

Symmetric Inclined Grid Mobility Analyzer

SIGMA

REDUCED MANUAL for the Instrument No. 1

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FOREWORD

The Symmetric Inclined Grid Mobility Analyzer SIGMA (Tammet, 2011) is an instrument for atmospheric aerosol nucleation research with special attention to long-term routine measurements in natural atmospheric conditions. It was elaborated using the experience of the preceding scanning mobility analyzers IGMA and BSMA. The range of electric mobility is 0.032–3.2 cm²V⁻¹s⁻¹. The particle diameter distribution in the range of 0.4–7.5 nm is resolved considering the size-mobility correlation at air temperature and pressure measured with built-in sensors. The scanning technique ensures that possible peculiarities in the recorded size or mobility distribution are not caused by the technical troubles of individual mobility channels. High rate of air flow and isopotential principle suppress the disturbing effect of external electric fields and assure the representative sampling of air ions. The sheath air is coming from the atmosphere together with the sample air and is deionized by means of electrostatic filters. The residence time of the air in the instrument is less than 0.2 s and the temperature change does not exceed 0.5 K. The positive and negative particles are sampled from the same inlet air flow and measured exactly simultaneously. The sensitivity of the instrument allows measuring the mobility fraction concentrations of charged fine nanometer particles in atmospheric air at a standard 5-minute time resolution with random errors of about 1 cm⁻³.

The present manual is long and requires a lot of time to learn everything. However, different users of the instrument may learn only limited information:

- People who are responsible for the installing and maintenance of the instrument need information from Chapters 2–4 and Appendices 4, 6–9.
- People attending to routine measurements should read Section 2.3, be well familiar with Chapter 3, Sections 4.1–4.2, and read the first three sections of Appendix 4.
- People processing the recorded data can acquire main information from Chapters 5 and 6, and additional information from Appendices.
- People supervising the measurements and interpreting the recorded data can skip only Appendices 7 and 8 and should pay special attention to the first chapters.

The present manual is compiled considering the Instrument No. 1 manufactured for the University of Tartu. The principles of SIGMA are universal but some technical issues and calibration information will be different in case of the newer instruments. Fresh information about SIGMA and recent revisions of documents and software are available in SIGMA website <http://ael.physic.ut.ee/tammet/sigma/>.

1. INTRODUCTION

1.1. Air ions and nanometer aerosol particles

Charged molecular clusters and nanometer particles act as carriers of the electric current in the air. Therefore, they are called *air ions* in many documents including the present manual. Hörrak et al. (2000) carried out long-term measurements of natural air ion mobility distribution in the rural air. The dataset was analyzed using the method of principal components, producing a classification of atmospheric ions according to their mobility and size in such a way that the variation of the values of the size distribution function is well correlated inside the classes and ill correlated between the classes. The principal component classification is in good accordance with the earlier intuitive classification of air ions (Israël, 1970) and the classes are called the cluster ions (diameters 0.4–1.6 nm), intermediate ions (charged fine nanometer particles of the diameter of 1.6–7.4 nm) and large ions (charged aerosol particles of the diameter above 7.4 nm). The physical background of the 1.6 nm threshold is the transition from elastic collisions with gas molecules characteristic to the electron structure of a molecular ion to the inelastic collisions characteristic to the condensed matter electron structure of aerosol particles (Tammet, 1995). The physical background of the empiric 7.4 nm correlation threshold has no definite theoretical explanation.

The size range of an instrument for applications in atmospheric aerosol nucleation studies must include cluster ions and intermediate ions up to the diameter of at least 7.4 nm. The nonlinear character of the size-mobility relation makes the relative range of ion mobility wider than the size range. As a minimum, it should cover two magnitudes of mobility. The size range of the SIGMA 0.4–7.5 nm and the mobility range of 0.032–3.2 cm²V⁻¹s⁻¹ cover the region of cluster ions and charged small nanometer particles.

1.2. Measuring methods and special requirements

Traditional methods of measuring ions in atmospheric air were reviewed from the viewpoint of atmospheric electricity by Israël (1970) and Tammet (1970), and from the viewpoint of aerosol science by Flagan (1998). Three methods of mobility spectrometry are dominating in atmospheric aerosol nucleation studies: single-channel method with a CPC-detector (traditional DMAS, see Flagan 1998), multichannel method (Tammet et al., 1973; Mirme et al., 2007), and single channel method with an internal electrometric detector e.g. the BSMA (Tammet, 2006).

