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Particle mobility size spectrometers: harmonization of technical standards and data structure to facilitate high quality long-term observations of atmospheric particle number size distributions

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Abstract

Particle mobility size spectrometers often referred to as DMPS (Differential Mobility Particle Sizers) or SMPS (Scanning Mobility Particle Sizers) have found a wide application in atmospheric aerosol research. However, comparability of measurements conducted world-wide is hampered by lack of generally accepted technical standards with respect to the instrumental set-up, measurement mode, data evaluation as well as quality control. This article results from several instrument intercomparison workshops conducted within the European infrastructure project EUSAAR (European Supersites for Atmospheric Aerosol Research). Under controlled laboratory conditions, the number size distribution from 20 to 200 nm determined by mobility size spectrometers of different design are within an uncertainty range of ±10% after correcting internal particle losses, while below and above this size range the discrepancies increased. Instruments with identical design agreed within ±3% in the peak number concentration when all settings were done carefully. Technical standards were developed for a minimum requirement

- of mobility size spectrometry for atmospheric aerosol measurements. Technical recommendations are given for atmospheric measurements including continuous monitoring of flow rates, temperature, pressure, and relative humidity for the sheath and sample air in the differential mobility analyser. In cooperation with EMEP (European Monitoring and Evaluation Program), a new uniform data structure was introduced for
- ²⁰ saving and disseminating the data within EMEP. This structure contains three levels: raw data, processed data, and final particle size distributions. Importantly, we recommend reporting raw measurements including all relevant instrument parameters as well as a complete documentation on all data transformation and correction steps. These technical and data structure standards aim to enhance the quality of long-term size
- ²⁵ distribution measurements, their comparability between different networks and sites, and their transparency and traceability back to raw data.



1 Introduction

Over the past 10–15 years, mobility size spectrometers have been increasingly used for long-term observations of atmospheric particle number size distributions in the sub-micrometer diameter range. Depending on the instrument, mobility size spectrometers
 ⁵ usually cover diameter ranges 3–800 nm or slightly less. Particle mobility size spectrometers are commonly referred to as DMPS (Differential Mobility Particle Sizer) or SMPS (Scanning Mobility Particle Sizer). The number size distribution of atmospheric aerosol particles is a basic, but essential parameter required in calculations of the effects of aerosols on climate, human health, and eco-systems. It is also an important parameter with regard to the description of aerosol dynamical processes as well as heterogeneous chemical reactions in the atmosphere.

The first atmospheric long-term measurements with mobility size spectrometers were realized in the 1990s at urban observation sites (Tuch et al., 1997; Woo et al., 2001; Charron and Harrison, 2003; Wehner and Wiedensohler, 2003) as well as in the

- ¹⁵ rural and remote areas (Mäkelä et al., 2000; Birmili et al., 2001; Weingartner et al., 1999). Ever since, their application has expanded to a current number of more than 20 ground-based continuous observation points across the globe, whose measurement data are used for a wide range of purposes. Within the infrastructure EUSAAR (European Supersites for Atmospheric Aerosol Research), EMEP (European Monitoring and Content and Con
- Evaluation Program), and the research project EUCAARI (European Integrated project On Aerosol Cloud Climate Air Quality Interactions), particle mobility size spectrometers were implemented at a selection of 20 ground-based atmospheric observation sites ("super-sites") in Europe. As more and more measurement sites and institutions become involved, technical standardization of custom-built and commercial instruments,
- harmonization of the data evaluation, quality control, and data dissemination is needed to obtain a minimum comparability and accuracy of the data sets.

Most modern particle mobility size spectrometers operate a differential mobility analyzer (DMA; for a general theoretical background see, e.g., Knutson and Whitby, 1975;



Liu and Pui, 1974; Flagan, 1999; Stolzenburg and McMurry, 2008) upstream of a condensation particle counter (CPC; Agarwal and Sem, 1980; Stolzenburg and McMurry, 1991; Wiedensohler et al, 1997; Hermann et al, 2007), which records particle concentrations as a function of the electrical mobility by varying the DMA voltage. Mobility spectrometers measuring particle number size distributions were developed over the last 30 years (e.g., ten Brink et al., 1983; Fissan et al., 1983; Kousaka et al., 1985; Winklmayr et al., 1991; Wang and Flagan, 1990; Chen et al., 1998).

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Commercial mobility size spectrometers have been available since the 1980s and with automatic flow control only during the past 10 years. Two examples of commer-

- ¹⁰ cial devices are the SMPS instruments from TSI Inc. (Shoreview, MN, USA) or Grimm GmbH (Ainring, Germany). Commercial instruments are generally not optimized for long-term atmospheric measurements. Several groups in the atmospheric research community have thus developed custom instruments for such applications, (e.g., Jokinen and Mäkelä, 1997; Birmili et al., 1999).
- ¹⁵ Although mobility size spectrometry was established in the 1970s, published reports on the actual quality of the data as well as instrumental comparisons are scarce. The World Meteorological Organisation issued recommendations for the design and the operation of continuous atmospheric aerosol measurements in general (WMO-GAW Report 153), but recommendations for mobility size spectrometers were omitted. Few
- intercomparisons of mobility size spectrometer measurements have been published. Khlystov et al. (2001) identified systematic differences between three different mobility size spectrometers used for atmospheric measurements. During a workshop investigating diesel exhaust, eleven instruments were compared for the first time (Dahmann et al., 2001). They concluded that different instruments (of the same and also dif-
- ferent designs) may yield similar number size distributions, but emphasized that the constant supervision of the equipment in the laboratory by qualified personnel was necessary. Imhof et al. (2006) compared four different mobility spectrometers in a road tunnel experiment. They found discrepancies between the concentrations of number size distributions of 30% in the nanoparticle size range (Dp < 50 nm), approximately</p>



25% for the size range 60 < Dp < 120 nm, and 10% for Dp > 200 nm. The reasons for these discrepancies were not clear. Helsper et al. (2008) published the results from a VDI-initiated study ("Verein Deutscher Ingenieure", The Association of German Engineers), which compared four commercial mobility size spectrometers (TSI long-SMPS,

- ⁵ TSI nano-SMPS and two Grimm-SMPS) and a custom-made TDMPS (Twin-DMPS) from the Leibniz Institute for Tropospheric Research. Integrated particle number concentrations compared well (±12%) over the limited size range of 40–350 nm for all instruments with the exception of the TSI nano-SMPS, which had a limited size range up to 200 nm. The concentrations of the number size distributions differed by a maximum
- ¹⁰ of 25% within the size range of 20–200 nm although larger discrepancies occurred outside this range. The reasons for the instrumental deviations could not be rigorously assessed, because of unknown differences in the multiple charge inversion algorithms used by the different manufacturers.

To our knowledge, no comprehensive intercomparison of custom-built mobility spectrometers has been published to date, particularly in comparison to commercial instruments. Furthermore, we could not find literature where inversion routines of mobility size spectrometers were compared. Inversion routines are the vital basis for converting measured mobility distributions into final particle number size distributions taking into account the bipolar charge distribution as well as the DMA (Differential Mobility

- 20 Analyzer) transfer function. Possible sources of uncertainty in number size distribution measurements include DMA transfer functions, particle losses, size- and compositiondependent CPC counting efficiencies, or differences in the bipolar charge distribution. In practice, instabilities in the aerosol or sheath flow rates as well as an unmeasured relative humidity in an instrument are likely to cause additional uncertainties in number
- ²⁵ concentrations and sizing. Inconsistent practices among the scientific groups regarding the measurement and automatic monitoring of such instrumental parameters made it difficult to evaluate the quality of reported size distributions. Most scientists usually report final size distributions only, making it difficult to trace instrumental differences back to the level of raw concentrations.



The described inconsistencies and uncertainties in technical performance and undefined data quality lead to a need to harmonize technical standards of mobility size spectrometers and the data structure to ensure high data accuracy, comparability, transparency, and traceability of the measured particle number size distribution.

- In this paper, we recommend minimum technical standards for the design and the operation of mobility size spectrometers including the submission of data and metadata to data archives. This work was accomplished by the European infrastructure project EU-SAAR, the network of Excellence ACCENT (Atmospheric Composition Change: A European Network), the WMO-GAW programme (Global Atmosphere Watch), and EMEP
- ¹⁰ (European Monitoring and Evaluation Program). An additional goal is that our recommendation should be adapted for commercial mobility size spectrometers in the future.

2 The principles of mobility size spectrometers

This section briefly outlines the physical principles of modern mobility size spectrometers. For more details, the reader is referred to reports on the development of spectrometers (e.g., ten Brink et al., 1983; Fissan et al., 1983; Kousaka et al., 1985; Wang and Flagan, 1990; Winklmayr et al., 1991; Chen et al., 1998; Jokinen and Mäkelä, 1997; Birmili et al., 1999; Hinds, 1999), or the documentation provided by commercial manufacturers. Most mobility size spectrometers consist of a sequential set-up of a bipolar charger (or traditionally named neutralizer), DMA, and CPC. By setting different voltages in the DMA, particles of different mobility are selected and their concentration can be measured. Ramping or stepping the voltage yields an electrical mobility size distribution, which can later be inverted into a particle number size distribution.

Before the aerosol particles enter the DMA, they are brought to a bipolar charge equilibrium using a bipolar charger. Positive and negative ions are produced continuously in this charger, for instance by a radioactive source. The radioactive sources used in field observation include ⁸⁵Kr and ⁶³Ni (both β -radiators), and ²⁴¹Am and ²¹⁰Po (both



number concentration should be significantly lower than the equilibrium ion pair concentration, which is believed to be approximately 10^7 cm^{-3} . Mobility size spectrometers can be thus used for total number concentrations up to 10^6 cm^{-3} only. A complication is that α -particles emitted from radioactive sources such as ²⁴¹Am and ²¹⁰Po lose their $_5$ energy within 5 cm traveling distance in air and produce beside air ions also ozone leading to the production of unintended nucleation particles.

A DMA is usually built as a cylindrical capacitor. The charged aerosol is thus injected through an annular slit closed to the outer electrode into the DMA and then laminarly merged with the particle-free sheath air flow. In the DMA, charged particles are separated according to their electrical mobility. The electrical mobility depends primarily on particle charge (positively) and particle diameter (inversely), but also on the gas viscosity (see Appendix A.5), particle shape, Cunningham correction factor, and hence also indirectly temperature and pressure of the gas flowing inside the DMA (e.g., Hinds, 1999). The smaller the particle diameter and the higher the number of

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- charges, the larger is the particle's electrical mobility. By knowing the dimensions of the DMA (length and radii of the inner and outer electrode), one can calculate the voltage between the electrodes needed to transport charged particles with a certain electrical mobility from the entrance to a annular slit in the centre rod of the capacitor. The sample flow carrying particles with the same small range of electrical mobility is
- ²⁰ drawn through this small slit. The particle number concentration in this sample flow is measured in a CPC. The mobility distribution is determined by scanning the voltage through the entire particle mobility range to be investigated.

