

## **WCCAP Report**

### **H-TDMA workshop at the Leibniz-Institute for Tropospheric Research in Leipzig, Germany, February 2006**

Hygroscopic properties of aerosol particles are crucial parameters for the determination of the understanding of the particle's impact on regional and global climate. One of the most common methods to examine these properties is the H-TDMA (Hygroscopicity-Tandem Differential Mobility Analyzer) technique. The principle of the H-TDMA has been used for more than twenty years to investigate hygroscopic properties of submicrometer aerosol particles with high temporal resolution. In general, these systems are home-made set-ups („custom built system“) and show substantial differences in technical details, used quality criteria, and applied data evaluation.

A workshop comparing six European H-TDMA systems was held in February 2006 at the Leibniz-Institute for Tropospheric Research in Leipzig, Germany. The workshop was divided into an experimental section and a discussion section.

It was the aim of the experimental part to intercompare the performance of those systems in terms of particle dry sizing, RH (Relative Humidity) control and RH stability (certainty of growth factor determination), and determination in observed number fractions in case of external mixtures.

Additionally, the set-up of a standardized H-TDMA system and a standardized data evaluation procedure was discussed within the workshop.

In general, 18 scientists participated in the workshop, that was funded by ACCENT (Atmospheric Composition Change: The European Network of Excellence).

Within the H-TDMA workshop from 06.02.07 – 10.02.07 the following institutions represented by the listed scientists were present.

<b>European Institute</b>	<b>Workshop participant (experiment)</b>	<b>Workshop participant (discussion)</b>
Lund University (Sweden)	Erik Swietlicki, Erik Nilsson, Jakob Löndahl, Jenny Rissler (one system)	Erik Swietlicki, Erik Nilsson, Jakob Löndahl
University of Helsinki (Finland)	Kaarle Hämeri, Mikael Ehn (one system)	Kaarle Hämeri, Mikael Ehn
Laboratoire de Météorologie Physique (France)	Paolo Villani (one system)	Paolo Laj, Karine Sellegri, Paolo Villani
University of Manchester (United Kingdom)	Nick Good (one system)	Nick Good
Leibniz-Institute for Tropospheric Research (Germany)	Tabea Hennig, Nicole Kaaden, Andreas Nowak, Andreas Massling, Alfred Wiedensohler (two systems)	Tabea Hennig, Nicole Kaaden, Alfred Wiedensohler, Andreas Massling,
Stockholm University (Sweden)	-	Johan Ström, Radovan Krejci
University of Kuopio (Finland)	-	Petri Vaattovaara, Laura Hiltunen
Academy of Sciences of the Czech Republic (Czech Republic)	-	Pavla Dohanyosova

# **Minutes: H-TDMA workshop in February 2006 at the Leibniz-Institute for Tropospheric Research in Leipzig, Germany (Part I)**

Within the discussion section of the H-TDMA workshop the following points were discussed.

## **Points of discussion:**

- Differences in system set up
- Differences in quality criteria
- Differences in data evaluation
- Differences in data inversion schemes / recalculation algorithms
- How can a future system look like?
- Evaluation of the workshop data
- Publication
- Time schedule

## **Differences in system set up**

The discussions showed, that all systems were based on substantially different technical details.

	Technical detail	Differences in technical details
1	DMA	Type: Hauke short, Hauke medium, TSI type
2	CPC	Some systems contained only one CPC, others contained two CPCs. Only TSI butanol based CPCs were used. Different models were used.
3	Sheath air DMA 1 and sheath air DMA 2	Some systems were built using a closed loop set up, others were built as open systems - need of pressurized air and of a vacuum pump.
4	Sheath air DMA 2 and aerosol sample air humidification	Some systems used perma pure humidifiers (gas to gas and water to gas humidifiers), others used gore tex tubing with temperature control for humidification.
5	Sheath air DMA 1 drying	Systems without closed loop set up needed dry compressed and oil-free air, drying sections of systems with a closed loop set up were based on silica gel dryers.
6	RH measurement	RH sensors ranged from capacitive sensors from Vaisala and Rotronic to self-made sensors. Some systems contained a dew point mirror, others did not contain a dew point mirror.
7	T stabilization	Some systems contained a temperature controlled or temperature-uncontrolled water bath for DMA 2, others contained a temperature controlled water tubing around DMA 2. Most systems did not have a temperature control of DMA 2.
8	Computer language	Most systems were based on the computer language LabView.
9	Voltage control	All systems were based on a procedure stepping the voltage of DMA 2.

### Differences in quality criteria

The discussions showed, that all groups were applying substantially different quality criteria evaluating their H-TDMA data.

	Quality criteria	Differences in quality criteria
1	Flows	Only some systems monitored flows and those were checked for individual measurements.
2	Temperatures	Only some systems monitored temperatures and those were checked for individual measurements.
3	RH	All systems monitored RH values. Different procedures were used to check the data for quality: dew point mirror, capacitive sensors, salt calibrations, etc.
4	CPC 1 /CPC 2 closure	For some systems containing two CPCs, an internal particle number closure was provided.

Quality control and quality assurance were found to be two different things. Quality control is what is done during the experiment and quality assurance is what is done before and also after the experiments to check the obtained data for quality.

### Main points - in terms of flow control we note/need:

- water vapor changes density of air; thus, the volume changes, that is given by the MFC
- for the sheath air the change of the volume flow is just few %, but it has to be taken into account for the aerosol/sample flow

- flow needs to be measured at the operating RH of the system
- pressure drop inside the system has to be taken into account

**As a solution we note/propose:**

- independent measurements are necessary
- inline measurements of flows are useful
- inline measurements of flows should be no problem for closed-loop systems

**Main points - in terms of temperature monitoring we note/need:**

- calibration in the lab (normally: long-term stability of the temperature sensors / calibration)
- fluctuation in growth due to temperature fluctuations (therewith RH fluctuations)
- measurement of gas temperature is tricky
- accuracy of the temperature sensors  $\pm 0.1$  K (or better)
- define temperature in the 2<sup>nd</sup> DMA  $\pm 0.2$  K/ $\pm 0.5$  K (?)

**As a solution we note/propose:**

- useful to have more massive DMAs for higher temperature stability
- better: measure body temperature of the DMA; better place temperature sensors in the housing of the 2<sup>nd</sup> DMA
- useful to measure the temperature inside the DMA – not possible up to now – temperature measurements should be close to the inside of 2<sup>nd</sup> DMA
- measure as many temperature as possible

**Main points - in terms of RH control and RH monitoring we note/need:**

- dew point mirror is best instrument to measure relative humidity (RH), because it is a temperature measurement
- dew point mirror is too slow to regulate the RH – dew point mirror is only used to monitor relative humidity
- if dew point mirror is included in the set up – no need for high quality humidity sensor for regulation
- if no dew point mirror is available – compare humidity sensors against each other
- never trust calibrations from company for capacitive sensors
- salt measurements are useful to check the right RH in terms of sensor shifts and temperature fluctuations – gives RH inside the DMA
- calibration with salts is temperature dependent
- calibration by DRH is useful to do in the lab – maybe it is not useful to do it in the field (question of time)
- for calibration by DRH – use at least 3 different salts
- importance of a dew point mirror when using salt calibration

**As a solution we note/propose:**

- dew point mirror should be placed in the excess air flow
- use fast sensors for humidity regulation
- calibrate humidity sensors regularly against the dew pointer
- if no dew point mirror is available, compare humidity sensors against each other

- in the field and in the lab: regularly scans with  $(\text{NH}_4)_2\text{SO}_2$  for calibration is recommended (use same salt for calibration in lab and in field)

**Main points - in terms of DMA sizing we note/need:**

- quality control for all DMA measurements
- calibration of high voltage supply
- calibration of a possible size shift due to DMA geometry and other issues

**As a solution we note/propose:**

- calibration with PSL spheres in the lab
- in the field: dry  $(\text{NH}_4)_2\text{SO}_2$  measurements with shape factor 1
- for hardware characterization do not use ambient aerosol
- for hardware characterization do not use agglomerates
- compare systems, that measure the same particles at the same time

**Differences in data evaluation**

The discussions showed, that all groups used substantially different corrections in terms of the data evaluation of their H-TDMA data.

	<b>Data evaluation</b>	<b>Differences in data evaluation</b>
1	Sizing correction	All groups corrected their data for size shifts of the used DMAs.
2	Losses correction	Some groups corrected their data for losses within then system.
3	Parameterization	All groups fitted log normal functions to their hygroscopic growth distributions.
4	gf correction	Some groups usually correct their data for deviations of RH from the set value.

**Main points - in terms of data evaluation**

- for urban aerosol: fitting of 3 modes is useful ( $2^{\text{nd}}$  mode is in most cases very small)
- borders for nearly hydrophobic ,less hygroscopic and more hygroscopic particles change slightly from data set to data set
- soluble volume fraction is an additional issue
- principle of “soluble volume fraction” works out in practice
- heated inlets can change hygroscopic behavior (different GF) and stands against the principle of “soluble volume fraction”
- for GF classes: soluble volume fractions over the whole size distributions can be calculated (soluble volume classes)
- small fluctuations / differences in RH from the set point can be recalculated to the set point of RH

**As a solution we note/propose:**

- instead of GF you can also define certain growth factor classes
- GF of certain modes and GF classes are two possible ways to analyze the data

## Differences in data inversion schemes / recalculation algorithms

The discussions showed, that all groups used different data inversion schemes / recalculation algorithms when processing their H-TDMA data.

	Algorithm	Differences in algorithms
1	Multiply charged particles	No routine did take into account multiply charged particles.
2	Singly charged particles	Some groups use an algorithm taking into account singly charged particles calculating the humidified log normal particle number size distribution upstream of DMA 2. Others used an inversion routine according to McMurry and Stolzenburg. Some did not use any algorithm.
3	Transfer function	Some groups took into account the ideal transfer function of DMA 2 in their algorithm. Some groups took into account the real transfer function of DMA 2 in their algorithm.
4	CPC efficiency	Some groups took into account the CPC efficiency in their algorithm.