Traditional DMA systems equipped with CPC-detectors are not appropriate for the measuring of cluster ions. The calibration of CPC-s below 3 nm is complicated and can include large systematic errors. Thus, the electrometric detectors of ions are preferred when the clusters should be included into the size range.

Multichannel instruments have some known advantages. The measuring information is collected simultaneously with many electrometers and the full distribution is measured as fast as the signal from a single channel. A weak point is that in this case it is difficult to identify the events where signals of one or a few channels include a moderate systematic error.

However, the main factor limiting the usage of multichannel instruments is their complicated construction and calibration, bringing about a high price and complex maintenance. Thus the single-channel scanning may appear to be a good alternative method in many situations.

Measurement of intermediate ions in the natural atmospheric air is a hard challenge for any instrument. The charging probability of a neutral-born nanometer particle is very low and the concentration of fine intermediate ions is often less than ten particles per cm³. If the particles are divided into narrow mobility or size fractions, then the concentration of charged particles may appear less than 1 cm⁻³ in some fractions. The electric current carried by the collected charged particles may come out very low. An example: if a size fraction contains 10 charged

particles per cm^3 and the sample air flow rate is 1 liter per minute, then the collected current of about 3×10^{-17} amperes is much less than the noise level of the best electrometric instruments when applied in atmospheric conditions. Hence, to collect more particles and make the measurement possible, the sample air flow rate should be much larger.

The clusters and the smallest nanoparticles are subjects to rapid transformations and their composition can be changed when the air is heated during passage through the instrument. If the subject of the measurement is the size distribution of particles in natural conditions then the residence time of the air in the instrument should be short and the temperature and humidity of the sheath air should be preserved as in the atmosphere. This is a high-priority requirement at many applications.

The necessity of sampling the finest particles from the open atmospheric air generates some extra difficulties. The high diffusivity causes a loss of ions in the inlet tract. The numerical correction of the diffusion loss may include considerable uncertainty and complicate the calibration of the instruments. The higher the loss, the greater is the uncertainty. Minimizing of the inlet loss of highly diffusive clusters and nanoparticles is an essential requirement for the instrument.

Some hardly controlled distortions can appear due to the effect of the external electric field on the sampling of ions. A typical fine-weather atmospheric electric field over the flat ground is $100\text{--}200 \text{ V m}^{-1}$ and it can be largely increased during the presence of convective clouds in the neighborhood. The electric field-driven speed of a cluster ion near the instrument inlet may exceed tens of cm s^{-1} , which can modify the amount of sampled ions. The effect is asymmetric: the amount of ions forced by the electric field opposite the intake air flow is decreased but the sampled amount of ions attracted to the inlet remains unchanged (Tammet, 1970).

The control voltage required for the classification of intermediate ions at a high flow rate is thousands of volts. Keeping the electrometric collector on high potential is technically inconvenient and can lead to troubles in exploitation. On the other hand, the inlet of the instrument must be grounded to avoid the effect of the electric field on the sampling of ions. The distortion caused by the electric field in the inlet channel is known as the edge effect in atmospheric electric instruments (Israël, 1970; Tammet, 1970). Labowsky and Fernández de la Mora (2006) introduced the term *isopotential* to mark the DMAs where both the inlet and outlet are on the same potential and can be connected to the ground. Instruments for the research of intermediate ions in natural atmospheric air must be isopotential. This requirement is satisfied in modern air ion analyzers designed for atmospheric research (e.g. Tammet, 2006; Mirme et al., 2007).

1.3. Predecessors of the SIGMA

The uniform increase in the DMA characteristic mobility during a scan requires exponential decay of the control voltage. In the SIGMA and its predecessors, the exponential decay of the voltage is produced by the discharge of a RC-circuit. This is called the RC-technique and introduced in an early instrument UT-7509 (Tammet et al., 1977), which is, even today, still used for the measuring of artificially generated high concentration cluster ions (Parts and Luts, 2004). Immediate predecessors of the SIGMA (Symmetric Inclined Grid Mobility Analyzer) are the RC-scanning mobility analyzers of natural atmospheric ions IGMA (Inclined Grid Mobility Analyzer, Tammet, 2003) and BSMA (Balanced Scanning Mobility Analyzer, Tammet, 2006).

The IGMA was the first application of the method of inclined grids, which carries out the idea by Loscertales (1998) to design a DMA with the electric field inclined relative to the air flow. Loscertales proposed the inclined field with the aim to improve the diffusion-limited mobility resolution. However, the IGMA is a low-resolution instrument, where the diffusion of ions is of secondary importance. The configuration with inclined grids was chosen in consideration of some engineering aims and for testing the new principle in the air ion mobility analysis.