Usually, the set-up of a mobility spectrometer requires upstream of the DMA the deployment of a pre-impactor. The role of this pre-impactor is to remove all particles larger than the upper size limit of the mobility spectrometer. This ensures that no multiply charged particles with sizes larger than the measurement range can interfere with the measured mobility spectrum, which is a prerequisite for a correct multiple charge inversion scheme. The impactor can be omitted for most atmospheric applications, if



the size range of the mobility size spectrometer extends up to 800 nm or more. The

reason is that the atmospheric number size distribution declines very steeply towards larger particle sizes, making the contributions of multiple charged particles much less. Sensitivity tests have shown that multiple charges on particles larger than 800 nm diameter are only important in the case of exceptional amounts of these large particles,
 ⁵ such as during dust events (Birmili et al., 2008; Schladitz et al., 2010).

The particle number size distribution is derived from the measured electrical mobility distribution with an inversion routine (Hoppel, 1978; Knutsen, 1976; Stolzenburg and McMurry, 2008). Here, the knowledge of the bipolar charge distribution (Wiedensohler, 1988; Baron and Willeke, 2005; see also Appendix A.5), the DMA-transfer function (Birmili et al., 1997), and the CPC counting efficiency function (e.g., Wiedensohler et al., 1997; Hermann et al., 2007) are used.

In the field, the quality of mobility spectrometer measurements depends essentially on the stability of the aerosol and sheath air flow rates, as well as the performance of the CPC. An error in the sheath air flow rate of 1% corresponds to a shift of 1% in the

- ¹⁵ selected electrical particle mobility. For the typical ratio of sheath air flow rate to sample air flow rate, 10:1, a leak in the loop of the sheath air flow of 1% would cause a 10% error in the aerosol flow rate. A change in the aerosol flow translates directly into the measured number concentrations. Furthermore, particle-contaminated sheath air or leaks between DMA and CPC cause rather large errors. It is also essential to calculate
- ²⁰ the DMA voltages for the actual temperature and pressure conditions encountered at the field station. It is appropriate to calculate the voltages on the basis of mean indoor temperature and pressure at the station, and additionally to monitor pressure and temperature in the instrument as confirmation. A pressure variation of ±20 hPa leads to an uncertainty in sizing of 1% and is thus tolerable, while temperature fluctuations within $\pm 10^{\circ}$ C are not critical in sizing. It is thus vitally important to use the correct mean
- pressure when operating instruments at high altitudes.

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Another important issue for atmospheric size distributions is the relative humidity (RH) in the instrument. Atmospheric aerosol particles containing water-soluble material may contain significant amounts of water well below the deliquescence RH of the



individual compounds. Ambient air samples can increase considerable their RH when cooled down after entering an air-conditioned laboratory. The typical growth of atmospheric particles larger than 100 nm in diameter is 1.3–1.6 at 90% RH (Swietlicki et al., 2008) depending on the mass fraction and nature of water-soluble particle material.

⁵ The solution to achieve comparability between measurements is to limit the relative humidity by drying the sample aerosol. Preferably, the RH should be kept below 40%, which minimizes diameter changes due to hygroscopic growth of typically less than 5% (Swietlicki et al., 2008).

3 Harmonization of the technical standard

- Within the EUSAAR project, we developed technical standards for mobility size spectrometers. To improve a world-wide comparability and accuracy of measurement data, it is suggested that future long-term observational measurements adhere to these standards. A schematic of our recommended, closed-loop-based, mobility size spectrometer is given in Fig. 1. The set-up includes dryers for aerosol flow and sheath air, a heat
 exchanger, high efficiency particle filters, and sensors for aerosol and sheath air flow rate, relative humidity and temperature of aerosol flow and sheath air, and absolute pressure in the aerosol flow before entering the DMA. All recommended system parameters should be recorded and stored with at least the same time resolution as the measured size distributions. Further details for operation of the standardized mobility spectrometer include:
 - The aerosol size distribution is measured under "dry conditions", i.e. RH (relative humidity) <40%. This low humidity regime ensures that changes in particle size due to hygroscopic growth remain negligible. We recommend the use of a membrane dryer (e.g. Nafion), or a silica-based aerosol diffusion drier that also features minimum particle losses. Generally, a dry aerosol sample is needed in the bipolar charger to ensure the bipolar charge equilibrium of the aerosol particles. Particle losses in the dryer, e.g. by Brownian diffusion, are characterized</p>



and accounted for in the data analysis. The RH of the aerosol flow should be monitored by a calibrated humidity sensor with a maximum uncertainty of 5% RH across the range of 10–90%.

- The aerosol volumetric flow rate is monitored with a calibrated flow meter. This is preferably a calibrated pressure transducer measuring the pressure drop across a laminar flow element (" Δp capillary"). It is not recommended to use mass flow sensors for the aerosol flow because they exhibit undesired particle losses. In the running instrument, the aerosol flow should not deviate systematically more than 5% from the set-point on daily average. In addition, the aerosol sample flow should be checked as often as possible using a reference flow meter, but at least at each service occasion.

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- The sheath air flow rate is also dried below 40% RH in a similar fashion as the aerosol inlet flow to prevent the accumulation of moisture (i.e. by a membrane or diffusion dryer). Importantly, the RH in the sheath air flow rate determines the equilibrium particle size during the mobility measurement. The RH of the sheath air has to be monitored with a calibrated humidity sensor as stated above. The RH sensor should preferably be installed either at the excess air outlet or at the sheath air entrance of the DMA, i.e. at temperature and pressure that are the closest to those in the DMA. The temperature of the sheath air RH sensor should not differ more than 1 K from the temperature in the DMA.
- In the case of a closed-loop technique for the sheath air flow, a heat exchanger is needed to remove the excess heat generated by the pump or blower. A closedloop system employs two HEPA filters (High Efficiency Particle Filter), one before entering and another after leaving the DMA. The pressure drop across the HEPA filters should be minimal to ensure a correct flow measurement in the sheath air loop. For a critical orifice/pump set-up, the absolute pressure downstream of the critical orifice should be monitored to ensure critical flow conditions (pressure downstream less than half of the upstream pressure).



- The sheath air flow rate is monitored with a calibrated flow meter. This is preferably a capillary measuring the pressure drop or a mass flow meter. Capillaries or mass flow meters should be installed at the sheath air inlet (before the HEPA filter) or excess air outlet (after the HEPA filter) to monitor the flow rate at a pressure representative for the DMA. A mass flow meter should be calibrated for volumetric flow. The mean sheath air flow should be kept as constant as possible, with a maximum deviation of 2% around the set-point value. This criterion can be met either by a critical orifice/pump set-up or by a software-controlled blower.

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- Temperature and absolute pressure representative of the conditions inside the DMA should be monitored because they are needed to ascertain the correct sizing of the particles and to correct the final data to standard conditions afterwards (see also Appendix A4.3). These are preferentially monitored at the aerosol inlet of the DMA.
- Electrical mobility distributions are stored as particle number concentrations as a function of nominal particle size (equivalent Stokes diameter for singly charged particles). This allows measurement data to be traced back to their instrumental origins, and permits direct intercomparisons regardless of the subsequent inversion routines and correction schemes. If necessary, other evaluation steps could be applied. The DMA dimensions (rod diameter and cylinder inner diameter and length) sheath and aerosol and flow rates, and possibly the serial numbers of the DMA and CPC should be supplied in the meta-data.
 - When dual SMPS or DMPS systems such as a TDMPS (Twin Differential Mobility Particle Sizer) are used to encompass a wider particle size range (such as below 10 nm), the system parameters for each DMA need to be monitored and recorded.
- To calculate the dynamic viscosity and mean free path of air as function of temperature and pressure, we recommend to use the constants and formulas of ISO 15900, which are given in Appendix A5.



 We generally recommend the bipolar charge distribution of ISO 15900 to be used in inversion routines. This bipolar charge distribution is based on the approximation formulas and the Gunn equation (1956) in Wiedensohler (1998) with corrections to two approximation coefficients published in Baron and Willeke (2005) and also listed in Appendix A5.

4 Intercomparison studies

4.1 Inversion routines

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Our intercomparison of inversion software includes 12 different variants of routines operated by the project participants (research groups and commercial companies). As a test bed, one day of mobility size spectrometer measurements of ambient aerosol was used. Identical copies of the raw data set (number concentrations versus mobility diameter classes as well as DMA and CPC specifications) were supplied to each group to run their individual inversion routine.

All inversion routines used the multiple charge correction for negatively charged particles. The pragmatic reason was that most participating groups apply positive voltages to their mobility size spectrometers. This also meant that some groups ran their inversion program in a non-standard fashion for this investigation. Short descriptions of all inversion routines are given in Appendix A.2 (Table 2).

To make the calculations comparable, an ideal DMA transfer function for all inversion programs was used (Knutson and Whitby, 1975). Apart from "Grimm old" (using Boltzmann charging probabilities for large particles), all inversion routines used the ISO 15900 recommendations (determination of particle size distribution – differential electrical mobility analysis for aerosol particles) for the bipolar charge equilibrium. As shown in the upper plots of Fig. 2a and b, the number size distributions determined

²⁵ by the different inversion routines visually agreed with some small differences. The shaded area represents the $\pm 5\%$ range around the results of IFT inversion routine.



To see small difference as function of particle size, we plotted the ratio of the results of any individual inversion routine against the IFT one (lower plots in Fig. 2a and b). Apart from "Old Grimm", all inversion routines agreed within 4% over the size range. Only towards the end of the size range, some discrepancies occur probably due to uncertainties by transferring the EUSAAR data format to individual formats for the individual inversion routines. Note: the "Old Grimm" inversion routine is not any used anymore in newer software revisions. The results give us the confidence that significant differences in number size distribution by comparing mobility size spectrometers are not due to the applied inversion routines.

10 4.2 CPC counting efficiency curves

Another important part of the mobility size spectrometer is the detector, which is most often a condensation particle counter (CPC). Correct measurement of the number size distribution for the smallest particles depends critically on accounting for the size-dependent particle detection efficiency of the CPC. In order to ensure long-term

¹⁵ quality-assured data, the CPC should be technically checked and/or calibrated preferably against a reference standard every year. Ideally, the CPC is compared to a reference instrument at a calibration centre or at the manufacturer.