### Main points - in terms of data inversion schemes/recalculation algorithms we note/need:

- different ways for inversion/recalculation are done
- here, inversion means correction concerning transfer function of the 2<sup>nd</sup> DMA
- final output from the inversion is the real size distribution upstream of 2<sup>nd</sup> DMA
- transfer function of the 2<sup>nd</sup> DMA influences measured size distribution
- transfer function of Vienna type DMA is broader than the ideal transfer function compared to the TSI type DMA
- no correction of multiply charged particles is possible
- does everybody recalculate  $dN/d\log D_p$ ?

### As a solution we note/propose:

- inversion is needed for the true values of the number fraction

### How can a future system look like?

- temperature stabilized system (humidity regulation, 2<sup>nd</sup> DMA) leads to well defined RH during measurements
- careful temperature and RH monitoring is needed
- RH in /out of 2<sup>nd</sup> DMA should agree very well (temperature and RH have to be measured)
- drifts of the sensors should be controlled
- temperature sensors should be placed in the housing of the 2<sup>nd</sup> DMA
- optimized system for humidification control
- separate aerosol and sheath air humidification for hysteresis measurement
- self calibrating (regularly automated measurements with  $(\text{NH}_4)_2\text{SO}_2$ ) system
- closed loop systems: operating 1<sup>st</sup> DMA as a closed loop is no problem
- closed loop systems: operating 2<sup>nd</sup> DMA as a closed loop leads to slow RH change
- better time resolution: scanning instead of stepping systems (counting statistics should be better because of efficient timing)

- scanning system can be used as a stepping system by scanning slowly (only problem: delay time in CPC leads to smearing)
- scanning system especially needed for measurements in air crafts
- for scanning and stepping systems the same DMA type can be used
- OMAC instead of DMA: is not yet fully developed and comparable to DMA (size range is smaller)
- remote control and surveillance (internet-based software) is suggested
- recovery from power failures is suggested
- automated fitting for online data evaluation is suggested
- automated fitting needs to be controlled
- it is useful to agree on one format for the data
- common platform for data processing and standard procedure for data processing is needed

### **Evaluation of the workshop data**

- data from workshop will be presented with GF (growth factors) for modes (no GF classes) and NF (number fractions) for modes
- data will be processed with individual inversion algorithms
- IfT will do data processing using the same procedure for all data (procedure: IfT-routine)
- raw data are needed in a certain format for all systems
- IfT will provide a spread sheet for the needed format

### **Publication**

- protocol for quality control / quality assurance and data evaluation is important for H-TDMA community
- comparison of the different systems & protocol will be published in one paper (shows comparison of the systems and comes up with the protocol and what the results mean to the future work in the conclusion)
- writing of the manuscript will be done by Nicole Kaaden, Andreas Massling, Kaarle Hämeri, Erik Swietlicki, Alfred Wiedensohler, ...

### **Time schedule**

- beginning of April 2006 students meet in Leipzig for data comparison (evaluation of the data processed with the individual procedures and the IfT routine)
- final report to ACCENT by the end of February 2007

# **Results: H-TDMA workshop in February 2006 at the Leibniz-Institute for Tropospheric Research in Leipzig, Germany (Part I)**

## **1 SYSTEMS**

### **1.1 Helsinki – Finland**

#### **1.1.1 Set up**

#### **1.1.2 Quality criteria**

#### **1.1.3 Inversion scheme / recalculation algorithm**

#### **1.1.4 Data evaluation**

### **1.2 Lund – Sweden**

#### **1.2.1 Set up**

#### **1.2.2 Quality criteria**

#### **1.2.3 Inversion scheme / recalculation algorithm**

#### **1.2.4 Data evaluation**

### **1.3 Manchester – England**

#### **1.3.1 Set up**

#### **1.3.2 Quality criteria**

#### **1.3.3 Inversion scheme / recalculation algorithm**

#### **1.3.4 Data evaluation**

### **1.4 Clermont - Ferrand – France**

#### **1.4.1 Set up**

#### **1.4.2 Quality criteria**

#### **1.4.3 Inversion scheme / recalculation algorithm**

#### **1.4.4 Data evaluation**

### **1.5 Leipzig – Germany**

#### **1.5.1 Set up**

#### **1.5.2 Quality criteria**

#### **1.5.3 Inversion scheme / recalculation algorithm**

#### **1.5.4 Data evaluation**

## **1.6 Leipzig – Germany**

### **1.6.1 Set up**

### **1.6.2 Quality criteria**

### **1.6.3 Inversion scheme / recalculation algorithm**

### **1.5.4 Data evaluation**

## **2 EXPERIMENTAL SET UP**

## **3 EXPERIMENTAL METHODS AND RESULTS**

### **3.1 Dry Ammonium Sulfate**

#### **3.1.1 Problems**

#### **3.1.2 Experiment**

#### **3.1.3 Data Evaluation**

#### **3.1.4 Results**

#### **3.1.5 Advices**

### **3.2 90 % Ammonium Sulfate**

#### **3.2.1 Problems**

#### **3.2.2 Experiment**

#### **3.2.3 Data Evaluation**

#### **3.2.4 Results**

#### **3.2.5 Advices**

### **3.3 DRH Ammonium Sulfate**

#### **3.3.1 Problems**

#### **3.3.2 Experiment**

#### **3.3.3 Data Evaluation**

#### **3.3.4 Results**

#### **3.3.5 Advices**

### **3.4 90 % soot + Ammonium Sulfate**

#### **3.4.1 Problems**

#### **3.4.2 Experiment**

#### **3.4.3 Data Evaluation**

#### **3.4.4 Results**

### 3.4.5 Advices

## A APPENDIX: Tables

## B APPENDIX: Basic considerations

### B.1 Influence of the inversion scheme / recalculation algorithm on the growth factor

### B.2 The instability of temperatures within the H-TDMA

## 1 SYSTEMS

This paragraph will describe the individual systems in more detail. It is subdivided into small sections about the set up, the chosen quality criteria, inversion scheme / recalculation algorithm and the data evaluation procedure of each system.

### 1.1 Helsinki – Finland

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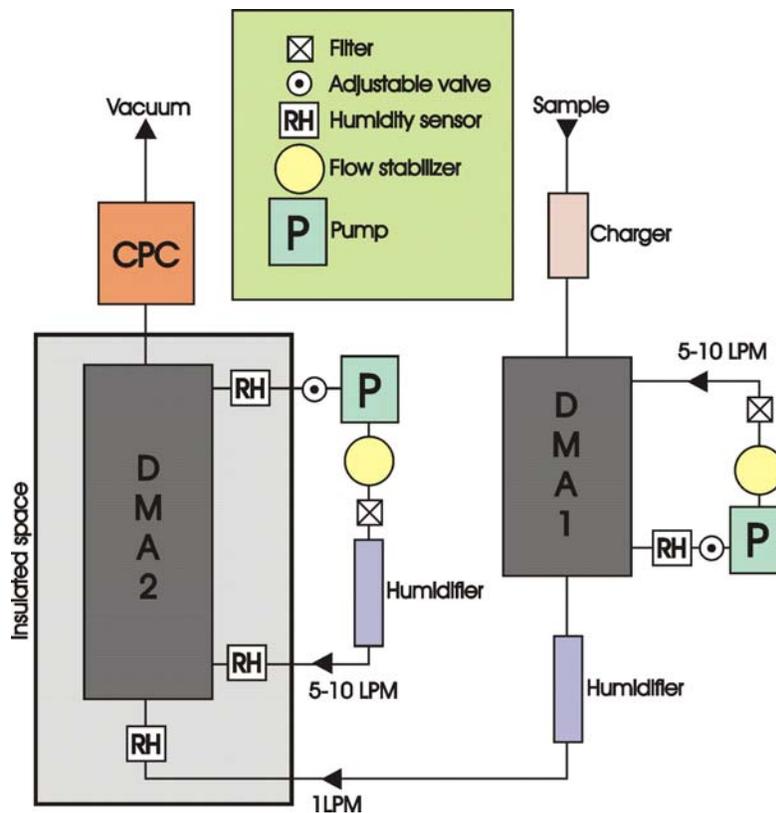


Figure 1.1: Scheme of the H-TDMA from Helsinki, System 1.

### **1.1.1 Set up**

1. No CPC downstream of DMA 1 is used; thus, no internal closure (CPC1 / CPC2) can be calculated.
2. A dew point mirror is used.
3. This is a closed-loop set up.

### **1.1.2 Quality criteria**

1. From the dew point mirror the mean RH is calculated.
2. Data with variation in RH larger than a certain value ( $\pm 1\%$ ) are sorted out.
3. Ammonium sulfate calibration is done once a day (there are plans to automate and do this more often in the future), but they are not used right now.
4. If the integrated total number concentration of CPC 2 is lower than a certain value, the scans are sorted out.
5. Losses are not taken into account for the data evaluation (relative measurements are taken, size-dependent losses may be important, but distance DMA 1 to DMA 2 is short, so there is no big difference in losses between 10 and 20nm).

### **1.1.3 Inversion scheme / recalculation algorithm**

1. A DMPS inversion scheme / recalculation algorithm is used calculating the humidified lognormal size distribution upstream of DMA 2.
2. No multiply charged particles are considered.
3. An ideal transfer function for DMA 2 is taken into account.

### **1.1.4 Data evaluation**

1. Data are parameterized by fitting of Gaussian functions (new start values have to be set for every new scan if the distribution changes).
2. The correction of growth factors calculated at the set RH is done with a two component model.
3. At the beginning of a measurement campaign, dry scans are taken and the data are corrected for shifts afterwards.