The IGMA was used in a few research projects studying the atmospheric aerosol nucleation (Iida et al., 2006).

An alternative instrument intended for the same applications is the Balanced Scanning Mobility Analyzer BSMA (Tammet, 2006). Different of the IGMA, the aspiration condensers of the BSMA have a classic configuration, which is familiar in atmospheric electric research. One instrument contains two identical aspiration condensers connected as the Komarov bridge. The BSMA was applied in studies of atmospheric aerosol nucleation (Kulmala, Tammet, 2007).

Exploitation experience of the instruments showed that easier maintenance is the strength of the BSMA, but its drawback is a higher noise level when compared with the IGMA. There are two reasons: two collectors are connected to the same electrometer and the fluctuations of the bridge balance are generating an extra noise component, especially at high humidity that deteriorates the insulation of the analyzer electrodes.

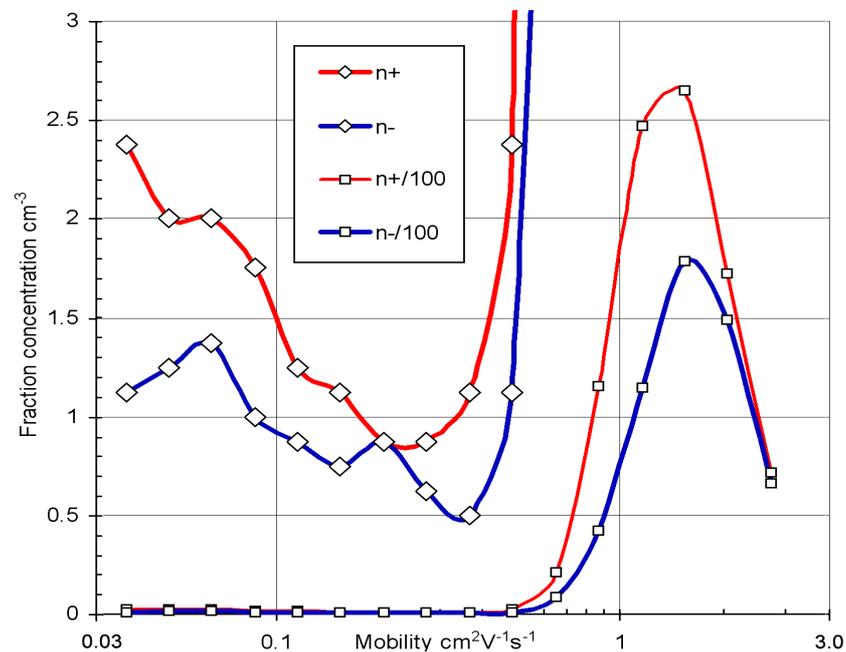


Figure 1.1. Mobility distribution presented by fraction concentrations measured at Tammemäe 20090920 from 16:00 until 24:00, a period without nucleation events. The BSMA was not able to detect the nanometer particles on the occasion of extra low concentrations like that.

1.4. Distinctive properties of the SIGMA

Some distinctive properties of the instrument are:

- The positive and negative air ions are sampled from the same inlet air flow and measured exactly simultaneously.
- A high rate of air flow more than 30 dm³/s and the isopotential principle suppress the disturbing effect of the external electric field and assure the representative sampling of air ions.
- The sheath air is sucked into the instrument directly from the atmosphere together with the analyzed air and ions pass only the unaffected atmospheric air during the analysis.
- The loss of ions in the inlet tract is only 5% at the mobility of 1 cm²V⁻¹s⁻¹.
- The residence time less than 0.2 s and low heating of air less than 0.5 K suppress the risk of the changing of the ions during the measurement.
- High sensitivity allows measuring of 8 fractions of intermediate ions on a mobility decade in atmospheric air at a standard 5-minute time resolution with random errors of about 1 cm⁻³. The sensitivity is demonstrated by an example in Figure 1.1.

- Time resolution up to 20 s is available at random errors in the mobility fraction concentrations between 4 and 10 cm⁻³, see example in Figure 1.2.
- The ions are independently distributed into the mobility fractions and the size fractions while the mobility borders of the size fractions are determined considering the simultaneously measured air temperature and pressure.
- The scanning technique assures that possible peculiarities in the recorded distributions are not generated by the technical troubles of individual mobility channels.
- The methods of measurement and instrument calibration are explicitly described in the attached documentation and the source code of the internal data processing is open and available for the user.