An accurate calibration of the detection efficiency of a CPC depends strongly on the traceability of the reference instrument such as an aerosol electrometer, which mea-

- ²⁰ sures the electric current of charged particles in an air flow. Attempts were recently taken to provide defined charged aerosol particles for CPC calibrations (Yli-Ojanperä et al., 2010; Fletcher et al., 2009). From electric current measured by the electrometer, the particle number concentration can be directly calculated by knowing the exact aerosol flow rate and assuming only singly charged particles. Since the electrical current current aerosol has be low as 10^{-15} A the signal has to be highly amplified by a factor of 10^{12} . The
- rent can be low as 10⁻¹⁵ A, the signal has to be highly amplified by a factor of 10¹². The uncertainty of the measurements depends thus on the uncertainty of the resistance in the amplifier circuit.



In the following, we describe the determination of the counting efficiency of CPCs as presently done at the WMO-GAW-WCCAP in Leipzig (World Calibration Centre for Aerosol Physic) within the EUSAAR and GAW networks (Wiedensohler et al., 1997). The results of the CPC calibration in the frame of third EUSAAR DMPS/SMPS inter-⁵ comparison workshop (Sect. 4.3) held in Leipzig 2009 are shown here an example. Briefly, we nucleate a silver aerosol in a tube furnace (Scheibel and Porstendörfer, 1983) and select monodisperse particles in the range from 3–40 nm using a DMA. For the size range up to 40 nm, we achieve after DMA mainly singly charged particles.

Due to the relative small polydisperse aerosol (geometric standard deviation of 1.3– 1.4), the concentration of double charged particles at 40 nm is negligible. After dilution, the response of each CPC was then compared against a reference electrometer as a function of particle diameter. All particle counters and the aerosol electrometer were connected to a common manifold that was designed to minimize particle losses.

In the first calibration step, we used ten particle counters (models TSI 3010 and 3772) and compared the number concentration for 40 nm particles. Taking into account

- ¹⁵ 3772) and compared the number concentration for 40 nm particles. Taking into account the actual flow rate of each particle counter, the unit-to-unit variability was maximum $\pm 3\%$ at 40 nm around the mean value. Since the exact amplification of our aerosol electrometer is not known, we *defined* the average counting efficiency of these CPCs as 100%. This is a necessary assumption to define an absolute measure. The average
- number concentration was then compared to the aerosol electrometer. Herewith, we determined the gain error of the electrometer, which was used to determine the CPC counting efficiencies for the size range smaller than 40 nm. Presently, the accuracy was not determined. This could only be done by a measurement of the resistance of the amplifier with a very high precision.
- The resulting detection efficiency curves are shown in Fig. 3. Four CPCs were set to a temperature difference of 25 °C between saturator and condenser, while the other CPCs were operated at the nominal, factory-set temperature difference of 17 °C. The counting efficiency curves can be used later to correct the ambient aerosol size distribution measurements. It can be seen that the lower cut-off size of the different CPCs



depends significantly on the particular design, but also on operational parameters, particularly the condenser/saturator temperature difference. Furthermore, CPCs operating with the nominal, factory-set temperature difference may also differ by few Nanometres in their actual 50% detection efficiency diameter.

5 4.3 Upgraded mobility size spectrometers

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To evaluate the performance of complete mobility size spectrometers, several custombuilt instruments from the EUSAAR network were compared. The goal of this comparison was to understand the unit-to-unit variability of mobility size spectrometers obeying comparable technical standards under controlled laboratory conditions. We emphasize that all mobility spectrometers under study were upgraded according to the technical harmonization described in Sect. 3, with a long-term deployment at EUSAAR measurement sites in mind.

Our results are based on a series of three intercomparison workshops conducted between 2006 and 2009 at the WCCAP facilities in Leipzig in the frame of the EUSAAR,

- ACCENT (Atmospheric Composition Change: a European Network), and WMO-GAW. The first workshop in November 2006 aimed at obtaining the status of comparability of all participants' instrumentation, and to plan the technical harmonization of all instruments for high quality long-term size distribution measurements. During the second workshop in March 2008, we checked whether all instruments were correctly upgraded,
- ²⁰ how their performance had improved, and what kind of discrepancies remained. After further improvements, a third intercomparison workshop was held in June 2009 to evaluate the comparability of the improved mobility size spectrometers, including commercial and custom-built instruments. Specific details of the mobility size spectrometers used during the third workshop are listed in Table 3 in Appendix A.3. To aim for a max-
- imum comparability between instruments, we repeated some of the experiments in August 2010 using identical mobility size spectrometers incl. the two reference instruments.



In the following sections, results of the calibrations and intercomparisons of the third workshop are given describing the status of the mobility size spectrometer under controlled laboratory conditions.

4.3.1 Sizing accuracy using PSL particles

- A mandatory part of a performance check of a mobility spectrometer concerns the sizing accuracy. This task can be accomplished using monodisperse PSL (polystyrene latex) spheres, whose diameters are certified by the manufacturer to be within ±2.5% of the nominal diameter. For size calibration, monodisperse PSL spheres of one or more particle sizes should be used (Mulholland et al., 2006). For practical reasons, we recommend to use at least 200 nm PSL particles: on one hand, a sufficient particle
- number concentration will remain after nebulisation, while on the other hand, a suncient particle number concentration will remain after nebulisation, while on the other hand, a minimum amount of residual material will usually be left on the surface of the particles after emerging from the aqueous suspension. Taking into account an additional uncertainty of the sheath air flow rate of $\pm 1\%$ for the individual spectrometers, we defined that a deviation of $\pm 3.5\%$ from the nominal diameter of the PSL particles is tolerable for an
 - instrument to pass the sizing quality test.

In practice, the nebulized PSL particles were pre-neutralized using a bipolar charger (85 Kr; 370 MBq) and then fed into a well-mixed 0.5 m³ mixing chamber, which fed all mobility size spectrometers through equivalent connecting tubes. Mixing in the cham-

²⁰ ber was achieved with a fan. During the experiment, the resulting number concentration of latex particles could be modified by changing the supply rate of dry, particle-free air into the chamber.

Inverted number size distributions of 200 nm PSL spheres are plotted in Fig. 4. As indicated above, the width of the measured latex particle distribution depends mainly on

the ratio of the aerosol to sheath air flow rate (see legend of Fig. 4) and on the standard deviation of the size of the PSL particles. The centroid diameters of all mobility size spectrometers were within the uncertainty range of 3.5% except the TNO instrument, which underestimated the 200 nm PSL size, which was due to an erroneous sheath air



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flow rate. We adjusted the flow rate after the experiments and the TNO instruments works well again.

4.3.2 Particle number concentration and size distribution

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In a final step, the comparability of mobility size spectrometers was evaluated by simul taneous sampling of ambient aerosols. For the EUSAAR network, the WCCAP built two "reference mobility size spectrometer" dedicated to instrumental intercomparisons during laboratory workshops and site visits. Note that the nomenclature "reference instrument" is not intended here to play the role of an absolute standard. An absolute standard for particle number concentration can only be defined by linkage to SI standards, which has not yet been successfully achieved. Therefore, the EUSAAR (and WCCAP) "reference instruments" can only be regarded as an intermediate comparison standard. As a second "reference instrument", particularly with regard to total particle number concentration, a total particle counter (CPC 3010 from TSI) with a standard saturator/condenser temperature difference of 17 °C and flow rate of 11 min⁻¹ was

Comparison experiments involved sampling ambient aerosols through the 0.5 m³ mixing chamber described above. All mobility size spectrometers sampled from this chamber through equivalent connecting tubes. Each participating group processed their electrical mobility distributions using their individual inversion routine. To take into account particle losses by diffusion in the mobility size spectrometers, we agreed on the standardized method described in detail in Sect. 4.4. Briefly, particle losses across individual components of the individual instruments were simulated by the loss across a laminar flow tube with a certain equivalent length. All size distributions shown in the following section corrected for particle losses following this standardization.

²⁵ We present results from run# 4 of the intercomparison workshop, since for this experiment, several instrumental problems were finally solved that had occurred before. Unfortunately, however, not every mobility size spectrometer present in the workshop took part in run# 4. Figure 5 shows the final number size distributions of ambient aerosol run# 4. These particular periods were selected because a) a maximum of instruments were available at this time and b) the ambient aerosol concentration showed limited variability.

- The shaded area in Fig. 5 represents a ±10% range around the number size distribution measured by the EUSAAR reference instrument. Visually, all number size distributions agree from 20 to 200 nm within the shaded ±10% range of the reference instrument. However, outside the 20–200 nm diameter range, the discrepancies increase. Three spectrometers were specifically designed to measure the number size distribution of nucleation mode particles (IFT-TDMPS, FMI, and UHEL). They deviate
- a distribution of nucleation mode particles (IFFTDMFS, FM, and OFEL). They deviate significantly below 10 nm. The reason for these differences could not be clarified during this work, and needs to be investigated in a dedicated study in the future. For the other mobility size spectrometers, deviations below 20 nm are probably due to unconsidered additional losses and increased uncertainties in the DMA voltage. Above 200 nm, low
- counting statistics become an issue and the uncertainty range between the instruments obviously increases towards larger particle sizes. However, we were not able to firmly evaluate the true reasons for the deviations at the upper end of the size distribution.

Figure 6 compares the integral number concentration of the mobility size spectrometers for particles >10 nm with the value measured by the reference CPC 3010. The

shaded area represents the ±10% range around the directly measure number concentration by the CPC. In the beginning of run# 4, the readings of the mobility spectrometers fell below the reference CPC concentration. Our explanation is that a significant number of nucleation mode particles around 10 nm were present, which were only partially detected by the mobility spectrometers. Note: some mobility spectrometers showed no sensitivity at all to particles <15 nm. After the nucleation mode particles disappeared, the number concentration of all instruments differed maximum ±12% compared to the directly measured number concentration.

Figure 7 shows a comparison of the integral number concentrations larger than 100 nm for non-diffusive aerosol particles. The shaded area is the $\pm 10\%$ range around



the concentration given from the reference instrument. Number concentrations from all instruments agree within this range compared to the reference instrument. Note: the major number fraction of the sampled particles is contained in the size range 100–200 nm.

- ⁵ Often, number size distributions are used to calculate properties of a higher moment of the size distribution, such as the surface area, volume, or mass concentration, or the light scattering coefficient. Increasing sizing and counting uncertainties in the number size distribution towards larger particles can lead to significant inaccuracies of these higher moments. In Fig. 8, we plot volume size distributions derived from the number
- ¹⁰ size distributions of run# 4, assuming spherical particles. Again, the shaded area represents a ±25% range around the particle volume size distribution of the reference system. All spectrometers are within this 25% range at the volume peak (250 nm) beside the TNO instrument (~40%). Towards the end of the size range, the discrepancy of two mobility spectrometers increases up to 70% compared to the IFT reference system. Since there is no absolute standard of aerosol volume distribution, we cannot
- judge which system provides the correct value. At present, we are however not able to firmly conclude what the underlying reasons for the observed divergences are.