## **1.2 Lund – Schweden**

Responsible Scientists:

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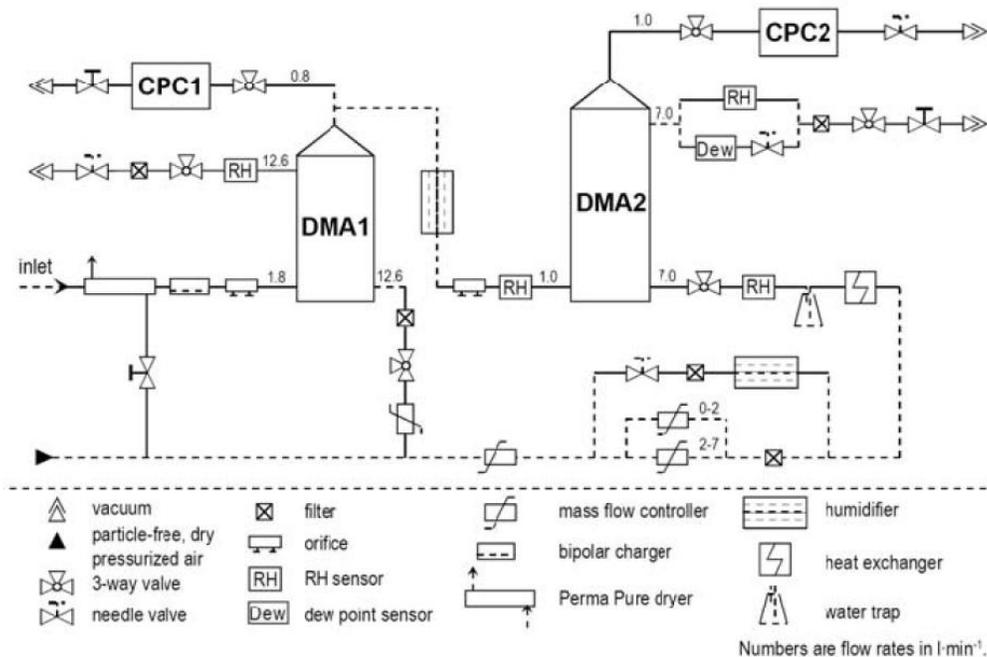


Figure 1.2: Scheme of the H-TDMA from Lund, System 2.

### 1.2.1 Set up

1. A CPC is used downstream of DMA 1; thus, an internal closure (CPC1 / CPC 2) can be calculated.
2. A dew point mirror is used.
3. This is a non closed-loop set up.

### 1.2.2 Quality criteria

1. Relative humidities, flows, pressure drops, and temperatures are monitored and checked for stability.
2. Ammonium sulfate salt scans are taken several times over the day to check the RH stability.
3. The number concentration over DMA 1 is checked for stability.

### 1.2.3 Inversion scheme / recalculation algorithm

1. An inversion scheme / recalculation algorithm according to McMurry and Stolzenburg is used.
2. It is assumed, that the Cunningham correction (which is used here) of the peak of the transfer function is the same over the width of this function.
3. The inlet concentration of DMA 2 is assumed to be triangular (a theoretical function is used).
4. No multiply charged particles are considered.

### 1.2.4 Data evaluation

1. Data are parameterized by fitting of log normal functions (program can be used to set outlier points of a mode equal to zero, or ignore them altogether).
2. The loss functions of DMA1 and DMA2 are considered.

- The temperatures evaluated from the ammonium sulfate scans are plotted with the best T sensors in the system; for the RH assignment, the RH from the salt scans and from the dew point mirror are combined; thus, the real temperature in the DMA2 can so far be calculated for every time and the real RH in DMA2 can thus be calculated from the dew point measurement and the temperature of DMA2 for every time.
- The correction of growth factors calculated at the set RH is done with a two component model.

### 1.3 Manchester – England

Responsible Scientists:

(Gordon McFiggins)

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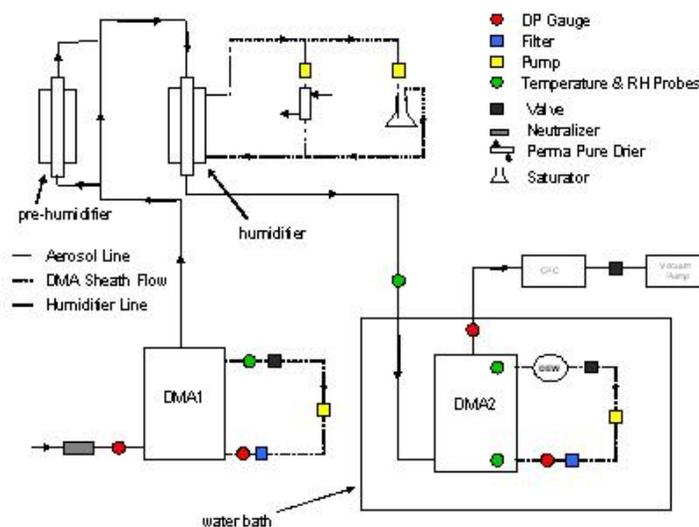


Figure 1.3: Scheme of the H-TDMA from Manchester, System 3.

#### 1.3.1 Set up

- A dew point mirror is used.
- This is a closed loop set-up.

#### 1.3.2 Quality criteria

- The flows, temperatures, and count statistics for one day of measurements in terms of time series are checked.

#### 1.3.3 Inversion scheme / recalculation algorithm

- The DMA 2 transfer function is calculated out of the dry scans for every new experiment with a new substance.

#### 1.3.4 Data evaluation

- The shift of DMA1 is corrected based on a calibration of DMA 1 with PSL spheres, which has been done once.

2. Calibration of RH: T sensor in the bottom of DMA 2 is compared with the dew point measurement and salt scans in between; the RH assignment is similar to Lund (see system 2).
3. A fit program is used, where the data points are connected and it can be decided afterwards, which function is used (Gaussian or log normal function); the fitting program does a fit back through to the original distribution from the retrieval to see, if the fit has worked correctly.

## 1.4 Clermont - Ferrand – France

Responsible Scientists:

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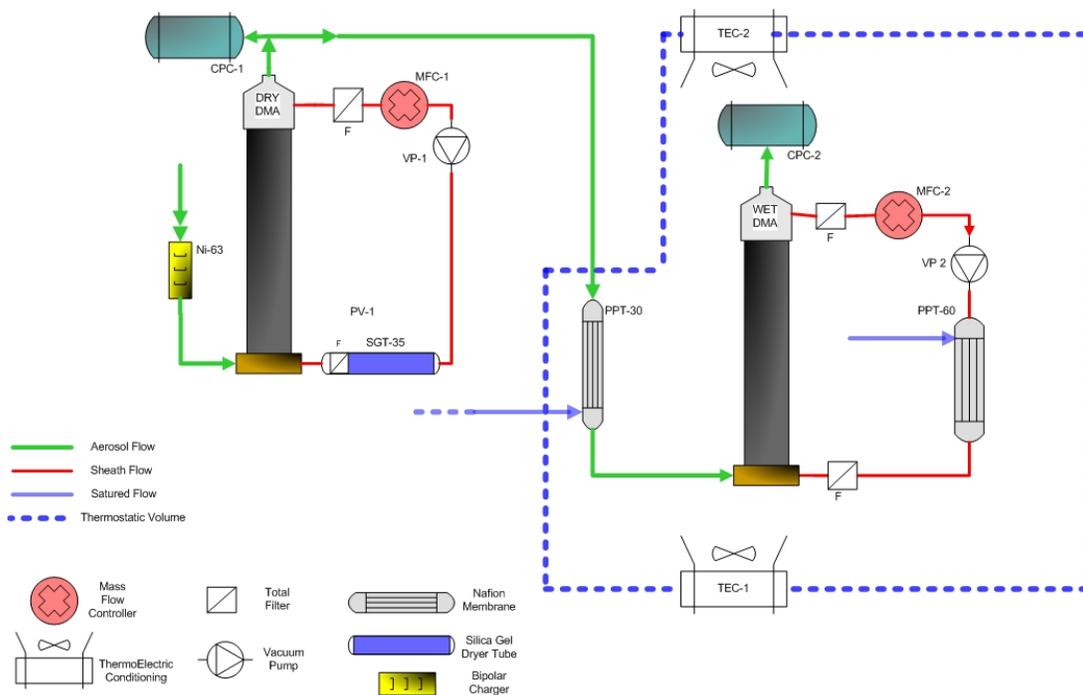


Figure 1.4: Scheme of the H-TDMA from Clermont Ferrand, System 4.

### 1.4.1 Set up

1. A CPC is used downstream of DMA1 (CPC2 is calibrated with respect to CPC1).
2. No dew point mirror is used.
3. This is a closed loop set up.

### 1.4.2 Quality criteria

1. No quality check is used for the determination of the stability of RH or T.
2. A quality check is done for the sheath/excess flows in both DMA loops and for the aerosol flows entering each DMA by measurement of pressure drops.

### 1.4.3 Inversion scheme / recalculation algorithm

1. Until now, no inversion scheme / recalculation algorithm is used.

#### 1.4.4 Data evaluation

1. Dry scans are taken before each experiment.
2. A shift of DMA2 is corrected by the information of dry scans only, if the deviations are larger than a certain value ( $\pm 1\%$ ).
3. The concentration of CPC2 is normalized by the concentration of CPC1.
4. A calibration of the RH sensors is done in the lab with saturated salt solutions (3 points:  $K_2SO_4$ ,  $NaCl$  and  $MgCl_2$ ).
5. No ammonium sulfate calibration is done during field measurements (there is a plan to add and automate this calibration 2 - 3 times per day).
6. The RH given in all data corresponds with the mean RH measured during each scan by the DMA2 excess flow sensor and the aerosol sample flow sensor, respectively.
7. A spline fit algorithm is used calculating the most 3 maxima in the observed spectra.

#### 1.5 Leipzig– Germany

Responsible Scientists:

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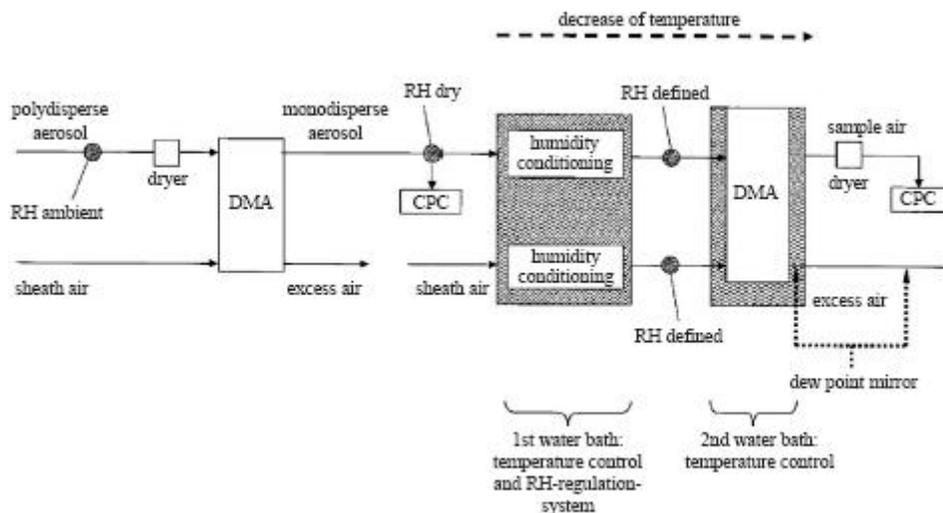


Figure 1.5: Scheme of the HH-TDMA from Leipzig, System 5.