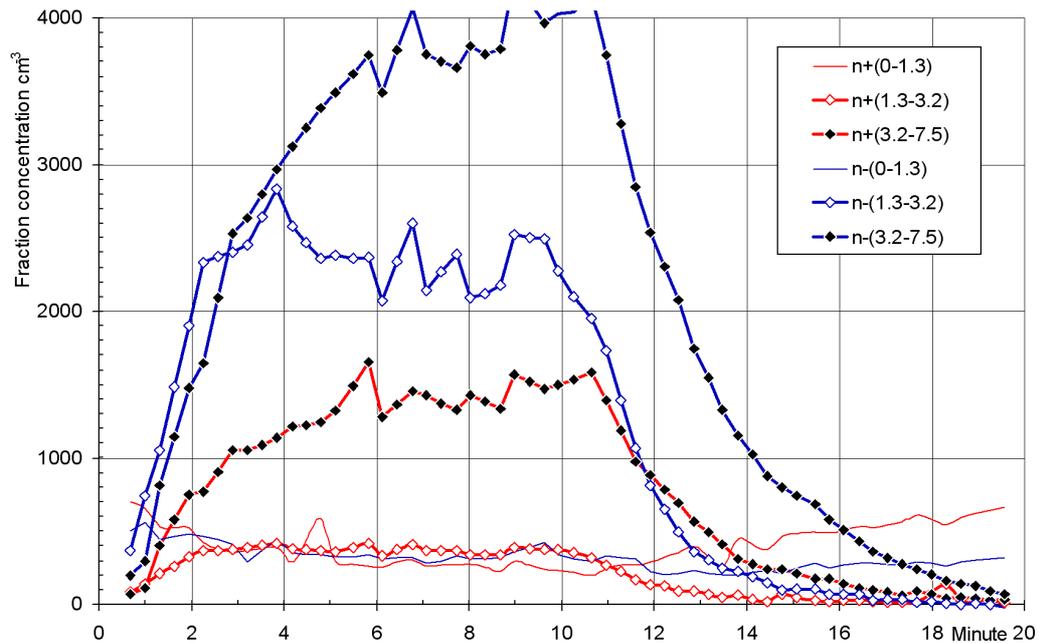


Figure 1.2. Concentrations of three size groups of ions during a water jet experiment. Blue lines show negative and red lines positive ions. The numbers in the legend show the diameter limits of the groups expressed in nanometers. The water jet was opened at 0 and closed at 10 minutes from the beginning of the experiment. During the first half-period (minutes 0–10), the concentrations are fluctuating due to the turbulent transfer of ions from the splashing point to the instrument. In the previous experiments (Tammet, Hõrrak, Kulmala, 2009) the dynamics of the processes remained unknown due to the limitations of the BSMA.

2. DESCRIPTION OF THE INSTRUMENT

2.1. Aspiration condenser

The analyzer includes a plain aspiration condenser equipped with an inlet electrostatic filter for preparing the deionized sheath air. The SIGMA analyzes simultaneously both the positive and negative ions that enter the instrument through the common inlet slit and are separated according to the polarity into two symmetric sections of the instrument. The ions are collected in two electrostatic filters shielded from the variable electric field of the mobility analyzer. A section of a SIGMA plain aspiration condenser is illustrated in Figure 2.1.

The atmospheric air enters the instrument through the inlet grid. The disturbing effect of external electric fields near the inlet is inversely proportional to the total air-flow rate and the extra high air-flow of about 30 dm³s⁻¹ is helpful in suppressing the distortions. The grid is made of a perforated metal sheet with the perforation diameter of 1 mm and the transparency of 46%. The inlet grid is necessary to prevent the entering of spiders, insects, fuzz, and hairs.