In August 2010, we repeated some experiments using five mechanically identical mobility size spectrometers, including the two EUSAAR reference instruments (IFT

- ²⁰ REF 1 and REF 2). We calibrated all flow rates with extreme care, and confirmed that the sizing accuracy of the instruments was within $\pm 2\%$ of the nominal PSL (200 nm) particle diameter. Size distributions of the ambient aerosol sampled through the mixing chamber are shown in Fig. 9. The maximum deviation of all different mobility size spectrometers was found to be $\pm 3\%$ from the average at the number peak (40 nm) of
- ²⁵ the size distribution. The light grey shaded area represents the ±10% range around the average of the five mobility size spectrometers. Additionally, we plotted the calculated volume size distributions of all mobility size spectrometers. The dark grey shaded area is again the ±10% range around the average of all systems. All spectrometers are within this range. This unit-to-unit variability of identical mobility size spectrometers



represent the accuracy that can be achieved under controlled laboratory conditions at the current the state of knowledge and technology.

Based on the experience of three comprehensive intercomparison workshops and on-going efforts in comparing different instruments at the WCCAP, we conclude that

- ⁵ the instrumental uncertainties presented in this paper can be regarded as a lower limit for the uncertainties encountered in long-term, field measurements. Considering the span of environmental conditions found at the variety of field stations in operation, it is likely that effects relating to, e.g., temperature and humidity fluctuations in the field laboratories, instabilities in line power, differences in atmospheric pressure, the lower
- degree of maintenance as compared to the laboratory workshops, and changes to the instrumentation during their transport to remote locations might add up to uncertainties that affect the comparison of global atmospheric data. While many of the latter effects can probably not be entirely avoided, it appears even more crucial to ensure a homogeneous technical standard of the instrumentation, which is a matter that can be well planned in advance.
 - Consequently, we recommend standard operating procedures described in Appendix A.1 and to perform (a) frequent on-site intercomparison to a reference instrument (CPC or mobility size spectrometer of a central calibration facility) and (b) additionally sizing calibrations using a PSL standard. Only then can the quality of ambient aerosol measurements be expected to fall within the range of uncertainty determined in the work here.

4.4 Correction of particle losses

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As mentioned in the Sect. 4.3, particle losses, particularly due to diffusion, occur in all sub-parts of the mobility size spectrometer. Here, we report on the details of the particle loss quantification.



4.4.1 Plumbing

Particle losses by Brownian diffusion in a straight pipe can be corrected by analytical formulae derived for the laminar flow regime (Hinds, 1999). For fully developed laminar flow, these losses depend only on the pipe length, the flow rate through the pipe, and the particle diffusivity. Enhanced diffusional particle losses may occur in sampling

5 pipes containing bends or elbows, which depend however on the radius of the bend or elbow (Wang et al., 2002). It is also vital that the plumbing consists of fully conducting material, preferably stainless steel. Non-conductive tubing may remove a considerable number of charged particles by electrostatic forces.

4.4.2 Neutralizer and DMA 10

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Losses also occur in bipolar chargers. It is the safest practice to experimentally verify these losses as a function of particle size. Then, a loss correction can be directly applied based on the experimentally determined loss function. A more widely used approach is to fit the loss function using the diffusional deposition formula (Hinds, 1999) and to obtain an equivalent length of a straight pipe. Once the equivalent length of 15 a device is available, particle losses can be easily computed. Covert et al. (1997) determined particle losses for sub-10 nm particles across ⁸⁵Kr bipolar chargers, reporting an equivalent pipe length of 1 m (Table 1). In Wang et al. (2007), the penetration efficiencies through ten different types of bipolar chargers were experimentally and numerically investigated.

The probability of a particle penetrating through a DMA depends on the losses in the DMA inlet and outlet and the transfer function in the DMA classification region. These losses can be either simulated by diffusional deposition models, or estimated experimentally (e.g., Stolzenburg, 1988; Fissan et al., 1996; Birmili et al., 1997). Again,

the losses across these devices can be simulated by an equivalent length of a straight 25 pipe (e.g., Reineking and Porstendorfer, 1986; Karlsson and Martinsson, 2003; Jiang et al., 2010). Some relevant lengths are reported in Table 1.



4.4.3 Condensation particle counter

Each CPC may have a rather individual particle counting efficiency, which can be determined experimentally (Sect. 4.2). The real counting efficiency of a CPC can depend on many individual factors, such as CPC geometry, or the actual supersaturation profile

- ⁵ inside the condenser. If such data are not available, a counting efficiency curve of the manufacturer can be applied. In any case, it is preferable to calibrate CPCs individually, as our experience suggests that the performance of the CPC alters after approximately one year of continuous ambient measurements due to laser deterioration or contamination of the optics. When calibrating CPCs against an aerosol electrometer as a reference (Liu and Pui, 1974), particle losses inside the CPCs are implicitly included in
- erence (Liu and Pui, 1974), particle losses inside the CPCs are implicitly included in the reported counting efficiency. Particle losses inside CPCs are primarily caused by diffusion and can be estimated using diffusional deposition models (Stolzenburg and McMurry, 1991).

5 Traceability of mobility size spectrometer data

In the EMEP data base (called "EBAS"), particle number size distributions have traditionally been stored as hourly average concentrations for standard temperature (273.15 K) and pressure (1013 hPa). Because of the previous lack of harmonization of the multiple charge inversion routines as well as subsequent correction procedures, the quality of the previously published data sets is poorly known. Traceability of the data and evaluation of uncertainties are only possible if the data originators supply raw mobility distributions, system parameters, and documentation of the data evaluation steps along with the final measurement data.

To encourage such a traceability of measurement data, an extended data structure for particle number size distribution measurements was developed within EUSAAR.

²⁵ The new structure consists of three levels, basically representing "raw", "intermediate", and "final" data. "Raw" encompasses raw values measured directly by the instrument,



including instrumental status parameters. "Intermediate" contains particle number size distributions at the maximum time resolution, and after applying multiple charge inversions and all instrumental corrections. "Final" adds averaging to hourly values and standardization to standard condition of 273.15 K and 1013 hPa. At all stages, metadata, which contain extended descriptions and gualifiers of the data, are included. The

⁵ data, which contain extended descriptions and qualifiers of the data, are included. The three-level structure was designed to ensure traceability of published measurement data back to their instrumental origins.

Besides the traceability of measurement data and the harmonization of their quality assurance procedures, a number of additional objectives were kept in mind when defining the data structure and file formats:

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- The data format should be suitable for near-real-time transmission to archives, such as the EUSAAR/EMEP data centre (EBAS) and the WMO GAW-WDCA (*World Data Centre for Aerosols*) hosted at NILU (*Norwegian Institute for Air Research*). This is intended as a first step towards including aerosol data in online information systems.
- To this end, the data format should include all information necessary to allow for near-real-time, automated, data quality checks.
- The format should collect all instrument metadata so that they can be stored and archived together with the raw data, to allow for future reprocessing of the data similar to procedures already implemented for satellite observation data.
- While defining the data format, we attempted to anticipate a wider range of possible uses in the future, and thus make additional provisions in the data formatting, last but not least to avoid frequent redefinitions of the data structure and formats. An example for this is the near-real-time assimilation of aerosol data into weather forecast systems or comparisons of field data with climate models. Here, information about the uncertainty is required. The data structure therefore allows propagation of the data uncertainty through all data levels.



 To ease the adaptation to the new format by the data originators, it is defined to allow for gradual implementation, i.e. it distinguishes between compulsory and optional parameters.

A detailed description of the data formats for the three levels (raw, intermediate, and final) is provided in Appendix A.4. A description of how the data were processed should be given, either in an additional "read-me" file or in the commenting part of the meta-

data.

6 Summary

This work summarizes new experimental evidence from the intercomparison of custombuilt as well as commercial particle mobility size spectrometers (e.g. SMPS, DMPS, TDMPS), and provides recommendations to harmonize the instrumental standards and modes of operation of such measurements, including data evaluation and reporting. In the framework of the European infrastructure project EUSAAR, we standardized the technical set-up of mobility size spectrometers for long-term observations. This
standard set-up includes a recommendation to control, measure, and store all relevant system parameters, such as the flow rate, temperature, relative humidity in the aerosol and sheath air streams, and the absolute pressure close to the DMA. The relative humidity in the aerosol and sheath air should be kept below 40% at all times. Drying

of the aerosol sample flow is usually required if the dew point temperature exceeds 10 °C and the room temperature is kept to approximately 22 °C. Note: if the dew point temperature is occasionally high then the room temperature, the inlet flow has to be dried before entering the room.

Specific instrumental checks and calibrations are necessary at a regular frequency to allow long-term operation of mobility size spectrometers at field stations, as listed in the standard operation procedures in the Appendix A1. Most important are regular checks and calibration of the flow sensors, checking of the sizing accuracy by latex particles,



and comparisons against a reference mobility size spectrometer. Furthermore, proper function of the condensation particle counter, including the counting efficiency, has to be checked at least once per year.

It is also recommended to use common constants and equations determining the 5 electric mobility for the measurement and the bipolar charge distribution needed to convert the measured mobility charge distribution into the number size distribution. These constants and equations follow the recommendations of the new ISO standard 15900.

We compared different commercial and custom-programmed inversion routines that calculate, from a given set of mobility distributions, the final number size distribution for 10 an ideal transfer function, without correction for particle losses in the system. Most of inversion routines seem to perform in a similar fashion, with maximum deviations of 5% with respect to individual sections of the particle number size distribution. Deviations in the number size distribution between individual mobility size spectrometers are thus not attributable to the inversion programs. 15

In a series of intercomparison workshops, the range of uncertainty among commercial and custom-built SMPS and DMPS was determined. All participating mobility size spectrometers were upgraded to the new standard set-up and operated by experienced users. We recommended a common procedure for the correction of particle losses in the SMPS or DMPS. The conclusion is that we can reach uncertainties smaller that 20 10% with state-of-the-art mobility size spectrometers for the size range between 20 and 200 nm in particle diameter. For smaller particle sizes, the deviations become significantly greater. Divergences at the upper end of the size spectrum (300-800 nm) were also observed. The underlying reasons for these deviations are not well understood at present.

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We developed a new three-level data submission protocol for number size distribution data, which is implemented in the EBAS data base (EMEP and GAW). Level-0 data contain raw mobility distribution data and system parameters as given above. The mobility distribution should be provided as an array of number concentrations versus



particle diameters (electrical mobility for singly charged particles). Level-1 data are number size distributions as calculated after inversion and loss correction for the temperature, pressure and time resolution as measured. In level-2, the number size distributions are given as hourly averages corrected to standard temperature (0 °C) and pressure (1013.25 hPa).

