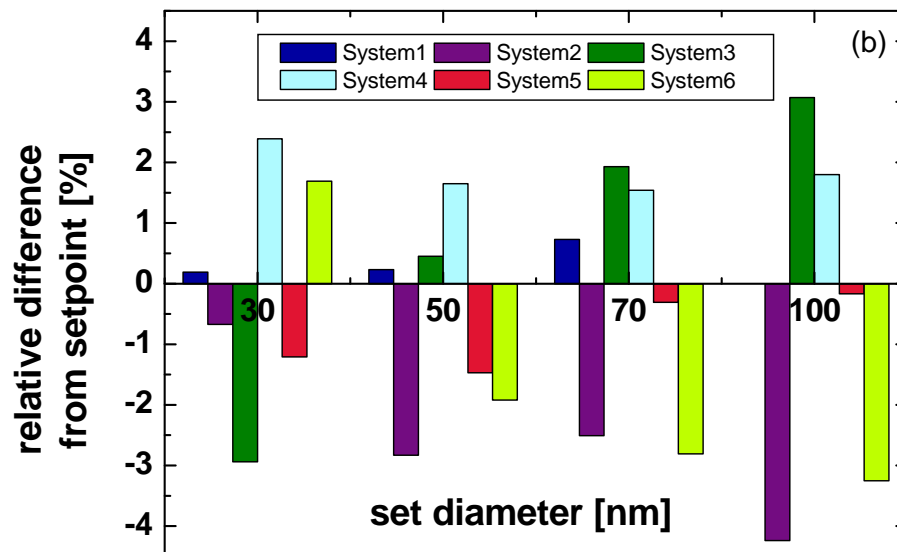


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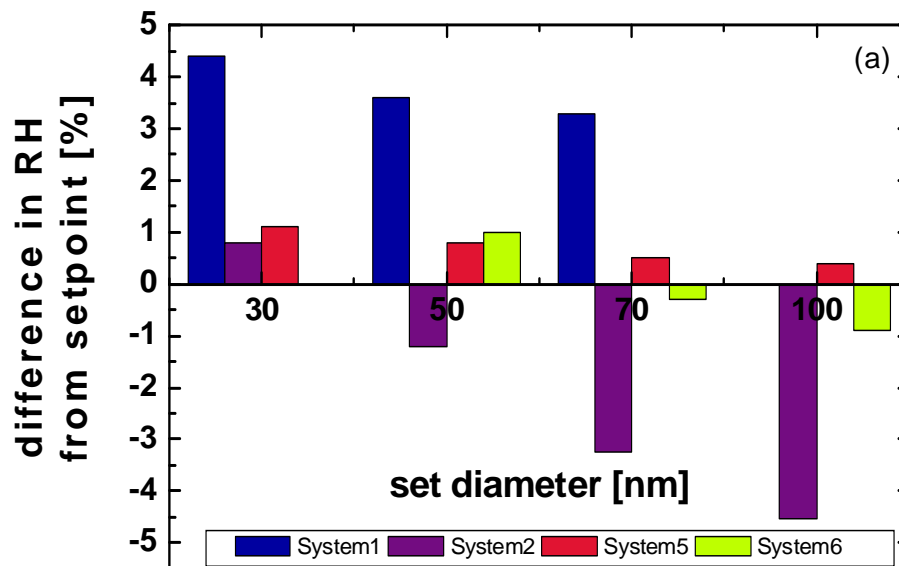


Fig. 3a. (a) Externally and (b) internally evaluated RH difference for all systems and $D_p=30$, 50, 70, and 100 nm measured at a setpoint of RH=90%.

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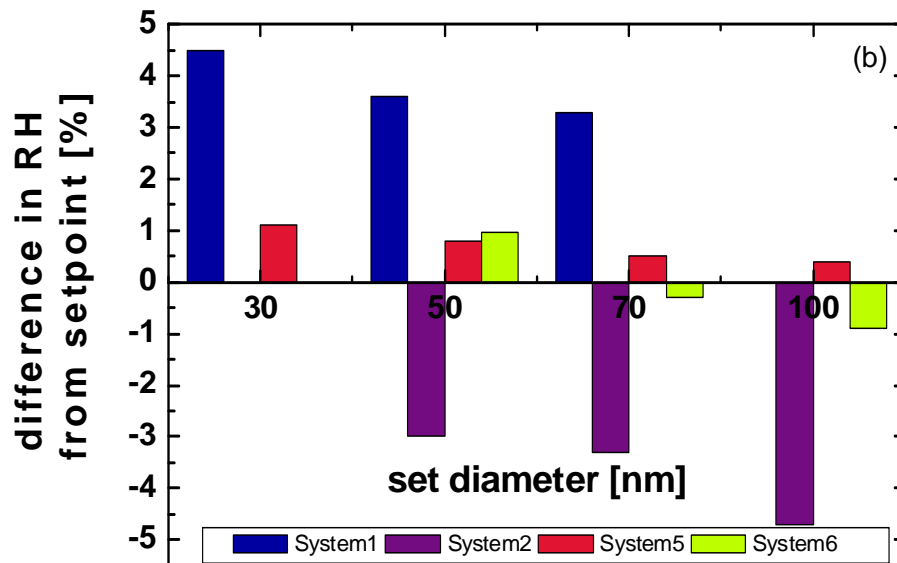


Fig. 3b. Continued.