##### 1.5.1 Set up

1. A CPC is used downstream of DMA1; thus, an internal closure (CPC1 / CPC2) can be calculated.
2. Two water baths are used for the temperature stabilization of the RH control unit and the second DMA, respectively.
3. A dew point mirror is used.
4. This is a non closed-loop set up.

### 1.5.2 Quality criteria

1. The stability of the inlet concentration of CPC1 and all relative humidities and temperatures is checked.
2. The difference in temperature of RH regulation sensors should be smaller than a certain value.
3. The aerosol RH should be within a certain value of the set RH value ( $\pm 2\%$ ).

### 1.5.3 Inversion scheme / recalculation algorithm

1. A DMPS inversion scheme / recalculation algorithm is used calculating the humidified lognormal size distribution upstream of DMA 2.

### 1.5.4 Data evaluation

1. The DMA shift for DMA 1 is corrected based on dry scans taken with ammonium sulfate before each experiment.
2. The data are corrected for the losses between CPC1 and based on a calibration curve.
3. The mean RH and the mean temperature is taken from the dew point mirror as main sensor.
4. The data are parameterized by fitting up to 3 log normal functions.

## 1.6 Leipzig– Germany

Responsible Scientists:

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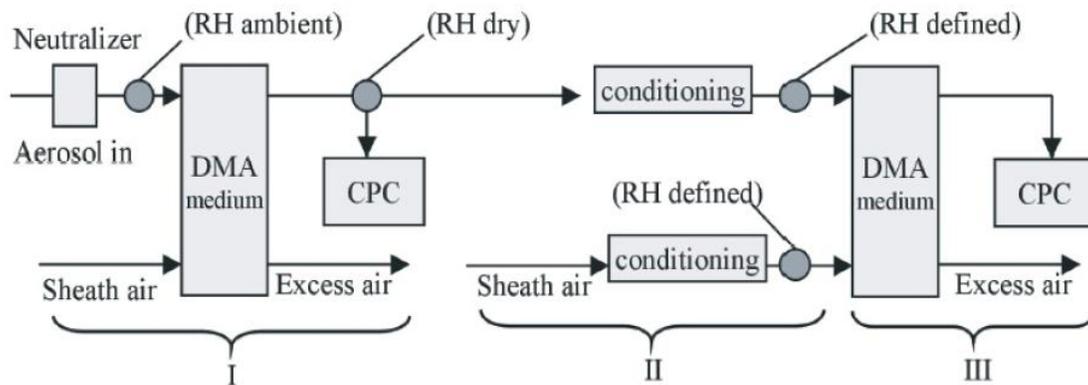


Figure 1.6: Scheme of the H-TDMA from Leipzig, System 6.

### 1.6.1 Set up

1. A CPC is used downstream of DMA1; thus, an internal closure (CPC1 / CPC2) can be calculated.
2. No dew point mirror is used.
3. This is a non closed-loop set up.

### 1.6.2 Quality criteria

1. The stability of RH, temperatures, inlet concentrations, and flows are checked.

### 1.6.3 Inversion scheme / recalculation algorithm

1. A DMPS inversion scheme / recalculation algorithm is used calculating the humidified log normal size distribution upstream of DMA 2.

### 1.5.4 Data evaluation

1. The DMA shift for DMA 1 is corrected based on dry scans taken with ammonium sulfate before each experiment.
2. There is no correction for the CPC efficiency used.
3. The data are corrected for losses between CPC1 and CPC2 in the interesting size range based on a calibration curve.
4. Salt scans of ammonium sulfate are taken every 1 - 2 hours during each experiment; the RH in DMA2 is calculated from theoretical growth curves; the interpolation between the RH points is done and each scan is assigned to the real RH in DMA2 during the measurement.
5. The concentration of CPC2 is normalized based on the CPC 1 inlet concentration.
6. The correction of growth factors calculated at the set RH is done with a two component model.
7. The data are parameterized by fitting up to 3 log normal functions.

## 2 EXPERIMENTS

A mixing chamber was used for all experiments except for experiment 6, when the ambient air was investigated. For experiment 6, the pressure drop in the chamber was too high resulting in instabilities in flows of some systems. Here, the ambient air was taken from the outside of the laboratories, room number 118 and room number 119. A scheme of both rooms where the systems were situated is shown in Figure 2.1. All pipes used for conducting the aerosol were 3/8" in size. One pipe was used for connecting the two rooms to apply the generated aerosol to all systems. Both rooms were air conditioned with a set value in temperature of about 20°C.

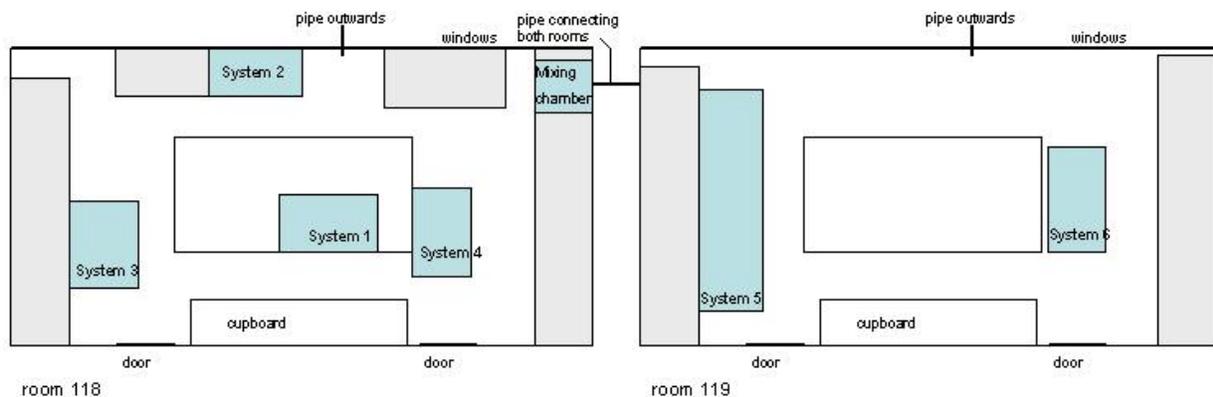


Figure 2.1: Scheme of both laboratory rooms 118 and 119.

In Table 2.1 all experiments performed during the workshop are listed (day of measurement and timeframe used for evaluation based on the number of participating systems that had good quality criteria).

	<b>Experiment</b>	<b>Date</b>	<b>Timeframe measurement</b>	<b>Timeframe evaluation</b>
1	dry Ammonium Sulfate	Tuesday, 07.02.06	05 PM - 10 PM	06 PM - 09 PM
2	90% Ammonium Sulfate	Wednesday, 08.02.06	11 PM - 12 PM	02 AM - 10 AM
3	DRH Ammonium Sulfate	Wednesday, 08.02.06	01 PM - 09 PM	01 PM - 09 PM
4	90% soot + Ammonium Sulfate	Thursday, 09.02.06	11 PM - 02 PM	12 AM - 08 AM
5	75 and 85% soot + Ammonium Sulfate (not evaluated yet)	Thursday, 09.02.06	02 PM - 04 PM	02 PM - 04 PM
6	90% ambient (not evaluated yet)	Friday, 10.02.06	08 PM - 10 AM	02 AM - 10 AM

Table 2.1: Introduction of experiments with belonging timeframes of measurement and evaluation periods.

The following table gives an overview, from which systems data could be provided for the evaluation of the different experiments during the chosen timeframes.

	System 1	System 2	System 3	System 4	System 5	System 6
Experiment 1	x	x	x	x	x	x
Experiment 2	x	x			x	x
Experiment 3	x	x		x	x	x
Experiment 4	x	x			x	x
Experiment 5	x	x		x	x	x
Experiment 6	x	x		x	x	x

Table 2.2: Scheme, from which systems data could be provided for the different experiments during the chosen timeframes.

### 3 EVALUATION and RESULTS

The data have been processed in two different ways. In a first step, data have been processed using the routines that are originally used for each individual system (external evaluation). In a second step, all raw data were processed the same way using the IFT-internal data evaluation procedure (internal evaluation) for inversion scheme / recalculation algorithm and data fitting. As the same routine (internal and external) is used for System 5 and 6 from IFT, those data were just calculated once. All results shown in the following sections are mean values of the respective experiment for the individual system operated during the timeframe for evaluation. Some systems only were operated for time windows during the timeframes listed above.

#### 3.1 Dry Ammonium Sulfate

##### 3.1.1 Problem

The sizing of DMAs may differ from the set values because of several reasons. For TDMA's this problem occurs in both DMAs and results in a size-dependent internal growth factor for the individual TDMA. DMA shifts may occur because of the reasons listed in Table 3.1.

Uncertainties in sizing of a DMA	Low impact	Large impact
Sheath flow stability	X	
Ambient pressure in the DMA	X	
Ambient temperature in the DMA	X	
Penetration losses in the inlet and in the DMA	X	
DMA transfer function	X	
Relative humidity in the DMA		X
Voltage calibration		X

Table 3.1: List of properties, that may have an impact on particle sizing using a DMA.

##### 3.1.2 Experiment

Ammonium sulfate particles were generated based on a 0.1 M solution using a TSI atomizer (Model 3076). The particle flow of about 2 lpm was diluted by dry and oil-free synthetic air of about 2 lpm and conducted to the mixing chamber. Each individual system had a drying section before the particles entered the first DMA. The systems were operated at dry conditions yielding the individual internal growth factor for selected sizes, here  $D_p = 30, 50, 70$  and  $100$  nm (the finish system using  $10$  instead of  $100$  nm).