Presently, these technical standards and data protocols are implemented in a number of observational networks including the EUSAAR-sites, parts of the GAW network (e.g. Point Barrow, Alaska and Shangdianzi, China), the Nordic Network (Tunved et al., 2003), and the German Ultrafine Aerosol Network (11 stations in Germany; Birmili et al., 2009).

It is a goal of this publication that world-wide measurements of mobility size spectrometers become increasingly standardized and comparable, which adds to the reliability and usefulness of the global surface-based atmospheric observation network. Another incentive was to propose enhanced technical standards that will hopefully be adopted by manufactures and applied in commercial mobility size spectrometers.

Appendix A

A1 Standard operation procedure for calibrations and system checks for mobility size spectrometers

For long-term measurements, we recommend the following listed items to improve the quality of the measurements.

- Pressure transducers or mass flow meters used to measure aerosol and sheath air flow rates have to be calibrated at least twice a year. The aerosol and sheath air flow rates should be regularly measured once per month with an independent flow standard such as an electrical bubble flow. The reference standard should have a low pressure drop. The flow rate at the pressure within the DMA should be determined.



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- Humidity sensors for the aerosol flow and sheath air have to be calibrated prior their deployment and afterwards at least once per year.
- The response function of the high voltage supply should be calibrated. This should include the analogue output module, if high voltage supply is controlled through an analogue voltage. The calibration function of the high voltage should be implemented into the scanning software or the data analysis. Correct sizing of small particles is highly sensitive to accurate knowledge of the applied voltage. Particular care is hence required in the low voltage range. Please, use a HV-probe with ultralow impedance. Check the high voltage power supply as frequently as possible.

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- Furthermore, Condensation Particle Counters have to be calibrated regularly at least once per year to detect malfunctions such as degradation of the laser diode, temperature instabilities, or internal pollution. CPCs should be only used after determining the flow rate and after a calibration of the detection efficiency curve (see also Wiedensohler et al., 1997) and the absolute detection efficiency. The flow rate should be accurate within 2% and has to be checked on a monthly base.
- The sizing accuracy of mobility size spectrometers have to be verified using 200 nm PSL spheres frequently. Especially for SMPS systems, the correct delay time can only be determined by this calibration. The measured mean particle size should be within 3.5% of the nominal PSL size including uncertainties of the PSL spheres (±2.5%) and the sheath air flow (±1%). The delay time should not be set by matching the PSL size with the nominal size. Generally, the delay time is correct if up- and down-scans show the same result. The use of 200 nm PSL particles is a compromise obtaining a sufficient number concentration and a minimum of residual material on the particles.
- Mobility size spectrometers should also be regularly compared to a reference system for a period of few days once per year (if a reference system is available). This



intercomparison can be done either within an intercomparison workshop or at the sampling site. If a reference spectrometer is not available, also the total number concentration measured by a CPC can be also compared to the number integral of the size distribution. Compare the integral of the number size distribution to the total number concentration measured by a particle counter if no nucleation mode particles are present. Ideally, the difference in number concentrations should be not greater than 10%.

- The background of the system should be also checked every month. Please, connect an absolute particle filter to the system inlet and scan for several size distribution! Ideally, the background should be close to zero.
- DMAs have to be cleaned once per year. CPCs have to undergo a service of an experienced person to clean the saturator and the optics. Please, do not open the bipolar charger! Follow only the instructions of the manufacturer!
- The mobility size spectrometers should be operated in an environment of 15– 30°C to avoid a malfunction of the particle counter.

A2 Description of non-commercial inversion routines

In Table 2, the inversion routines of the different non-commercial mobility size spectrometers are described in detail.

A3 Description of the mobility size spectrometers

²⁰ In Table 3, the technical set up and software of the different non-commercial mobility size spectrometers are described in detail.

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A4 EBAS three-level data structure

A4.1 Level-0

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The level-0 data set contains the metadata of the mobility size spectrometer, all mandatory raw data, and system parameters, as well as optional status parameters.

- Metadata should be included in the header as comment lines describing the mobility size spectrometer (e.g. inlet type, inlet length and diameter, DMA types, DMA dimensions, CPC type, serial numbers, dryer type, etc), operational settings (e.g. aerosol flow and sheath air flow rate), and finally the number of size bins.
 - The mandatory raw data include the measured system parameters and the measured mobility distribution as described above. The data should be flagged according to the flag list as shown in Table 4, if measurements problems were identified.
 - Optional data provide status parameters of the system. These data are meant for a future-orientated online data transfer showing the actual status of the instrument. These status parameters (e.g. standard deviation of the different flow rates, CPC temperatures and flow/liquid status) are necessary and important for an automated quality assurance procedure.

The whole level-0 data structures for single and dual DMA mobility size spectrometers are listed in Table 5a and b, respectively. Data which do not fulfill the requirements must be flagged according to Table 4. False measurements should be removed from the data set and should be flagged as missing measurements.

In a "Read-Me" file belonging to level-0, filtering of raw data should be described according to predefined Quality Control (QC). Alternatively, this can be also described in the comments of the metadata.



Accepted limits for system parameters such as flow rates, and RH

- Aerosol flow rate maximum ±5% (recommendation)
- Sheath air flow rate maximum ±2% (recommendation)

A4.2 Level-1

- ⁵ Data in level-1 contain processed particle number size distributions with the original time resolution of the instrument (Table 6). The entire process includes:
 - Inversion from particle mobility to number size distribution (conversion to dN/dlogDp and multiple charge correction)
 - Correction for CPC counting efficiency
- Correction for particle losses inside the of the mobility size spectrometer (if known)
 - Correction for particle losses from the aerosol inlet to the instrument
 - (optional) correction after comparison against reference mobility size spectrometer

For dual DMA mobility size spectrometer only:

- Assimilation of DMA and UDMA data
 - Removal of redundant size channels in the overlap region of DMA and UDMA

A "Read-Me" file belonging to level-1 data should list details about the transformations and corrections done from raw to processed data. Alternatively, this can also be described in the comments of the metadata.

 For Dual Mobility Spectrometer only: which and how many bins form the overlap between DMA1 and DMA2 were used

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- Description which inversion program with which bipolar charge equilibrium were used
- Description of the correction functions for CPC efficiency, losses inside of the mobility size spectrometer, and losses in transport inlet system
- (optional) Description of the correction function from comparison against a reference mobility size spectrometer
 - Calibration date of the CPC and DMA (in case a measured DMA-transfer function is used in the inversion program)

A4.3 Level-2

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Data in level-2 are the final number size distributions, which were corrected to standard conditions of temperature (273.15 K) and pressure (1013 hPa), and averaged to a time resolution of 1 h (arithmetic mean). These final transformations are according to the standard in the EBAS data base. The data structure is described in Table 7 and consists of median values of the averaged the particle number size distributions as well as their 15.87 and 84.13 percentiles.

As the WMO GAW-WDCA (World Data Centre for Aerosols) is hosted at the EMEP data centre now, a minimum set of metadata parameters are required in the level-2 file header to meet the needs of WDCA. These requirements include station- and instrument metadata for classifying the dataset in the database, e.g. instrument serial number, station GAW-ID, site land use and station settings. It also includes details about the corrections and calculations done.

Details about the corrections and calculations could alternatively be listed in a "readme" file belonging to level-2 data.



The "readme" file should at a minimum include:

- Correction to STP
- Calculations of average and percentile

A5 Constants and relevant equations

The constants and equations follow the recommendations in the ISO15900 standard-5 ization (also given in Kim et al., 2005):

Dynamic gas viscosity at 296.15 K and 101.3 kPa:

$$\eta_0 = 1.83245 \times 10^{-5} \frac{\text{kg}}{\text{ms}}$$
$$\eta = \eta_0 \left(\frac{T}{T_0}\right)^{3/2} \left(\frac{T_0 + 110.4 \text{ K}}{T + 110.4 \text{ K}}\right)$$

Mean free path at 296.15 K and 101.3 kPa: 10

$$\lambda_0 = 67.3 \times 10^{-9} \,\mathrm{m}$$

$$\lambda = \lambda_0 \left(\frac{T}{T_0}\right)^2 \left(\frac{p_0}{\rho}\right) \left(\frac{T_0 + 110.4 \,\mathrm{K}}{T + 110.4 \,\mathrm{K}}\right)$$

Cunningham correction:

$$C_{\rm C} = 1 + \frac{2 \cdot \lambda}{d_{\rm P}} \left(1.165 + 0.483 \cdot \exp\left(-0.997 \frac{d_{\rm P}}{2 \cdot \lambda}\right) \right)$$

Bipolar charge distribution: 15

To calculate the bipolar charge distribution analytically, an approximation formula for lower charging states, n, (-2, -1, +1, +2) was developed (Wiedensohler, 1988). This formula is valid for particle size ranges 1 to 1000 nm or 20 to 1000 nm particle



diameter for *n* equal to -1, 0, +1 or -2, +2, respectively. The according approximation coefficients are given in Table 8.

Approximation formula:

$$F(n) = 10^{\left(\sum_{i=0}^{5} a_i(n) \left(\log \frac{D_P}{nm}\right)^i\right)}$$

⁵ For higher *n* (+3, -3, +4, -4 etc.), the Gunn formula below can be used. A ratio of the electrical mobility of positive to negative ions Z_{I+}/Z_{I-} of 1.4/1.6 was suggested in Wiedensohler (1988).

Gunn (1956) equation:

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$$F(n) = \frac{e}{\sqrt{4\pi^2 \cdot \varepsilon_0 \cdot D_{\mathsf{P}} \cdot k \cdot T}} \cdot \exp\left(-\frac{\left(n - \left(\frac{2\pi \cdot \varepsilon_0 \cdot D_{\mathsf{P}} \cdot k \cdot T}{e^2}\right) \ln \frac{Z_{\mathsf{I}_+}}{Z_{\mathsf{I}_-}}\right)^2}{\left(\frac{4\pi \cdot \varepsilon_0 \cdot D_{\mathsf{P}} \cdot k \cdot T}{e^2}\right)}\right)$$

A6 EUSAAR raw data format for commercial mobility spectrometers

A6.1 EUSAAR format: TSI-SMPS raw data

TSI-AIM raw data are particle counts versus time. The raw data are exported by AIM as shown below. The export also contains all parameters needed for all further calculations. The raw data time listed in this table is elapsed time since the start of the scan, the particle counts have been detected by the CPC at this elapsed time. As can be

the particle counts have been detected by the CPC at this elapsed time. As can be seen, the CPC is read 10 times per second.