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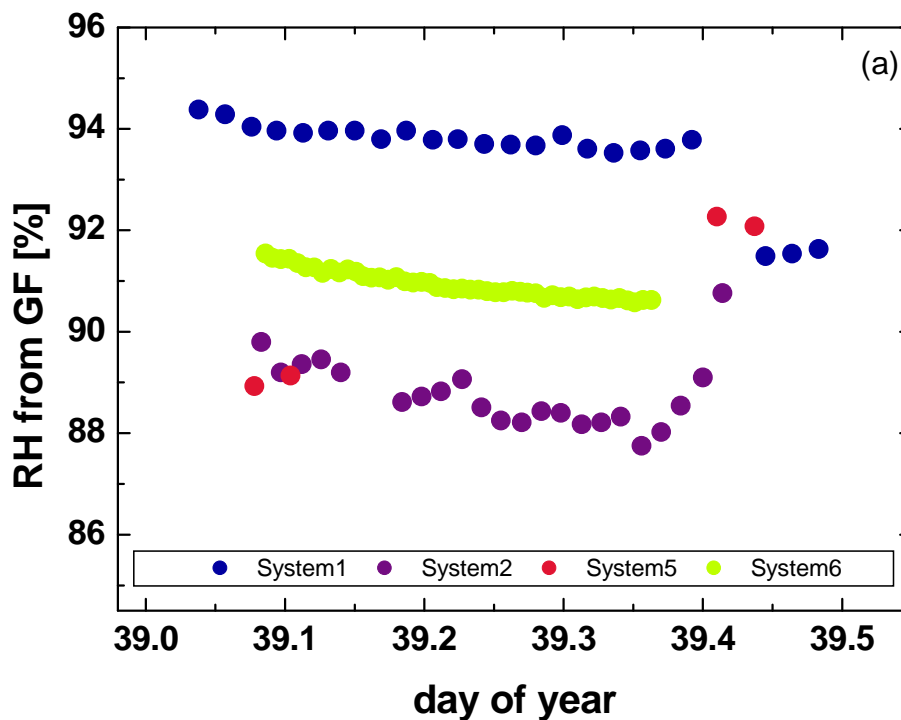


Fig. 4a. Time series of (a) externally and (b) internally evaluated RH difference for all systems measured at a dry diameter of $D_p = x$ nm. The set point during this experiment was RH=90%.

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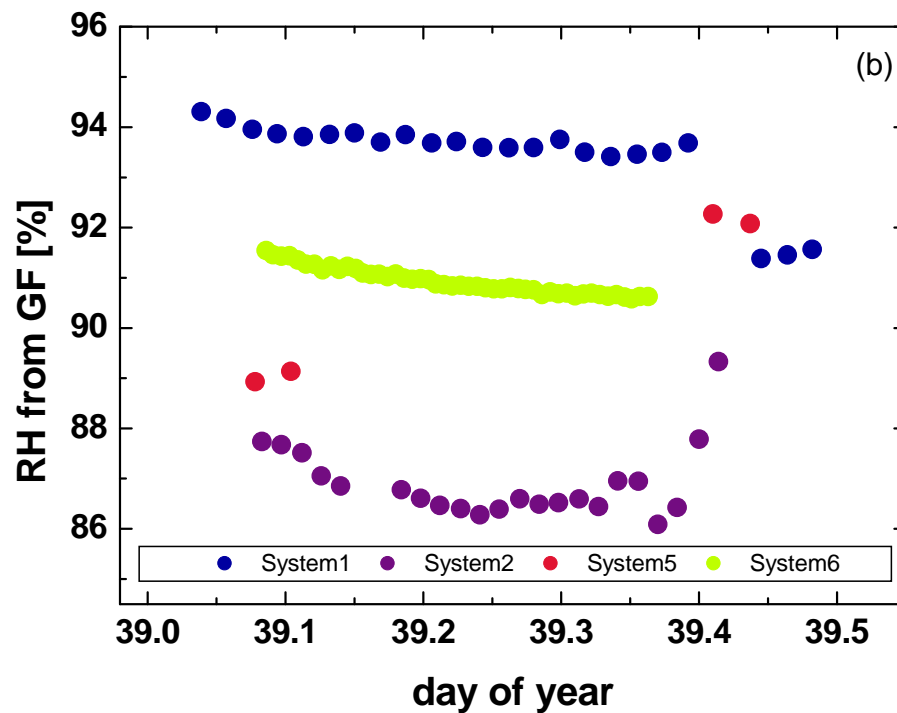