##### 3.1.3 Data evaluation

The individual internal growth factors  $gf_{int}$  were derived by the calculation of the ratio of the derived diameter of DMA2  $D_{p,DMA2}$  and the set diameter of DMA1  $D_{p,set}$ , which is reciprocal to the uncorrected GF  $GF_{uncor}$ .

$$gf_{int} = D_{p,set} / D_{p,DMA2} = 1 / GF_{uncor} \quad (\text{Equation 3.1})$$

For a better comparison, the following calculations were made out of the mean of the measured hygroscopic growth factors during this experiment.

Absolute difference in GF:  $dGF = 1.00 - GF_{uncor.}$  (Equation 3.2)

### 3.1.4 Results

In Figures 3.1 and 3.2, the results of this experiment are shown presenting the evaluation of both routines (internal and external evaluation).

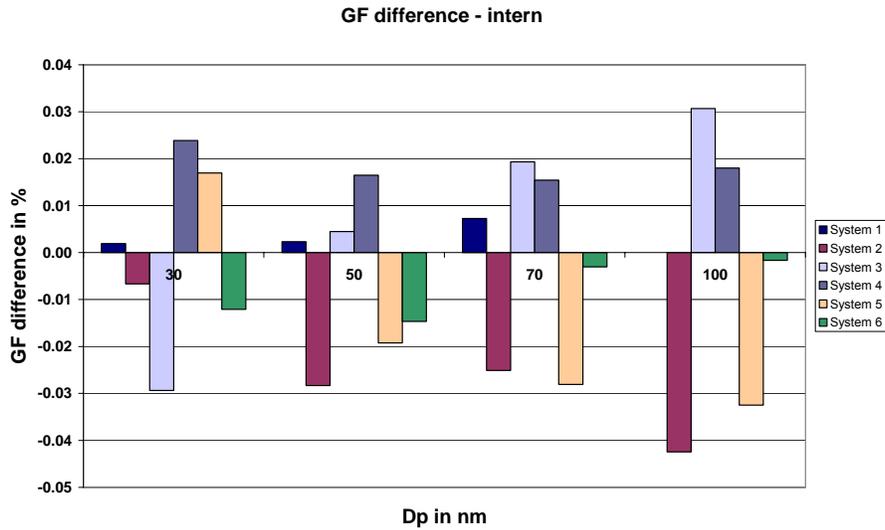


Figure 3.1: Internally evaluated growth factor difference for all systems.

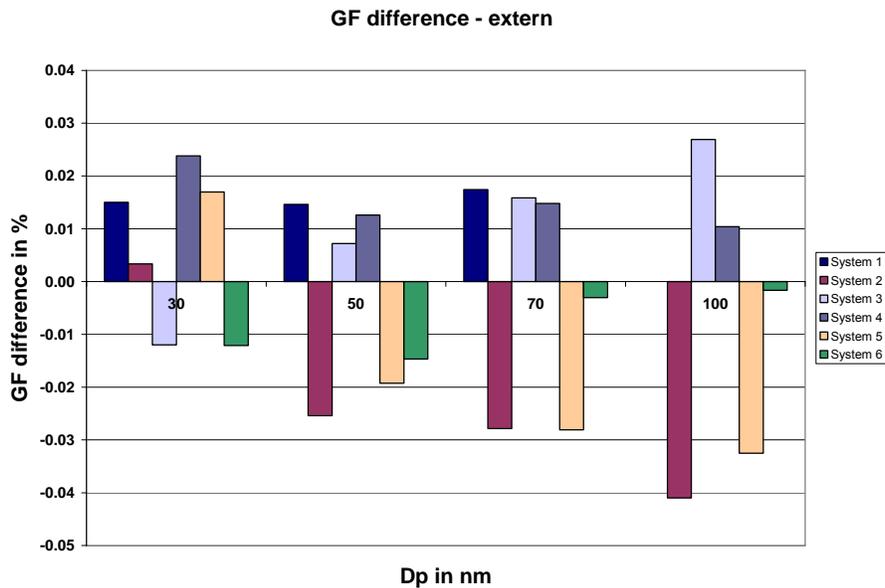


Figure 3.2: Externally evaluated growth factor difference for all systems.

- I. In general, shifts in sizing between the two DMAs of individual systems are smaller than 4% in relative deviation.

- II. The fourth column in Table A.1 and A.2 in the Appendix shows the values by fitting a regression curve to the measured values for all sizes and individual systems. Obviously, the shift in sizing behaves linear in dependence in size resulting in correlation coefficients around 1.
- III. Also, the external and internal evaluations do only differ within 2% of relative deviation.

### 3.1.5 Advices

- I. Because shifts in sizing between the two DMAs of individual systems are about 1 to 4 % in relative deviation, we propose to correct the growth factors for the individual shifts based on dry ammonium sulfate scans taken during each field campaign from time to time. We propose to take dry scans at the same diameters as the TDMA is operated during measurement. Thus, an internal growth factor can be calculated for each dry size and be applied to all data measured at that size.
- II. There is a second way correcting the growth factors for shifts in sizing by calculating a regression curve  $y = ax + b$  (y set diameter DMA 1, x measured diameter DMA 2). Here, the wet diameter can be corrected.  $GF = y / Dp_{set}$
- III. High voltage power supplies should be calibrated once the year especially for small voltages. The calibration of the power supplies should be done after a warm up period of at least one hour.
- IV. The DMA sizing of individual DMAs should be controlled by determination of sizes of PSL latex spheres in the operating size range before a TDMA is set to field the first time.

## 3.2 90 % Ammonium Sulfate

### 3.2.1 Problem

For a well calibrated system, the largest uncertainty in hygroscopic growth factor results from RH instabilities in the second DMA operated at humidified conditions. As the RH in the system is controlled by an RH sensor, that is not a part of the DMA itself, 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. DMAs have massive bodies and only follow slowly the temperature curve of the lab.

### 3.2.2 Experiment

Ammonium sulfate particles were generated based on a 0.1 M solution using a TSI atomizer (Model 3076). The particle flow of about 2lpm was diluted by dry and oil-free synthetic air of about 2 lpm and conducted to the mixing chamber. Each individual system had a drying section before the particles entered the first DMA. The systems were operated at 90% RH selecting initial dry diameters of  $D_p = 30, 50, 70$  and 100 nm (the finish system using 10 instead of 100 nm) yielding the individual hygroscopic growth factors of ammonium sulfate at 90 % RH for the selected dry sizes.

### 3.2.3 Data Evaluation

For a better comparison, the following calculations were made out of the mean of the measured hygroscopic growth factors during this experiment.

Relative uncertainty in GF:  $relUn = GF_{meas}(RH_{read})/GF_{theo}(RH_{read}).$  (Equation 3.3)

Here,  $GF_{meas}$  is the measured size-corrected growth factor,  $RH_{read}$  is the RH given from the individual system (set RH was always 90 %), and  $GF_{theo}(RH_{read})$  is the theoretically calculated growth factor at  $RH_{read}$  based on data taken by Tang and Munkelwitz (1994).

Absolute difference in RH:  $dRH = RH_{theo} - RH_{read}$ . (Equation 3.4)

Here,  $RH_{theo}$  is the RH expected in the DMA based on the measured growth factor  $GF_{meas}$  based on the theory of Tang and Munkelwitz (1994).

### 3.2.4 Results

Figures 3.3 and 3.5 show the above mentioned parameters for internally evaluated data listed by individual systems. Figures 3.4 and 3.6 show the same results for externally evaluated data. The GFs are summarized in Tables A.3 and A.4 in the Appendix.

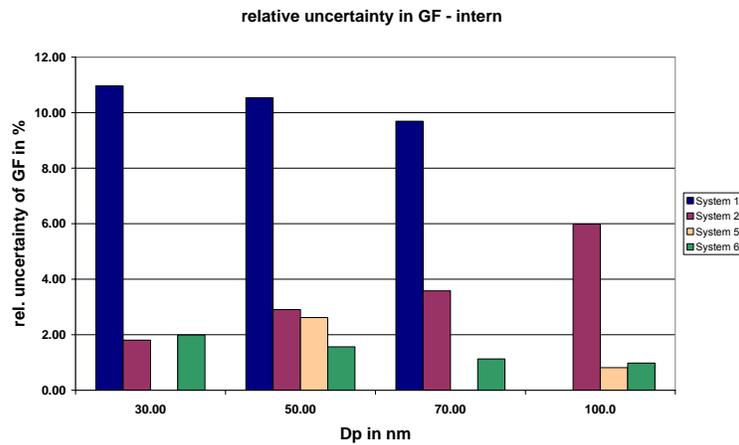


Figure 3.3: Internally evaluated relative uncertainty in growth factor for all measured diameters except 10nm for 4 of the 6 systems.

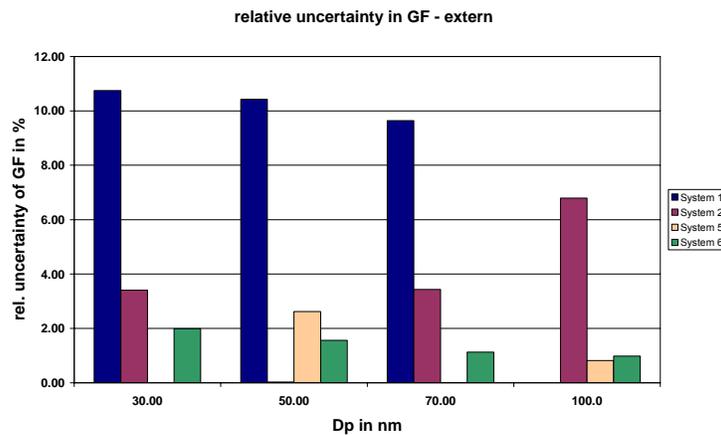


Figure 3.4: Externally evaluated relative uncertainty in growth factor for all measured diameters except 10nm for 4 of the 6 systems.