The raw data export table also lists the particle diameter for singly charged particles $(D^{1+}(t_{e,i}))$ which corresponds to the elapsed time $(t_{e,i})$. This diameter can be used to show raw counts versus particle size. The raw particle counts can either been accumulated in the time interval from $t_{e,i-1}$ to $t_{e,i}$ or in the diameter interval from $D^{1+}(t_{e,i-1})$ to $D^{1+}(t_{e,i})$. Knowing the effective aerosol flow rate of the condensation particle counter, the number can be converted to a number concentration. The mid particle diameter is

determined the geometric mean of the lower $D^{1+}(t_{e,i-1})$ and upper diameter $D^{1+}(t_{e,i})$ of the chosen channel width.

A6.2 EUSAAR format: Grimm-SMPS raw data GRIMM data formats for mobility size distributions

⁵ The Grimm mobility size spectrometers record the measured raw mobility distribution according to the EUSAAR Level-0 data format indicating as particle number concentration for each particle diameter bin.

A7 Abbreviation list

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In Table 9, the abbreviations for the institute or universities are listed, which participated in the intercomparison of inversion routines and instruments.

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Table 1. Equivalent lengths of a straight pipe for calculating the losses by diffusion for different parts of mobility size spectrometers.

Device	Equivalent pipe length
Hauke-type medium DMA (28 cm effective length) TSI long DMA (44.4 cm effective length) TSI nano DMA (4.987 cm effective length) SS24" Permapure Nafion dryer SS12" Permapure Nafion dryer Topas diffusion dryer Other diffusion dryers Bipolar charger	4.6 m (Karlsson et al., 2003) 7.1 m (Karlsson et al., 2003) 3.64 m (Jiang et al., 2011) 2.5 m (Dick et al., 1995) 1.25 m (Dick et al., 1995) 5 m (estimate) Unknown (to be estimated) 1 m (Covert et al., 1997)



Table 2. Descriptions of the different non-commercial inversion routines for mobility size spectrometer measurements.

	Input	Calculation
IFT (Stratmann and Wieden- sohler, 1996) NILU (Fiebig et al.,	- Number concentration versus mobility diameter calculated from count number, counting time,	 dN/dlogZp using the real area of the DMA trans- fer function (Note: Zp is the electrical mobility)
	and CPC flow rate (no certain distance between mobility bins required)	 Charge correction (Wiedensohler, 1988)
1996)	 Selection: DMA type for real transfer function 	- Conversion Zp to Dp (mobility or volume equiva-
NILU (Fiebig et al., 2005)	 Selection: Positive or Negative* charge 	
	 Selection: CPC efficiency correction Yes or No* 	 Conversion dividiog2p to dividiog2p (mobility of volume equivalent diameter)
	 Consider additional input data out of range of the instrument for charge correction 	 Split and interpolate in discrete logarithmic equidistant mobility or volume equivalent diam-
	 Consider of simple dynamic shape factor along the whole range (static or dynamic) 	eter bins
NILU (Fiebig et al., 2005)	 Particle number concentration (incl. optional un- certainty) as function of mobility particle diameter (incl. optional uncertainty), sample and sheath 	 Total DMA counting efficiency calculated on- line from input parameters by convolution of: – Stolzenburg (1988) DMA transfer function.
	air flows (incl. uncertainty), DMA internal tem- perature and pressure.	 Charge correction Wiedensohler (1988)
	 DMA inner and outer radius, DMA length 	 Baron and Willeke (2005) diffusion losses
	 Polarity of high-voltage 	 Problem formulated as underdetermined, ill- paged vector equation by dispertising in particle
	 CPC counting efficiency as function of particle di- ameter 	size independently of number of measured size bins (finer discretizing than measured size bins
	 Diffusion losses as function of particle diameter 	 Problem solved by Singular Value Decompo- sition on rank and least-squares with inequal- ity constraints on null space, solution is non- negative and smooth
		 No initial guess, no smoothing of transfer function or input data
		 May provide uncertainty of result, either by Gaussian or Monte Carlo error propagation
		 Cross-instrument inversions of CPC cascades, CPCs with diffusion screens, DMPS/SMPS, OPCs

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	Input	Calculation
JHEL/FMI	 DMA voltage versus concentration DMA dimensions DMA flow rates DMA voltage polarity DMA pressure and temperature Function for particle losses inside the DMA Function for the detection efficiency of the CPC 	 Inversion method choices are pseudo inversion, nnls and tikhonov Charge correction (Wiedensohler, 1988) DMA transfer function by Stolzenburg (1988)
νSI	 CPC count rate and DMA voltage as a function of time, sheath and aerosol flow rate through DMA, flow rate through detection volume of CPC Selection**: DMA type for ideal transfer function (TSI short, TSI long, Hauke short, Hauke long, TSI modified short, TSI modified long*) Selection: Positive* or Negative charge Selection: CPC efficiency correction Yes or No* Selection: resolution of diameter grid for inverted size distribution (log-even spaced, 2ⁿ bins per decade, integer "n" can be selected). ** Note: other DMA types and/or real transfer function properties can be added if data are available 	 Convert CPC counts into concentrations behind DMA Calculate equivalent constant voltage corre- sponding to each CPC reading Calculate diameters corresponding to voltages Correct raw measurements with CPC detection efficiency Invert measurement, i.e. distribute raw measure- ments to regular diameter grid and correct with DMA transfer probability Apply correction according to selection

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	Input	Calculation
ULUND (Zhou, 2001)	 Number concentration versus mobility diameter calculated from count number, counting time, and CPC flow rate 	 – dN/dlogZp using the ideal width of the DMA transfer function and area corrected for transmis- sion losses
	 Selection: DMA type for real transfer function (Ideal width, are adjusted for transmission effi- ciency Hauke short Hauke medium*) 	 Spline through dN/dlogZp data points Split in discrete logarithmic equidistant mobility
	 Negative charge 	bins (adjustable factor between mobility bins) – Charge correction (Wiedensohler, 1988)
	 Cubic spline 	 Conversion Zp to Dp
	 CPC counting efficiency as function of size 	 Spline through dN/dlogDp
	 Sampling line losses 	- Split in discrete logarithmic equidistant mobility
	 Bipolar charging probability 	diameter bins
ISAC	 Voltages versus number concentration calcu- lated from count number counting time, and CPC 	 dN/dlogZp using the real area of the DMA trans- fer function
	flow rate (No certain distance between mobility bins required)	 Spline through dN/dlogZp data points
	 Selection: DMA type for real transfer function (Ideal, Hauke short, Hauke medium*) 	 Split in discrete logarithmic equidistant mobility bins (factor 1.1 between mobility bins)
	- Selection: Positive or Negative* charge	 Charge correction (Wiedensohler, 1988)
	 Selection: Cubic or Linear spline 	 Conversion Zp to Dp
	 Selection: CPC efficiency correction Yes or No* 	 Spline through dN/dlogDp
	- * Selected for the instrument intercomparison	 Split in discrete logarithmic equidistant mobility diameter bins
LAMP	 Number concentration as a function of electrical mobility 	- Charge correction (Wiedensohler, 1988)
	- DMA flow rates	 Linear inversion Ideal transfer function
	- DMA polarity	 Integrate between set mobility's transfer func- tions
		Convert to dN/dlogDn
	- iemperature, riessure	

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	Input	Calculation
JRC	- Number concentration versus mobility diameter	 dN/dlogZp using the ideal DMA transfer function
	calculated from count number, counting time, and CPC flow rate (no fixed distance between mobility bins required)	 Use of discrete logarithmic equidistant mobility bins (factor 1.1 between mobility bins)
	 Ideal DMA transfer function 	- Charge correction (Wiedensohler, 1988)
	- Negative charge	 Conversion Zp to Dp
	 Linear spline 	 Linear Spline through dN/dlogDp
	 CPC efficiency correction: No 	
UMN	 Number concentration versus mobility diameter 	- Bipolar charging fractions: approximation by
	 Aerosol and sheath flow rates 	Wiedenschler (1988) or direct estimation using
	 DMA temperature and pressure 	Frick, 1986)
	 DMA voltage polarity 	- Diffusing or non-diffusing DMA transfer function
	 CPC counting efficiency 	(Stolzenburg and McMurry, 2008). An equivalent
	 Parameters for loss estimation: flow rates; actual and/or equivalent lengths in the sampling system 	efficiency through DMA inlet and outlet. Trans- fer function and penetration were experimentally determined
		 Diffusion losses through the sampling system
		 Linear inversion method with multiple charging correction was used to obtain dN/dlogDp
PKU	- Number concentration versus mobility diameter	- Iterantiv calculation for dN/dlogDp to fit the mea-
	 Sheath to sample flow ratio 	sured number concentration using the triangle
	 Default temperature (20 °C) and pressure (1013.25 hPa) is used, if not temperature and pressure input 	(Wiedenschler, 1988), assuming dN/dlogDp can be linear fitted in the scale of logDp



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	Input	Calculation
TNO	 Number concentration versus mobility diameter calculated from count number, counting time, 	 – dN/dlogZp using the real area of the DMA trans- fer function (Note: Zp is the electrical mobility)
	and CPC flow rate (No certain distance between mobility bins required)	- Charge correction (Wiedensohler, 1988)
	- Selection: DMA type for real transfer function	 Conversion Zp to Dp (mobility or volume equiva- lent diameter)
	 Selection: Positive charge 	 Conversion dN/dlogZp to dN/dlogDp (mobility or volume equivalent diameter)
		 Split and interpolate in discrete logarithmic equidistant mobility or volume equivalent diam- eter bins
UBIR	- TSI AIM Software Version 8.1	- TSI AIM Software Version 8.1



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Table 3. Description of the mobility size spectrometers used in the third intercomparison workshop.