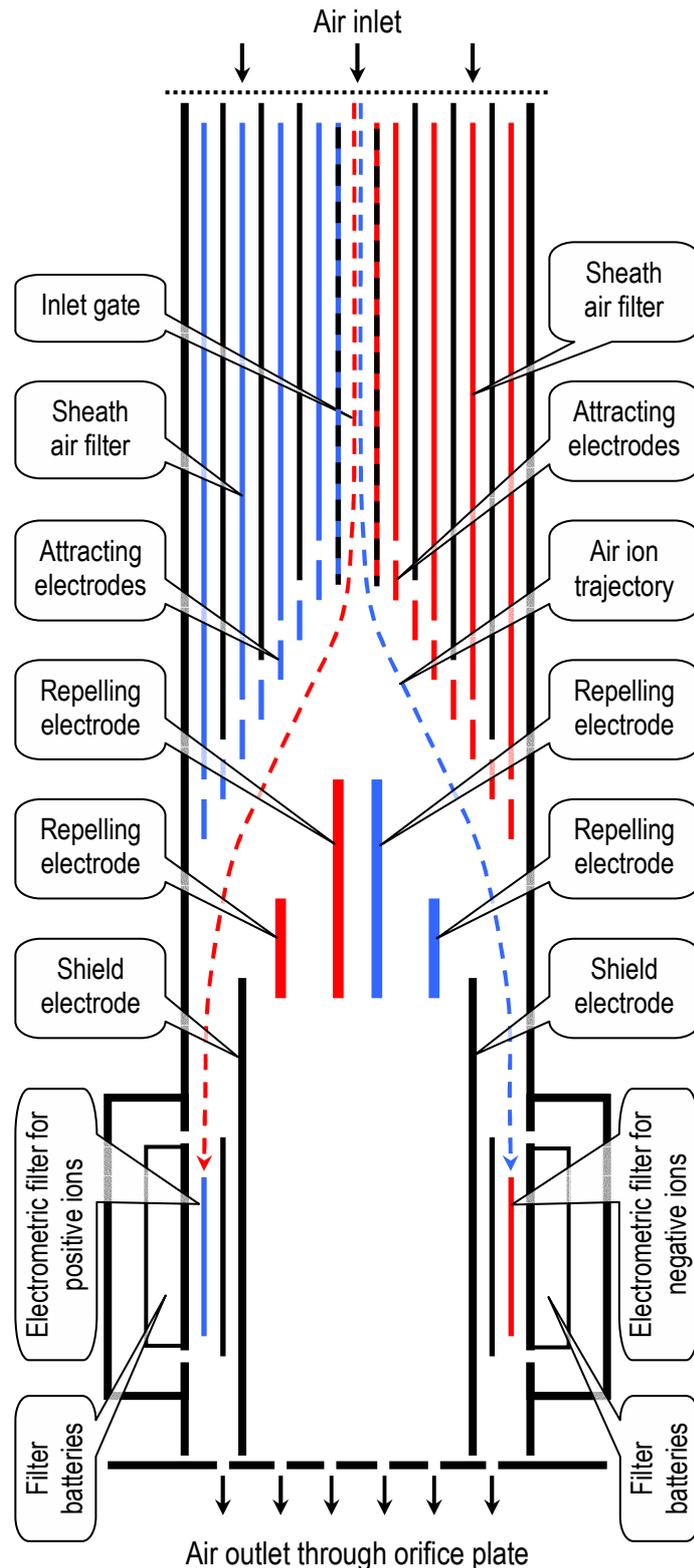


Figure 2.1. A simplified diagram of the SIGMA analyzer (M1:2). Air flow is controlled by exchangeable orifice plate and can be up to $40 \text{ dm}^3/\text{s}$. Positive electrodes are marked with red, negative blue, neutral black, and commutable with two colors. Dotted lines show two air ion trajectories. Positive ions are deflected to the left and negative to the right. The sheath air filter voltage is 520 V. Voltages of the attracting and repelling deflection electrodes are relaxing from 3000 to 20 V during each 20-second scan. Collectors of electrometric filters are supplied from 240 V internal batteries. A detailed drawing is presented in Appendix 7.

Next, the air will pass the inlet gate and the sheath air filter. The flat filter plates are 1 mm thick and the distance between the plates is 4 mm. The mean air flow speed in the filter is about 1.8 m s^{-1} and the Reynolds number between the plates of about 500 ensures the damping of the carried-on turbulence. The potential of the sheath air filter odd plates is -520 V (left side) or $+520 \text{ V}$ (right side), while all the even plates are on the zero potential. The low mobility ions passing the middle sections will not reach the collectors even if they are not precipitated in the filter. The crucial elements are the outermost sections, whose critical mobility is tenfold less than the lowest mobility of ions to be measured using the SIGMA. However, the concentration of low mobility ions can largely exceed the concentration of intermediate ions and still cause systematic errors in the collector signal. For correcting this systematic error, the zero level of the signal is to be determined and subtracted from the general measurement signal.