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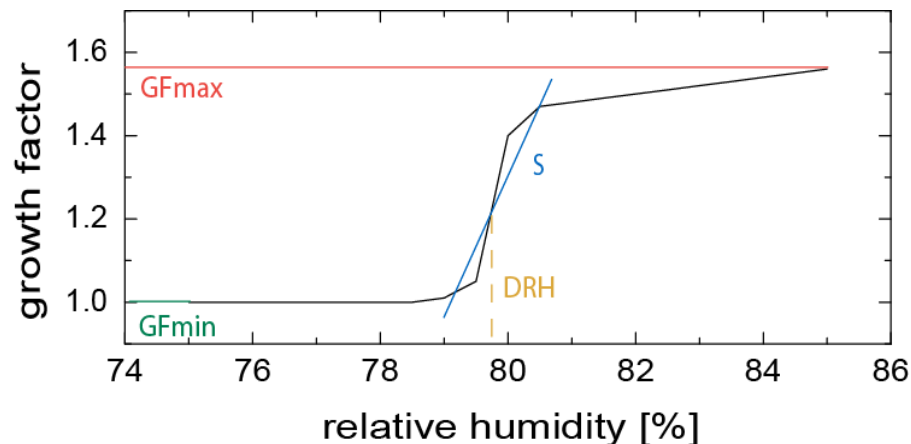


Fig. 5. Schematic of the determination of the DRH (deliquescence relative humidity) using the internal data evaluation procedure. The blue line represents the mean slope of the S-curve fitted to the data.

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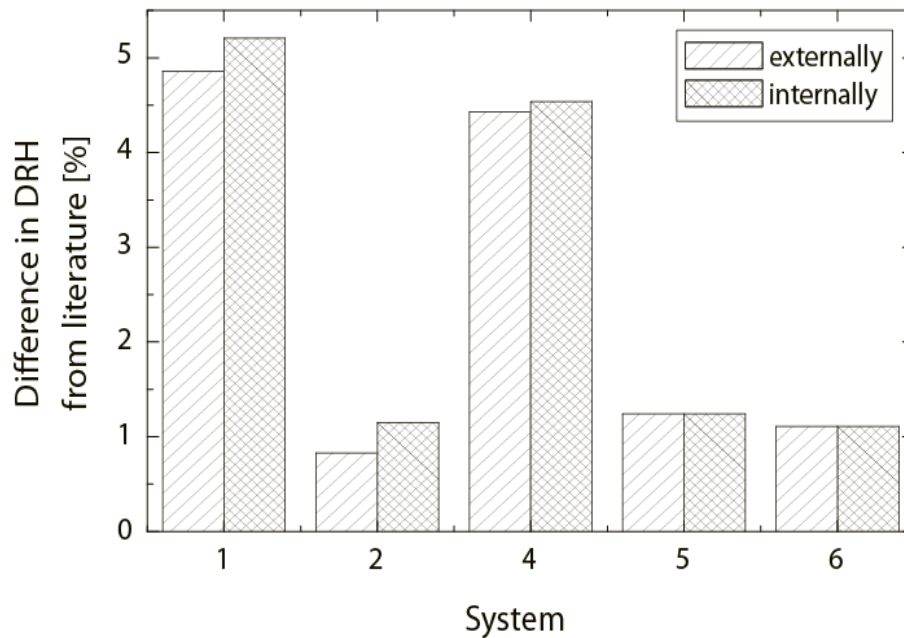


Fig. 6. Externally and internally evaluated DRH for all systems measured at a dry size of $D_p=70$ nm.

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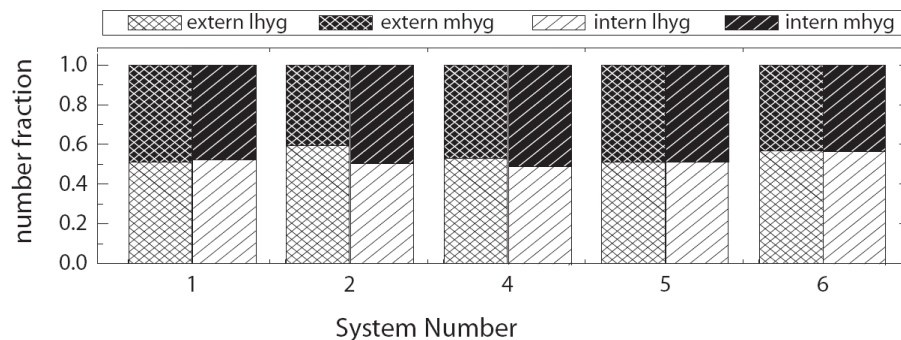


Fig. 7. Externally and internally evaluated mean number fractions of less (lhyg) and more (mhyg) hygroscopic particles of all systems measured at a dry diameter $D_p=50$ nm and at RH=85%.