Description IFT-SMPS Range: 10-800 nm (IFT-REF1. DMA: Hauke-type; inner diameter 50 mm, outer diameter 67 mm, length 280 mm Bipolar charger: 85Kr IFT-REF2. CPC: TSI model 3010 IFT-CH UBA-NG Software: IfT scanning program LB) Other hardware: aerosol and sheath air Nafion dryers; aerosol and sheath air T and RH sensors, blower for closed loop, heat exchanger for closed loops, power supply for positive HV, mass flow meter for sheath air flow, Δp capillary flow meter for aerosol flow Range 3-800 nm IFT-TDMPS DMA: Hauke-type; inner diameter 50 mm, outer diameter 67 mm, length 280 mm UDMA (Ultrafine DMA): Hauke-type; inner diameter 50 mm, outer diameter 67 mm, length 110 mm Bipolar charger: ⁸⁵Kr CPC: TSI model 3010 UCPC (Ultrafine CPC): TSI model 3025 Software: IfT stepping program Other hardware: aerosol and sheath air flows Nafion dryers; aerosol and sheath air T and RH sensors, blowers for closed loops, heat exchangers for closed loops, FUG power supplies for positive HV, Δp capillary flow meters for aerosol and sheath air flows NILU-DMPS Range: 10-550 nm DMA: Hauke-type, inner diameter 50 mm, outer diameter 67 mm, length 285 mm Bipolar charger: ⁶³Ni CPC: TSI 3010, running with 25K temperature difference between saturator and condenser Software: NILU DMPS software, target diameters specified, voltages calculated online from flows, temperature, pressure, uses EUSAAR file formats for data storage. Other hardware: closed loop sheath air flow controlled by mass flow controller, particle filters immediately at DMA sheath in and sheath out, heat exchanger between sheath pump out and sheath DMA in; p, T, and RH monitored in sample and sheath flow; aerosol flow: two 12" Nafion dryers; positive voltage FUG HV supply



Description UHFI -Range: 6-800 nm DMPS DMA: Hauke-type length 280 mm Bipolar charger: ¹⁴C CPC: TSI3772 Other hardware: Aerosol flow dried with Nafion drier, aerosol flow temperature and RH probe, Δp capillary flow meter for aerosol flow, sheath flow blower with volumetric flow meter in control loop, closed loop sheath flow configuration with heat exchanger, sheath flow pressure and temperature sensors, positive Spellman HV source 0-10 kV. possibility to switch between two DMA aerosol flow rates and virtually any sheath flow rate PSI-SMPS Range: 10-450 nm DMA: TSI modified-type; inner diameter 9.37 mm, outer diameter 19.61 mm, length 437 mm Bipolar charger: ⁸⁵Kr CPC: TSI model 3772 Software: PSI scanning program Other hardware: aerosol flow: Nafion dryer; sheath air T and RH sensors, pump for closed loop, heat exchanger for closed loops, FUG power supply for negative HV, mass flow meter for sheath air flow Range: 7-500 nm FMI-TDMPS DMA1: Hauke-type length 109 mm DMA2: Hauke-type length 280 mm Bipolar charger: ⁸⁵Kr CPC1: TSI3772 CPC2: TSI3772 Software: Custom-made Python code Other hardware: Aerosol flow dried with Nafion drier, Δp capillary flow meters for aerosol flow (x2), sheath flow blower with volumetric flow meter in control loop (x2), sheath flow in closed loop controlled separately from the measurement program (x2), sheath flow pressure and temperature sensors (x2), FUG positive power supply (up to 12500 and 3000 V)



	Description
JRC-DMPS	Range: 10–800 nm DMA: Hauke-type; inner diameter 50 mm, outer diameter 67 mm, length 280 mm Bipolar charger: ⁸⁵ Kr
	Software: stepping program from IFT, extensively modified at JRC for instrument control
	Other hardware: aerosol flow with Nafion dryer; sheath air flows with Nafion dryer; aerosol and sheath air T and RH sensors, blower for closed loop, heat exchanger for closed loops, FUG power supply for positive HV, mass flow meter for sheath air flow, Δp capillary for aerosol flow
LAMP-	Range: 10–450 nm
DMPS	DMA: inner radius 9.4 mm; outer radius 19.6 mm, length 444 mm
	Bipolar charger: "Ni
	CPC: TST model 3010 Software: Jahview, made by LAMP
	Other hardware: aerosol flows: diffusion dryer; sheath flow silica gel dryer, aerosol and sheath air T and RH sensors, blower closed loop, internal electronic box, power supplies for positive HV, Δp capillary for aerosol air flow, mass flow meter for sheath air flow
NUIG-	Range: 20–500 nm
DMPS	DMA: TSI model 3081
	Bipolar charger: TSI model 3077 ⁵⁵ Kr
	CPC: TSI Model 3010 Software: Jabview, made by University of Galway
	Other hardware: aerosol flows: Nafion dryer, aerosol and sheath air T and RH sensors, diaphragm pump with critical orifice for closed loop, heat exchanger, Bertram power supplies for positive HV, Δp capillary for aerosol air flow, mass flow meter for sheath and excess air flow. Electronic box made by University of Galway



			71. •••••••••••	0011
	Description	SCU		
ISAC-DMPS	Range: 10–500 nm	SS.		
	DMA: inner radius 25 mm; outer radius 33.5 mm, length 280 mm	on	Title	Pag
	Bipolar charger: ⁶³ Ni	σ		
	CPC: TSI model 3010	ap	Abstract	In
	Software: visual basic, made by University of Kuopio	Ē		
	Other hardware: aerosol flows: Nafion dryer; sheath flow silica gel dryer, aerosol		Conclusions	R
	and sheath air T and RH sensors, vacuum pump with critical orifice for closed loop,		Conclusions	
	heat exchanger for closed loop, FUG power supplies for positive HV, Δp capillaries	_	Tablaa	
	for aerosol air flow, TSI 4140 flow meter for sheath air flow		Tables	
TNO-SMPS	TSI SMPS 3034 with internal CPC	C		
	Bipolar charger: TSI model 3077 ⁸⁵ Kr	No.	14	
	IFT scanning software was used instead of the TSI-supplied software	<u> </u>		
UBIR-SMPS	TSI SMPS 3936			
	DMA: 3081	2		
	CPC: 3775	pe		
	Bipolar charger: TSI model 3077 ⁸⁵ Kr	-	Back	
	Other hardware: aerosol air flow: Nafion dryer			
			Full Scr	een /



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Table 4. Flag list for data of mobility size spectrometer measurements according to the EMEP flagging description.

Flag	Original description/commentary – EMEP data base	Comments especially for mobility spectrometers
Missir	ng data	
999	MMU I	Instrument not operational
	Missing measurement, unspecified reason	False measurements
980	MZSI	E.g. zero check with total particle filter or high volt-
	Missing due to calibration or zero check	age supply
Mech	anical problems	
699	LMUT	E.g. problems with flow, leaks, or HV supply
	Mechanical problem, unspecified reason	3 F
662	LFV V	E.g. aerosol flow or sheath flow rate out of range but
	Too high sampling flow, data considered valid	considered as valid
652	LCN V	Disturbance by other laboratory activity
	Construction/activity nearby	
Extre	me or inconsistent values	
499	INU V	E.g. inconsistency with total particle counter reading
	Inconsistent with another unspecified measurement	3
459	EUE I	Unexplained extreme values, technical problem is
	Extreme value, unspecified error	suspected
410	SDE V	
	Sahara dust event	
Flags	for aggregated data sets	
394	DC9 V	
	Data completeness less than 90%	
392	DC7 V	
	Data completeness less than 70%	
390	DC5 V	
	Data completeness less than 50%	
Excer	otion flags for accepted, irregular data	
189	LCS V	
	possible local contamination indicated by wind from contami-	
	nated sector (auto)	
188	LCW V	
	possible local contamination indicated by low wind speed	
	(auto)	
187	LCP V	
	possible local contamination indicated by occurrence of new	
	particles (auto)	
187	LCAV	
	possible local contamination indicated by single scattering	
	albedo (auto)	

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Table 5a. Level-0 data structure for single DMA mobility spectrometers including mandatory and optional data and system parameters.

mandatory

- Column 01 Start date (Day of Year/UTC starting 1 Jan, 00:00 UTC with 0.00)
- Column 02 End date (Day of Year/UTC starting 1 Jan, 00:00 UTC with 0.00)
- Column 03 Start Year
- Column 04 End Year
- Column 05 Internal system temperature (K)
- Column 06 Internal system pressure (hPa)
- Column 07 Flow rate aerosol flow rate (I/min) at DMA temperature and pressure
- Column 08 Flow rate sheath air flow rate(I/min) at DMA temperature and pressure
- Column 09 Relative humidity aerosol flow (%)
- Column 10 Relative humidity sheath air (%)
- Column 11 Number of particle size bins
- Column 12 Size Bin 01 median particle mobility diameter for singly charged (in μ m)
- Column ... /continue with all particle size bins
- Column ... Size Bin 01 (raw number concentrations in cm⁻³)
- Column ... /continue raw concentration for all particle size bins

optional

- Column ... Size Bin 01 Standard deviation median particle mobility diameter (in $\mu m)$
- Column ... /continue with all particle size bins
- Column ... CPC saturator temperature (K)
- Column ... CPC condenser temperature (K)
- Column ... Standard deviation aerosol flow rate(I/min)
- Column ... Standard deviation sheath air flow rate (I/min)
- Column ... CPC flow status, 0 ok, 1 flow error
- Column ... CPC liquid status, 0 ok, 1 liquid error
- $Column \ldots Sheath air status, 0-ok, 1-flow not critical/low/high$

mandatory

Column ... Numflag (data exist/don't exist/flagged \rightarrow 0/999/nnn)



Table 5b. Level-0 data structure for dual DMA mobility spectrometers including mandatory and optional data and system parameters.

mandatory	1
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Column 01 Column 02 Column 03 Column 04 Column 05 Column 06 Column 07 Column 08 Column 09 Column 10 Column 11 Column 12 Column 13 Column 14 Column 15 Column 16 Column Column	Start date (Julian Day/UTC – starting 1 Jan, 00:00 UTC with 0.00) End date (Julian Day/UTC – starting 1 Jan, 00:00 UTC with 0.00) Start Year End Year DMA temperature (K) DMA pressure (hPa) Flow rate – aerosol flow rate (l/min) – DMA1 Flow rate – sheath air flow rate(l/min) – DMA1 Flow rate – aerosol flow rate (l/min) – DMA2 Flow rate – sheath air flow rate (l/min) – DMA2 Flow rate – sheath air flow rate (l/min) – DMA2 Flow rate – sheath air flow rate (l/min) – DMA2 Relative humidity – aerosol flow (%) – DMA1 Relative humidity – sheath air (%) – DMA1 Relative humidity – sheath air (%) – DMA2 Relative humidity – sheath air (%) – DMA2 Number of size bins Size Bin 01 median particle mobility diameter for singly charged (in μm) … /continue with all particle size bins
optional	
Column Column Column Column Column Column Column Column Column Column Column Column Column Column Column Column	Size Bin 01 Standard deviation – median particle diameter /continue for all particle size bins Standard deviation – aerosol flow rate (l/min) – DMA1 Standard deviation – sheath air flow rate (l/min) – DMA1 Standard deviation – aerosol flow rate (l/min) – DMA2 Standard deviation – sheath air flow rate (l/min) – DMA2 Standard deviation – sheath air flow rate (l/min) – DMA2 CPC saturator temperature (K) CPC condenser temperature (K) CPC1 flow status, 0 – ok, 1 – flow error CPC2 liquid status, 0 – ok, 1 – liquid error CPC2 liquid status, 0 – ok, 1 – liquid error Sheath flow status DMA1, 0 – ok, 1 – flow not critical/low/high Sheath flow status DMA2, 0 – ok, 1 – flow not critical/low/high
mandatory	
• •	

Column ... Numflag (data exist/don't exist/flagged \rightarrow 0/999/nnn)



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Table 6. Level-1 processed particle number size distribution with the original time resolution.Midpoint bin particle mobility diameter is given in the title of the column.