The inlet gate is composed of the two central plates of the filter and the space between these plates (see Figure 2.1). The voltages of the inlet gate plates are computer-controlled. The distance between the gate plates is 10 mm and the mean air speed is about 2 m/s. The gate is open when the plates are on the zero potential and closed when the plates are switched to potentials -260 V and $+260 \text{ V}$. The critical mobility of the closed gate of about $0.03 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ is a little less than the mobility of the largest particles to be measured. A closed gate scan yields a zero level record, which includes the effect of large ions passed through the sheath air filter, as well as the effect of the residual electrostatic displacement current. The air flow between the gate plates at the Reynolds number of about 1400 does not suppress the incoming turbulence. However, the main part of an ion trajectory from the gate to the collector passes the sheath air, where large-scale turbulence is damped in the filter. The Reynolds number is high in the classification zone and turbulence is one of the main factors limiting the mobility resolution discussed in the Appendix 6.7. However, a high Reynolds number in the short zone cannot lead to large distortions because there is not enough time for developing the turbulent pulsations as shown by Tammet (1970) and comprehensively verified by Rosell-Llompart et al. (1996).

The ions, which have passed the inlet gate, are deflected in the electric field between the attracting and repelling electrodes depending on their polarity and mobility. In the IGMA both the attracting and repelling electrodes were made of a perforated sheet, which caused strong distortions in the air flow in the mobility classification zone. In the SIGMA, the attracting grid is a slat grid of the same step as the sheath air filter and the distorting effect is reduced. The repelling grid is replaced by particular repelling electrodes.

The geometry of the analyzer was optimized using a numerical model, which solves the Laplace equation according to the Jacobi-Seidel method on the uniform rectangular grid with a step of 0.1 mm. The ion trajectories were calculated integrating the displacements of ions when passing in an electric field and air flow from one grid cell into the next one. The diffusionless geometric transfer function was found out by repeating the calculation of trajectories at different control voltages. The calculations were carried out on the assumption of plug air flow. However, the diffusionless transfer function depends only on the fluxes of air flow and electric field and is not sensitive to the air flow profile while it remains laminar (Tammet, 1970). The model was the crucial tool that allowed discovering the possibility to separate the positive and negative ions from the common inlet flow with a minor internal edge effect. The internal edge effect, which means the loss of ions on the rear edges of the plates of the inlet gate, is considered in the numerical model when estimating the effective width of the gate.

Nearly all ions of the central critical mobility will travel to the electrometric collector filters. The electrometric filters are shielded from the variable electric field issued from the classifier electrodes with long zero-potential shield plates. A minor part of the electric field still penetrates the channel to the collector. The penetration capacitance of 45 aF is estimated

using the numerical model. The small signal of the residual displacement current is effectively removed by the procedure of subtracting the records of the closed-gate scans.

An electrometric filter consists of one attracting plate between the two zero-potential plates. The distance between the plates is 4.5 mm, the effective electrostatic length about 50 mm, the voltage 240 V, and the air flow velocity 180 cm s^{-1} . This ensures the critical mobility of about $0.03 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, which is just low enough to collect the ions in the instrument mobility range and high enough to minimize the collecting of the large ions penetrating the inlet sheath air filter.

The time of passage of the ions through the classification zone is about 0.07 s. The passage of the air of the total distance of about 27 cm from the inlet grid to the collector entrance requires about 0.16 s. The walls of the analyzer are thermally insulated by foam polystyrene. In an extreme situation of 20 K temperature difference, the heat flux causes the mean temperature change of the air flow by about 0.5 K.

A detailed drawing of the aspiration condenser is presented in Appendix 7.

2.2. Principles of operation

Performance of the SIGMA substantially depends on manipulating the instrument by a computer and internal processing of the recorded signals under the supervision of the control program. The computer is connected to the electronic circuits inside the instrument via the USB port. The data acquisition unit USB-1608FS, manufactured by the Measurement Computing Corporation, is built into the SIGMA. The control program, which was used during the test measurements, had been written for a PC-compatible computer running under Windows.