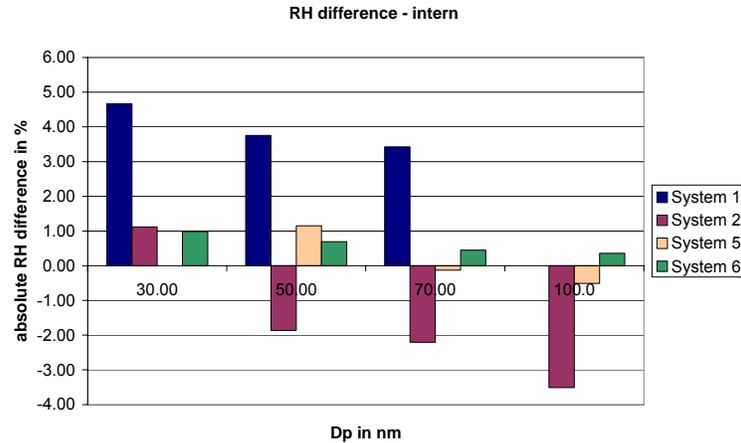


Figure 3.5: Internally evaluated RH difference for all measured diameters except 10nm for 4 of the 6 systems.

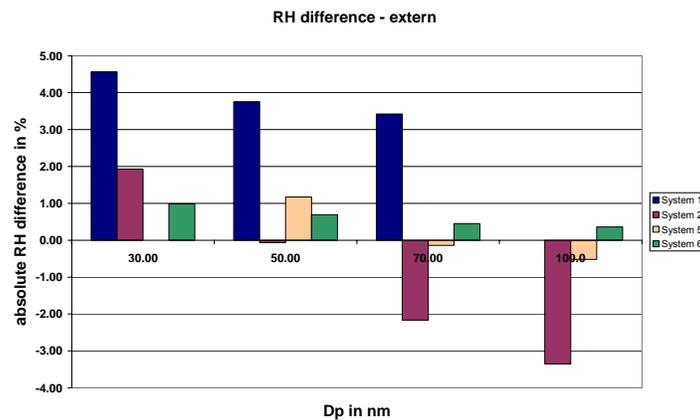


Figure 3.6: Externally evaluated RH difference for all measured diameters except 10nm for 4 of the 6 systems.

- I. In general, system 1 shows higher relative uncertainties in growth factor than other systems for both evaluations (high uncertainty within this experiment in relative humidity is known for system 1).
- II. Except for system 1, the relative uncertainties in growth factors are smaller than 7%.
- III. We see a size dependent error, which is decreasing for system 1, 5, and 6 with increasing investigated size and increasing for system 2 with increasing investigated size. This is observed for internal as well as for external evaluation.
- IV. The calculated relative difference in RH shows the same behavior as the relative uncertainty in growth factor as expected.
- V. Except for system 1, the absolute difference between measured RH and calculated RH based on measured growth factors is smaller than 3.5% RH for all systems independently of the type of evaluation.

### 3.2.5 Advices

- I. All relative humidity sensors should be calibrated by means of a dew point mirror or at least with a trustworth RH sensor using at least 4 to 6 RH values. The different

RH values can be produced with the sheath air set up of the individual system. We propose to do this evaluation once the year, at least before the beginning of a field campaign.

- II. To assure RH control during the field measurements, we propose one of the following techniques: First, you can use a dew point mirror in the excess air flow (the RH can not be regulated by means of a dew point mirror, but it can be determined later; you have to measure the temperature of the DMA as close as possible to the inner housing of the DMA in parallel. Second, you can automate the generation of salt particles of known theoretical growth and measure the particle growth in regular intervals (every 2 hours) during field measurements. This technique is actually the best method to obtain the real RH inside the DMA as the dew point method depends on the measurement of the real temperature inside the DMA. Third, a combination of method 1 and method 2 is the most appropriate technique. All methods provide a good knowledge about the real RH inside the DMA during measurement.

### **3.3 DRH Ammonium Sulfate**

#### **3.3.1 Problems**

Here, the operation of the TDMA was tested at one point in RH. The jump in hygroscopic growth should be observed at about 80% RH for ammonium sulfate particles. Based on RH regulation uncertainties and temperature differences between RH control sensor and DMA housing, the jump was observed at different RH values giving the hint, that the individual systems were not operating correctly at this point of RH.

In the transition zone of deliquescence relative humidity (DRH), sometimes more than one particle mode was observed, which could be due to a non-dissolved core coated by a water layer, or a contamination of the solution, or multiply charged particles, that behaved different because of the Kelvin effect.

#### **3.3.2 Experiment**

Ammonium sulfate particles were generated based on a 0.1 M solution using a TSI atomizer (Model 3076). The particle flow of about 2 lpm was diluted by dry and oil-free synthetic air of about 2 lpm and conducted to the mixing chamber. Each individual system had a drying section before the particles entered the first DMA. The systems were operated at  $D_p = 70\text{nm}$  increasing the RH individually from 75 % to 85 % yielding the DRH of the individual systems for the selected size.

#### **3.3.3 Data Evaluation**

The following procedure was chosen for the internal data evaluation to obtain the DRH point from the measured H-TDMA growth spectra. The distributions were processed in a conventional way and only one particle mode was fitted (only spectra showing one particle mode were considered). The following four parameter exponential equation was fitted to the observed growth curve in dependence on RH.

$$GF = a - [b / (1 + \exp[(RH - RH1) / RH2])]. \quad (\text{Equation 3.5})$$

The point, when the DRH occurred was defined to be the 50% growth factor  $GF_{50}$  of the growth factor reached after deliquescence.

$$GF_{50} = (GF - GF_{\min}) / 2. \quad (\text{Equation 3.6})$$

Thus, the DRH was calculated by the following formula:

$$RH_{50} = RH_2 * \ln \{ [b / (a / 2)] - 1 \} + RH_1 \quad (\text{Equation 3.7})$$

To determine the parameters a, b,  $RH_1$  and  $RH_2$ , some values for these parameters were assumed. 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 parameters for all systems are to be found in Table A.5 in the Appendix. For comparison of all systems, the following calculation of  $DRH_{\text{diff}}$  was provided:

$$DRH_{\text{diff}} = DRH_{\text{theo}} - DRH_{50}. \quad (\text{Equation 3.8})$$

Here,  $DRH_{\text{theo}}$  was about 79.97 % RH. For the external evaluation the DRH was obtained by different individual routines.

### 3.3.4 Results

Figures 3.7 and 3.8 show the difference in DRH measured for the individual systems based on both calculations for internal and external evaluation.

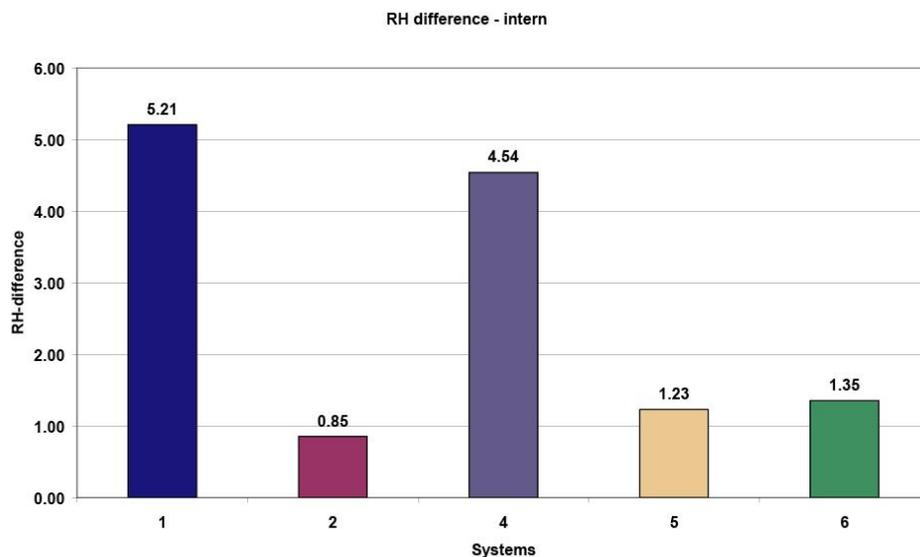


Figure 3.7: Internally evaluated DRH difference for 5 of the 6 systems.

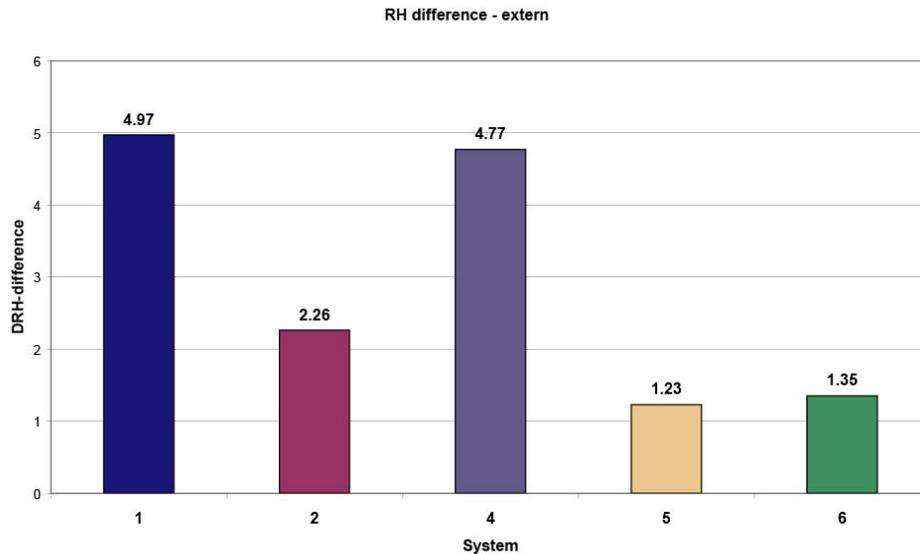


Figure 3.8: Externally evaluated DRH difference for 5 of the 6 systems.

- I. The maximum in difference of DRH was about 5 to 5.5% in RH independently of type of evaluation.
- II. For all systems, the measured DRH was lower than the theoretical value for both types of evaluation.
- III. Two out of four systems were close together and determined the theoretical DRH within less than 1.5 % RH.
- IV. Deviations in measured DRH between the two types of evaluation were small.
- V. The high deviation in DRH determined for system 1 can be explained by the problems observed during experiment 2.