Column 01	Start date (Julian Day/UTC - starting 1 Jan, 00:00 UTC with 0.00)
Column 02	End date (Julian Day/UTC – starting 1 Jan, 00:00 UTC with 0.00)
Column 03	Start Year
Column 04	End Year
Column 05	Internal system temperature (K)
Column 06	Internal system pressure (hPa)
Column 07	Number of particle size bins
Column 08*	Size Bin 01 (dN/dlogDp in cm ⁻³)
	/continue for all particle size bins
Column	Numflag (data exist/don't exist/flagged \rightarrow 0/999/nnn)

 * zero values are replaced by a very small value such as $10^{-5}.$

Table 7. Level-2 processed particle number size distribution with the one hour time average at standard temperature and pressure. Bin particle mobility size is given in the title of the column.

Column 01 Column 02	Start date (Julian Day/UTC – starting 1 Jan, 00:00 UTC with 0.00) End date (Julian Day/UTC – starting 1 Jan, 00:00 UTC with 0.00)
Column 03	Start Year
Column 04	End Year
Column 05	Internal system temperature (K)
Column 06	Internal system pressure (hPa)
Column 07	Number of particle size bins
Column 08* Column	Particle size Bin 01 arithmetic mean (dN/dlogDp in cm ⁻³)*/continue for all particle size bins
Column	Particle size Bin 01 15.87 percentile (cm ⁻³)*
Column	/continue for all particle size bins
Column … Column … Column …	Particle size Bin 01 dN/dlogDp 84.13 percentile $(cm^{-3})^*$ /continue for all particle size bins Numflag (data exist/don't exist/flagged \rightarrow 0/999/nnn)

* zero values are replaced by a very small value such as 10^{-5} .



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Table 8. Approximation coefficients for the formula describing the bipolar charge distribution for the charging states n (-2, -1, 0, +1, +2).

		Approxima	ation coeffic	eients $a_i(n)$	
i	<i>n</i> =–2	<i>n</i> =-1	<i>n</i> =0	<i>n</i> =+1	<i>n</i> =+2
0	-26.3328	-2.3197	-0.0003	-2.3484	-44.4756
1	35.9044	0.6175	-0.1014	0.6044	79.3772
2	-21.4608	0.6201	0.3073	0.4800	-62.8900
3	7.0867	-0.1105	-0.3372	0.0013	26.4492
4	-1.3088	-0.1260	0.1023	-0.1553	-5.7480
5	0.1051	0.0297	-0.0105	0.0320	0.5049

Table 9. Abbreviations of institutes and universities.

AbbreviationInstitute, University, Public AuthorityIFTLeibniz Institute for Tropospheric ResearchNILUNorwegian Institute for Air ResearchFMIFinnish Meteorological InstituteLAMPLaboratoire de Météorologie PhysiqueUHELUniversity of HelsinkiULUNDUniversity of LundPSIPaul Scherrer InstituteJRCJoint Research CentreUBIRUniversity of BirminghamNUIGNational University of IrelandISACInstitute of Atmospheric Sciences and ClimateTNOTNO Built Environment and GeosciencesUMNUniversity of MinnesotaPKUPeking UniversityUBA-NGUmweltbundesamt, Instrument NeuglobsowLBProvince Limburg		
IFTLeibniz Institute for Tropospheric ResearchNILUNorwegian Institute for Air ResearchFMIFinnish Meteorological InstituteLAMPLaboratoire de Météorologie PhysiqueUHELUniversity of HelsinkiULUNDUniversity of LundPSIPaul Scherrer InstituteJRCJoint Research CentreUBIRUniversity of BirminghamNUIGNational University of IrelandISACInstitute of Atmospheric Sciences and ClimateTNOTNO Built Environment and GeosciencesUMNUniversity of MinnesotaPKUPeking UniversityUBA-NGUmweltbundesamt, Instrument NeuglobsowLBProvince Limburg	Abbreviation	Institute, University, Public Authority
NILUNorwegian Institute for Air ResearchFMIFinnish Meteorological InstituteLAMPLaboratoire de Météorologie PhysiqueUHELUniversity of HelsinkiULUNDUniversity of LundPSIPaul Scherrer InstituteJRCJoint Research CentreUBIRUniversity of BirminghamNUIGNational University of IrelandISACInstitute of Atmospheric Sciences and ClimateTNOTNO Built Environment and GeosciencesUMNUniversity of MinnesotaPKUPeking UniversityUBA-NGUmweltbundesamt, Instrument NeuglobsowLBProvince Limburg	IFT	Leibniz Institute for Tropospheric Research
FMIFinnish Meteorological InstituteLAMPLaboratoire de Météorologie PhysiqueUHELUniversity of HelsinkiULUNDUniversity of LundPSIPaul Scherrer InstituteJRCJoint Research CentreUBIRUniversity of BirminghamNUIGNational University of IrelandISACInstitute of Atmospheric Sciences and ClimateTNOTNO Built Environment and GeosciencesUMNUniversity of MinnesotaPKUPeking UniversityUBA-NGUmweltbundesamt, Instrument NeuglobsowLBProvince Limburg	NILU	Norwegian Institute for Air Research
LAMPLaboratoire de Météorologie PhysiqueUHELUniversity of HelsinkiULUNDUniversity of LundPSIPaul Scherrer InstituteJRCJoint Research CentreUBIRUniversity of BirminghamNUIGNational University of IrelandISACInstitute of Atmospheric Sciences and ClimateTNOTNO Built Environment and GeosciencesUMNUniversity of MinnesotaPKUPeking UniversityUBA-NGUmweltbundesamt, Instrument NeuglobsowLBProvince Limburg	FMI	Finnish Meteorological Institute
UHELUniversity of HelsinkiULUNDUniversity of LundPSIPaul Scherrer InstituteJRCJoint Research CentreUBIRUniversity of BirminghamNUIGNational University of IrelandISACInstitute of Atmospheric Sciences and ClimateTNOTNO Built Environment and GeosciencesUMNUniversity of MinnesotaPKUPeking UniversityUBA-NGUmweltbundesamt, Instrument NeuglobsowLBProvince Limburg	LAMP	Laboratoire de Météorologie Physique
ULUNDUniversity of LundPSIPaul Scherrer InstituteJRCJoint Research CentreUBIRUniversity of BirminghamNUIGNational University of IrelandISACInstitute of Atmospheric Sciences and ClimateTNOTNO Built Environment and GeosciencesUMNUniversity of MinnesotaPKUPeking UniversityUBA-NGUmweltbundesamt, Instrument NeuglobsowLBProvince Limburg	UHEL	University of Helsinki
PSIPaul Scherrer InstituteJRCJoint Research CentreUBIRUniversity of BirminghamNUIGNational University of IrelandISACInstitute of Atmospheric Sciences and ClimateTNOTNO Built Environment and GeosciencesUMNUniversity of MinnesotaPKUPeking UniversityUBA-NGUmweltbundesamt, Instrument NeuglobsowLBProvince Limburg	ULUND	University of Lund
JRCJoint Research CentreUBIRUniversity of BirminghamNUIGNational University of IrelandISACInstitute of Atmospheric Sciences and ClimateTNOTNO Built Environment and GeosciencesUMNUniversity of MinnesotaPKUPeking UniversityUBA-NGUmweltbundesamt, Instrument NeuglobsowLBProvince Limburg	PSI	Paul Scherrer Institute
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NUIGNational University of IrelandISACInstitute of Atmospheric Sciences and ClimateTNOTNO Built Environment and GeosciencesUMNUniversity of MinnesotaPKUPeking UniversityUBA-NGUmweltbundesamt, Instrument NeuglobsowLBProvince Limburg	UBIR	University of Birmingham
ISACInstitute of Atmospheric Sciences and ClimateTNOTNO Built Environment and GeosciencesUMNUniversity of MinnesotaPKUPeking UniversityUBA-NGUmweltbundesamt, Instrument NeuglobsowLBProvince Limburg	NUIG	National University of Ireland
TNOTNO Built Environment and GeosciencesUMNUniversity of MinnesotaPKUPeking UniversityUBA-NGUmweltbundesamt, Instrument NeuglobsowLBProvince Limburg	ISAC	Institute of Atmospheric Sciences and Climate
UMNUniversity of MinnesotaPKUPeking UniversityUBA-NGUmweltbundesamt, Instrument NeuglobsowLBProvince Limburg	TNO	TNO Built Environment and Geosciences
PKUPeking UniversityUBA-NGUmweltbundesamt, Instrument NeuglobsowLBProvince Limburg	UMN	University of Minnesota
UBA-NG Umweltbundesamt, Instrument Neuglobsow LB Province Limburg	PKU	Peking University
LB Province Limburg	UBA-NG	Umweltbundesamt, Instrument Neuglobsow
	LB	Province Limburg



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Fig. 1. Schematic sketch of the recommended closed-loop-based mobility size spectrometer. The set-up includes dryers for aerosol flow and sheath air, heat exchanger, particle filters, and sensors for aerosol and sheath air flow rate, relative humidity and temperature of aerosol flow and sheath air, and absolute pressure.



Fig. 2. Comparison of the inversion routine of different custom-programmed and commercial mobility spectrometers using an ideal transfer function and no correction for particle losses due to diffusion. In the upper graphs of **(a)** and **(b)**, the shaded areas mark the $\pm 5\%$ range around the output of the IfT inversion. The lower graphs show the ratio the different inversion outputs to the IfT inversion.











Fig. 4. Latex sphere (200 nm) calibration of the modified mobility size spectrometer to determine the sizing accuracy under laboratory conditions and supervised by experienced users. The size distributions are normalized to the peak concentration of the reference mobility size spectrometer. The 3.5% uncertainty range includes the uncertainties of the latex particles from the nominal size (2.5%) and the sheath air flow rate (1.0%). The ratios given in the legend are aerosol to sheath air flow rate.























Fig. 8. Intercomparison of the volume size distribution of the mobility size spectrometers with ambient aerosol. The shaded area marks the $\pm 10\%$ range around the reference spectrometer.





Fig. 9. Intercomparison of the particle number size distribution of five identical mobility size spectrometers (custom-built by IfT according to the specifications given in Sect. 3 and Fig. 1) for urban aerosol. The shaded area marks the $\pm 10\%$ range around the average of spectrometers.