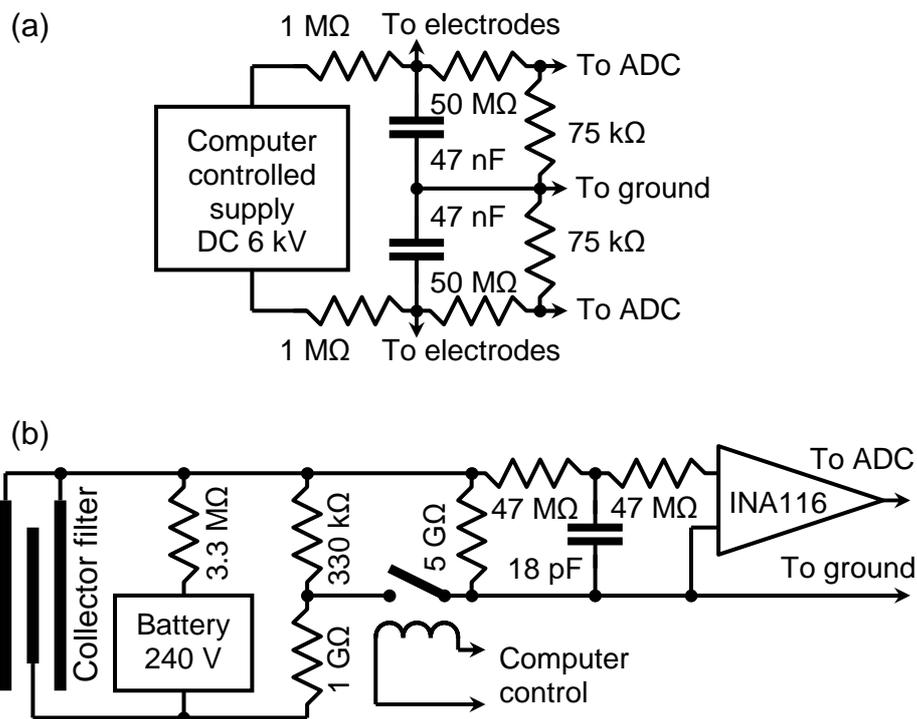


Figure 2.2. Simplified electric diagrams of the SIGMA. (a) High voltage relaxation RC-circuit, (b) electrometric collector and amplifier.

The circuit for the control of the voltage between the attracting and repelling electrodes of the mobility classifier is illustrated in Figure 2.2a. The high voltage up to 6 kV is generated by a well-insulated voltage converter U3-6PN manufactured by the Matsusada Precision Inc. The output internal capacitance of the converter is about 15 nF and the time constant of the RC-circuit is about 3.8 s. The positive output is connected to the attracting grid for the negative ions and to the repelling electrodes for the positive ions. The negative output is connected to

the attracting grid for the positive ions and to the repelling electrodes for the negative ions. The control computer can switch the inlet power of the converter on and off. About one second of power on is enough to charge the capacitors. After charging, the power is switched off and the classifier voltage will exponentially decay, which follows in a logarithmically uniform increase in the critical mobility. The decaying voltages are monitored using two ADC inputs of the data acquisition unit. The full mobility range with some reserve is passed during 18 seconds. The period composed of charging and discharging of the RC circuit is called a scan. Typically, three scans are performed during one minute. This provides the time resolution of about 20 seconds.

The circuit for collecting and recording the ion current is illustrated in Figure 2.2b. An electrometric collector filter is powered by twenty 12 V miniature batteries type GP27A. The batteries have the capacity to serve for at least one year without changing. They are enclosed into a metal box connected to the electrometric collector and well insulated from the other details. The computer-controlled electrometric relay shown in Figure 2.2b is open during the measuring of the ion current. The ion current is converted to the voltage on a precision 5 G Ω resistor and amplified with the electrometric instrumental amplifier INA116. The 1 G Ω load of the battery is required to make possible a regular check of the voltage using the computer-controlled electrometric relay. The checking procedure includes two measurements of the electrometer outlet voltage in the closed inlet gate regime. One measurement is made with the open relay and the second with the closed relay. The difference between these measurements corresponds to the voltage drop on the 330 k Ω resistor and allows estimating the actual voltage of the battery.

Detailed electric diagrams of the SIGMA are presented in Appendix 8.

During the routine measurement, the scans are performed alternately with the open inlet gate and the closed inlet gate. The schedule usually contains the groups of three scans where two scans are made with the open inlet gate and one scan with the closed inlet gate. The zero level is estimated according to the closed gate measurements. The zero level can drift due to the technical drift of the electrometric amplifier and the variation of the residual signal of large ions. The drift is slow, which allows integrating the zero signals over a three to five times longer time interval than the open gate signals. An additional benefit of repeated closed-gate-measurements is providing data for the estimate of the instrumental noise.

During a scan the classifier voltages and the electrometric signals are recorded every 0.01 s. After the required lower border of the voltage has been reached, the collected data are processed. Initially, the full mobility range is logarithmically uniformly divided into 16 narrow fractions per decade, altogether 35 fractions. The voltage range is split into subintervals corresponding to these mobility fractions and the fraction averages of the electrometric signals are calculated. Afterwards, the data are processed as explained in Appendix 4. The 16 wide fractions (see Table 5.2) are calculated on the basis of the initial narrow fractions. The parameters of the air are measured using built-in sensors during every scan and the size distribution of air ions is determined considering the air temperature and pressure according to the algorithm by Tammet (1995). The results are issued as output files described in Chapter 5.