### 3.3.5 Advices

- I. We recommend to perform DRH measurements at least once before the H-TDMA is operated within a field campaign, once during the field campaign and once after the field campaign. The DRH measurement gives a clear hint, if the system is operating correctly at the DRH value of the investigated salt.
- II. The DRH measurements can be extended to individual salt solutions containing sodium chloride, ammonium sulfate, and potassium chloride to cover the RH range from 74 up to 86%. In combination to advices recommended in chapter 3.2.5, these procedures will assure the quality of your measurements in terms of RH instabilities, temperature instabilities and wrong calibrations of RH sensors.

## 3.4 Soot and Ammonium Sulfate

### 3.4.1 Problems

The number fraction of individual hygroscopic growth groups is the second most interesting parameter provided by H-TDMA measurements. Thus, an experiment was performed to intercompare this parameter between the H-TDMA systems.

### 3.4.2 Experiment

Ammonium sulfate particles were generated based on a 0.1 M solution using a TSI atomizer (Model 3076). In parallel, soot particles (Type Printex 18) suspended in water were generated using a second atomizer of the same model. The particle flows of about 2 lpm of each atomizer were combined and were diluted by dry and oil-free synthetic air of about 4 lpm and conducted into the mixing chamber. The experiment was provided to generate an externally mixed aerosol showing two particle groups with different hygroscopic growth. The systems were operated at initial dry sizes of  $D_p = 10, 30, 50, 70$  and  $100$  nm under humidified conditions at  $RH = 90\%$ .

### 3.4.3 Data Evaluation

The number fractions of observed hygroscopic growth modes were calculated dividing the individual number of particles of the group by the number of all particles observed in the total humidified particle number size distribution. The number of particles of each hygroscopic growth mode are a result of the log normal fit of the  $dN / d\log D_p$  distribution after applying an inversion scheme / recalculation algorithm.

$$nf_i = N_i / \text{sum}(N_i). \quad (\text{Equation 3.9})$$

### 3.4.4 Results

In Figures 3.9 and 3.10, the calculated number fractions of all systems for different diameters are presented for internally and externally evaluated data. The data are summarized in Tables A.7 and A.8 in the Appendix.

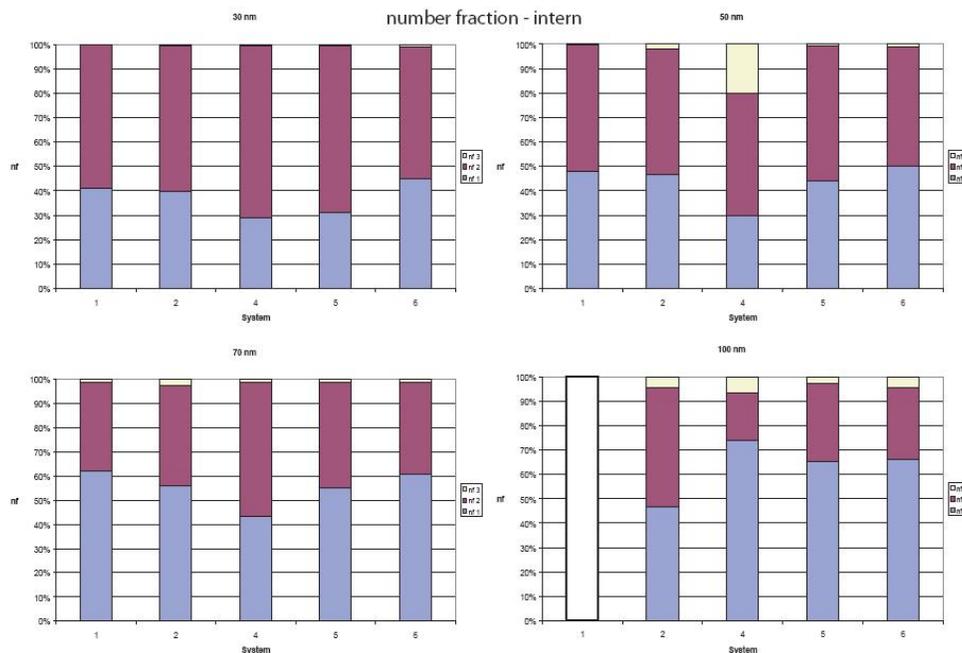


Figure 3.9: Internally evaluated number fraction of all measured diameters for 5 of the 6 systems.

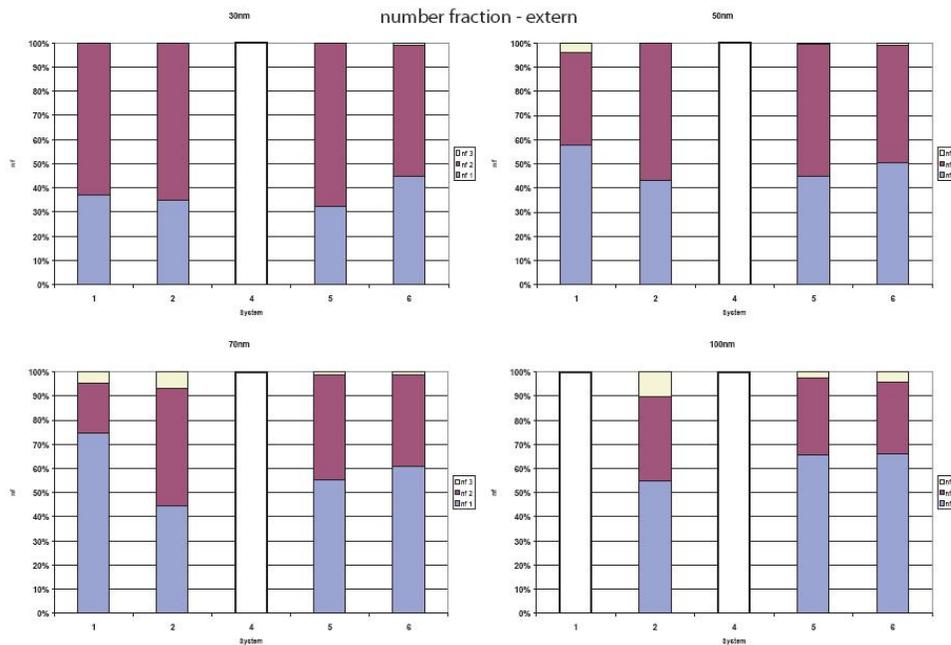


Figure 3.10: Externally evaluated number fraction of all measured diameters for 4 of the 6 systems.

- I. In general, for most systems, an external mixture yielding two dominant different hygroscopic growth groups was observed.
- II. Because of unknown reasons, sometimes a third mixed particle group was observed during some time periods. A pure soot mode showing only hydrophobic particles was never seen, maybe because of coagulation processes within the set up.
- III. All systems showed an increase in hydrophobic particle number fraction with ascending size for both types of evaluation.
- IV. The results show differences in calculated number fractions of all individual systems. These differences are within +/- 15% in calculated number fraction in between the individual systems for all internally evaluated and all externally evaluated data and within +/- 15% for each individual system with regard to the internal and external evaluation.

### 3.4.5 Advices

- I. We propose to invert the data in terms of a calculation of the humidified particle number size distribution upstream of DMA 2. The determination of the particle number size distribution  $dN / d\log D_p$  is necessary to obtain the real number of particles within different size ranges of the distribution.
- II. For the inversion scheme / recalculation algorithm, we recommend to implement the real, measured transfer function of the used DMA2. Differences in individual transfer functions may cause a smearing of the obtained particle number size distribution resulting in uncertainties in determination of number fractions of the observed hygroscopic growth modes.

## A APPENDIX: Tables

The following tables show the evaluated data for all experiments and both evaluation routines.

INTERNAL EVALUATION					
System	Dp set	Dp meas	reg. curve	correlation coefficient R	GF without correction
System 1:	10	9.85	$y = 1.0057x - 0.0211$	1	0.99
	30	29.94			1.00
	50	49.88			1.00
	70	69.49			0.99
System 2:	30	30.20	$y = 0.9474x + 1.4846$	0.9999	1.01
	50	51.42			1.03
	70	71.76			1.03
	100	104.24			1.04
System 4:	30	29.28	$y = 1.0598x + 0.127$	1	0.98
	50	49.18			0.98
	70	68.92			0.98
	100	98.20			0.98
System 5:	30	29.49	$y = 0.9499x + 1.788$	1	0.98
	50	50.96			1.02
	70	71.97			1.03
	100	103.25			1.03
System 6:	30	30.36	$y = 1.0049x - 0.677$	0.9999	1.01
	50	50.73			1.01
	70	70.21			1.00
	100	100.17			1.00

Table A.1: Comparison of DMA shift parameters for all systems with internal routine.

EXTERNAL EVALUATION					
System	Dp set	Dp meas	reg. curve	correlation coefficient R	GF without correction
System 1:	10	9.82	$y = 1.0173x - 0.0353$	1	0.98
	30	29.55			0.99
	50	49.27			0.99
	70	68.78			0.98
System 2:	30	29.90	$y = 0.9449x + 1.7358$	1	1.00
	50	51.27			1.03
	70	71.95			1.03
	100	104.10			1.04
System 4:	30	29.29	$y = 1.0056x + 0.518$	1	0.98
	50	49.37			0.99
	70	68.96			0.99
	100	98.96			0.99
System 5:	30	29.49	$y = 0.9499x + 1.788$	1	0.98
	50	50.96			1.02

	70	71.97			1.03
	100	103.25			1.03
System 6:	30	30.36	$y = 1.0049x - 0.677$	0.9999	1.01
	50	50.73			1.01
	70	70.21			1.00
	100	100.17			1.00

Table A.2: Comparison of DMA shift parameters for all systems with external routine.

<b>INTERNAL EVALUATION</b>						
System 1:			System 2:			
Dp set	GF (DP1 cor)	std GF	Dp set	GF (DP1 cor)	std GF	
10	1.59	0.03	30	1.61	0.02	
30	1.79	0.04	50	1.57	0.02	
50	1.82	0.05	70	1.58	0.01	
70	1.84	0.05	100	1.56	0.01	
System 5:			System 6:			
Dp set	GF (DP1 cor)	std GF	Dp set	GF (DP1 cor)	std GF	
30			30	1.64	0.05	
50	1.69	0.00	50	1.69	0.07	
70	1.67	0.00	70	1.70	0.07	
100	1.67	0.00	100	1.72	0.08	

Table A.3: Growth factor with standard deviation for internally evaluated data in experiment 2.