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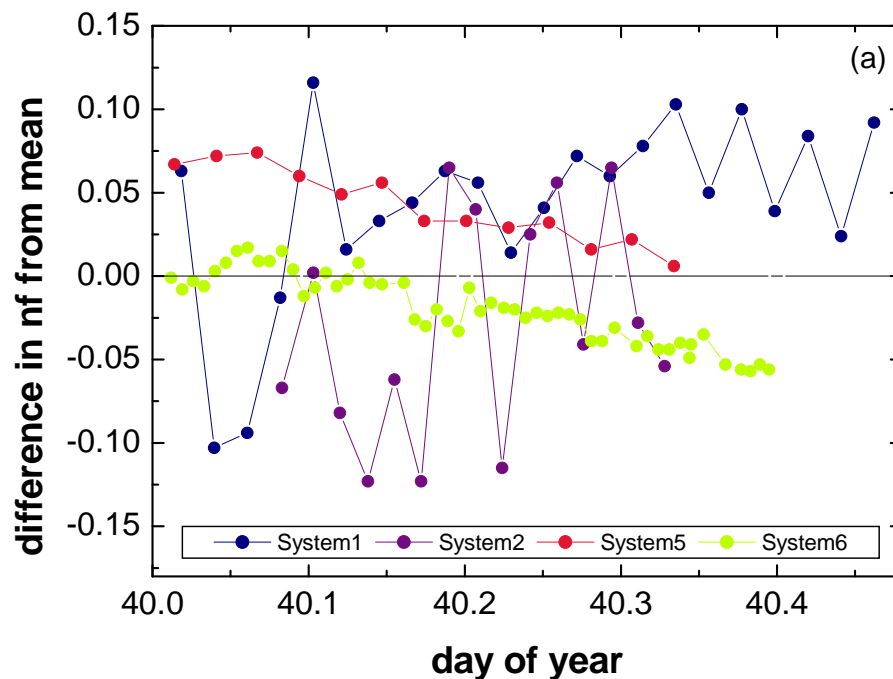


Fig. 8a. Time series of (a) externally and (b) internally evaluated difference in number fraction from the mean value of less hygroscopic and more hygroscopic particles measured at a dry diameter of $D_p=50$ nm and at RH=90%.

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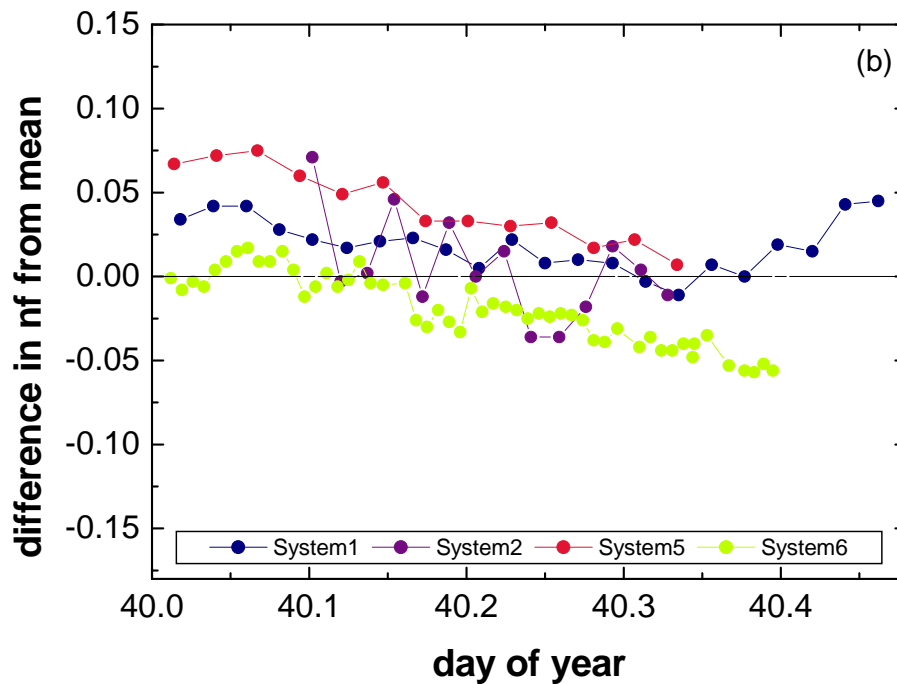


Fig. 8b. Continued.

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