<b>EXTERNAL EVALUATION</b>						
System 1:			System 2:			
Dp set	GF (DP1 cor)	std GF	Dp set	GF (DP1 cor)	std GF	
10	1.57	0.03	30	1.63	0.02	
30	1.78	0.04	50	1.62	0.02	
50	1.82	0.04	70	1.58	0.02	
70	1.84	0.05	100	1.57	0.02	
System 5:			System 6:			
Dp set	GF (DP1 cor)	std GF	Dp set	GF (DP1 cor)	std GF	
30			30	1.64	0.05	
50	1.69	0.00	50	1.69	0.07	
70	1.67	0.00	70	1.70	0.07	
100	1.67	0.00	100	1.72	0.08	

Table A.4: Growth factor with standard deviation for externally evaluated data in experiment 2.

<b>INTERNAL EVALUATION</b>	System1	System2	System4	System5	System6
DRH	74.45	79.12	75.43	78.74	78.62
GF-DRH	1.17	1.19	1.18	1.20	1.19

GF-MAX	1.39	1.39	1.40	1.44	1.42
a	1.3939	1.3867	1.3953	1.4406	1.4234
b	0.9350	0.3568	0.3801	0.4245	0.4144
RH1	74.6298	79.1364	75.5423	78.8590	78.7342
RH2	0.6963	0.0839	0.4292	0.4428	0.4075

Table A.5: DRH with growth factor at DRH, RH1, RH2, a, b, and maximum GF for internally evaluated data in experiment 3.

<b>EXTERNAL EVALUATION</b>	System 1	System 2	System 4	System 5	System 6
DRH	78.00	77.71	75.20	78.74	78.62
GF-DRH	1.20	1.14	1.23	1.20	1.19
GF-MAX	1.42	1.39	1.43	1.44	1.42

Table A.6: DRH with growth factor at DRH and maximum GF for externally evaluated data in experiment 3.

<b>INTERNAL EVALUATION</b>									
<b>System 1</b>									
Dp set	nf1	std nf1		nf2	std nf2		nf3	std nf3	
10.00		0.14	0.06		0.83	0.07		0.03	0.02
30.00		0.41	0.04		0.59	0.04		0.00	0.00
50.00		0.48	0.02		0.52	0.02		0.00	0.00
70.00		0.62	0.05		0.37	0.06		0.01	0.01
<b>System 2</b>									
Dp set	nf1	std nf1		nf2	std nf2		nf3	std nf3	
30.00		0.40	0.01		0.59	0.02		0.01	0.00
50.00		0.47	0.03		0.52	0.03		0.02	0.01
70.00		0.56	0.05		0.41	0.05		0.02	0.01
100.00		0.74	0.05		0.19	0.05		0.07	0.01
<b>System 4</b>									
Dp set	nf1	std nf1		nf2	std nf2		nf3	std nf3	
30.00		0.25	0.09		0.20	0.11		0.55	0.13
30.00		0.29	0.18		0.71	0.18		0.00	0.00
50.00		0.30	0.05		0.50	0.17		0.20	0.13
70.00		0.43	0.11		0.55	0.11		0.01	0.00
100.00		0.47	0.10		0.49	0.09		0.05	0.02
<b>System 5</b>									
Dp set	nf1	std nf1		nf2	std nf2		nf3	std nf3	
30.00		0.31	0.00		0.69	0.00			
50.00		0.44	0.02		0.55	0.02		0.01	0.00
70.00		0.55	0.01		0.44	0.01		0.01	0.00
100.00		0.65	0.06		0.32	0.06		0.03	0.00
<b>System 6</b>									
Dp set	nf1	std nf1		nf2	std nf2		nf3	std nf3	

30.00	0.45	0.02	0.54	0.02	0.01	0.00
50.00	0.50	0.02	0.49	0.02	0.01	0.00
70.00	0.61	0.02	0.38	0.02	0.01	0.00
100.00	0.66	0.04	0.29	0.04	0.05	0.01

Table A.7: Number fraction (nf1 = more hygroscopic; nf2 = less hygroscopic; nf3 = hydrophobic ) with standard deviation for internally evaluated data in experiment 4.

<b>EXTERNAL EVALUATION</b>							
<b>System 1</b>							
Dp set	nf1	std nf1	nf2	std nf2	nf3	std nf3	
10	0.07	0.06	0.55	0.14	0.38	0.14	
30	0.37	0.06	0.63	0.06			
50	0.58	0.05	0.38	0.05	0.04	0.02	
70	0.75	0.06	0.21	0.08	0.05	0.03	
<b>System 2</b>							
Dp set	nf1	std nf1	nf2	std nf2	nf3	std nf3	
30	0.65	0.05	0.35	0.05	0.00	0.00	
50	0.50	0.19	0.44	0.08	0.06	0.15	
70	0.07	0.02	0.49	0.06	0.44	0.07	
100	0.10	0.02	0.35	0.06	0.55	0.07	
<b>System 5</b>							
Dp set	nf1	std nf1	nf2	std nf2	nf3	std nf3	
30	0.33	0.00	0.67	0.00			
50	0.45	0.02	0.55	0.02	0.01	0.00	
70	0.55	0.01	0.44	0.01	0.01	0.00	
100	0.65	0.06	0.32	0.06	0.03	0.00	
<b>System 6</b>							
Dp set	nf1	std nf1	nf2	std nf2	nf3	std nf3	
30	0.45	0.02	0.54	0.02	0.01	0.00	
50	0.50	0.02	0.49	0.02	0.01	0.00	
70	0.61	0.02	0.38	0.02	0.01	0.00	
100	0.66	0.04	0.29	0.04	0.05	0.01	

Table A.8: Number fraction (nf1 = more hygroscopic; nf2 = less hygroscopic; nf3 = hydrophobic ) with standard deviation for externally evaluated data in experiment 4.

## **B APPENDIX: Basic considerations**

### **B.1 The influence of the application of an inversion scheme / recalculation algorithm on the growth factor**

The DMA selects particles with a specified electrical mobility by a probability function called transfer function. Each DMA has an individual transfer function that can be theoretically described by a triangle function. In Figure B 1, this ideal function is graphically illustrated.

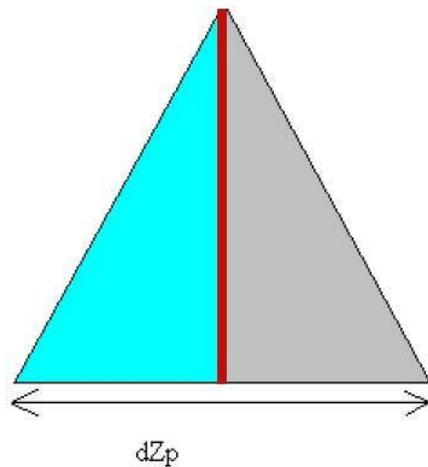


Figure B.1: Scheme of a transfer function.

By means of an inversion scheme / recalculation algorithm, the obtained data are corrected for the influence of the transfer function, the CPC efficiency, diffusion losses in the DMA and multiply charged particles. For TDMA inversion scheme / recalculation algorithms, multiply charged particles cannot be considered since the number of particles of larger particles than the determined particles is not known. Also, most TDMA inversion schemes / recalculation algorithms only take the transfer function of the second DMA into account, which means the inversion / recalculation is limited to the calculation of particle number size distributions downstream of DMA1 and upstream of DMA2 in humidified state. This calculation still leads to a broadening of the distribution since the broadening of the first DMA is not taken into account for this evaluation. We strongly recommend to invert / recalculate the data with the individual transfer function. At least, an inversion scheme / recalculation algorithm using the ideal transfer function has to be applied to receive the real number fractions of different hygroscopic growth modes. The inversion scheme / recalculation algorithm should correct for singly charged particles, because the charging probability for singly charging decreases with increasing diameter resulting in a shift of hygroscopic growth modes towards smaller diameters.

## B.2 The instability of temperatures within the H-TDMA

The instability of temperatures within the H-TDMA systems leads to large uncertainties in derived hygroscopic growth factors during measurements. The temperature at the RH control unit sometimes differs from the DMA2 temperature especially from the temperature inside the DMA2, where the measurement is taken and the particle growth is determined. In Figure B.2, this problem is schematically illustrated.

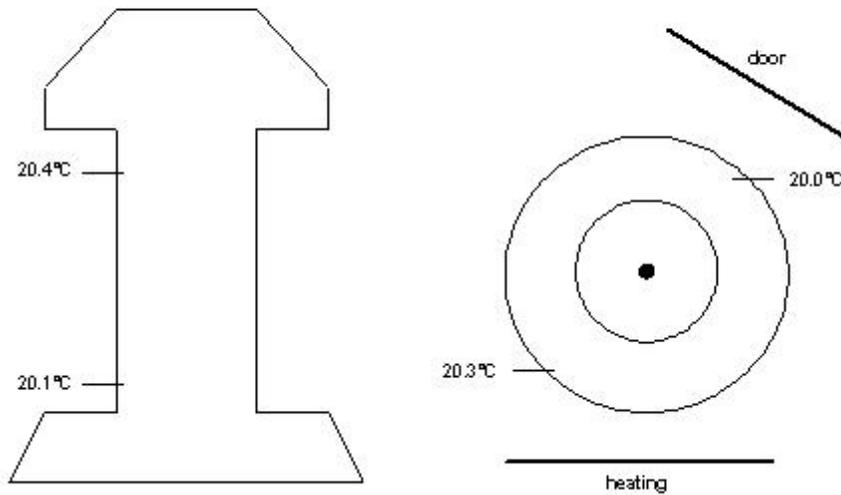


Figure B.2: Scheme of possible temperature differences inside a DMA.

We strongly recommend to thermally insulate the RH measurement control unit as well as the DMA2 itself. An exact measurement as close as possible to the inner housing of the DMA2 (sensors could be attached into the wall of the outer DMA housing) and an exact measurement at the RH control point (here the set RH is regulated) leads to an exact calculation of the RH inside DMA2. Deviations between these two temperatures and thus the control RH and real RH inside the DMA can thus be considered and be corrected for.