The measuring algorithm and internal data processing are independent of the hardware. In the present manual, the measuring process is described as specified by the control program SIGMA1A. A user of the instrument is free to modify the control program or write a new control program, which may specify completely different methodology of the measuring process and a new structure of the output data.

2.3. Installing of the instrument

The SIGMA package consists of

1. Main unit of the SIGMA.
2. 24 V power supply Mascot 2020 and the power cord.
3. USB-A to USB-B cable.
4. Documentation and software for SIGMA
5. Documentation and software for MCC DAQ CD.
6. Prepared control computer.

A new computer should be prepared for controlling the SIGMA. The computer should run under MS Windows (version XP with SP2 or SP3 or version 7) and have a free USB connector. The SIGMA includes built-in data acquisition unit USB-1608FS and the MCC DAQ software for USB-1608FS should be installed first. The SIGMA software and data folder "SIGMA" should be copied from the included SIGMA CD into the computer. This folder in the computer is called the *SIGMA folder*. The location of the SIGMA folder on the computer disk is arbitrary, most convenient location is the root folder of the main disk.

Be aware that hidden system programs have a higher priority than the SIGMA control program. Aggressive background programs, like many of Antivirus programs, may take over the processor and cause failures and timeout of measurements. Thus all processes of automatic scanning and software updates, as well as all unneeded simultaneous applications, should be turned off for the period of routine measurements. If necessary, the base priority of the control program can be increased using Windows task manager.

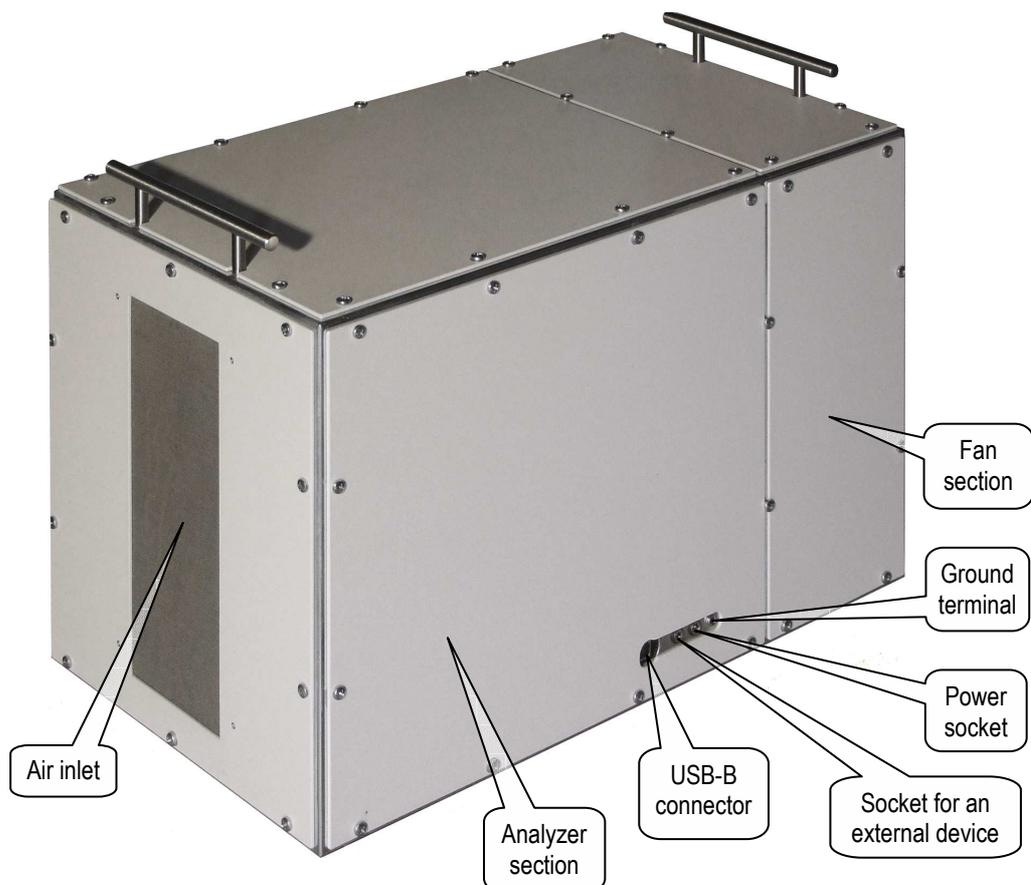


Figure 2.3. Main unit of the SIGMA.

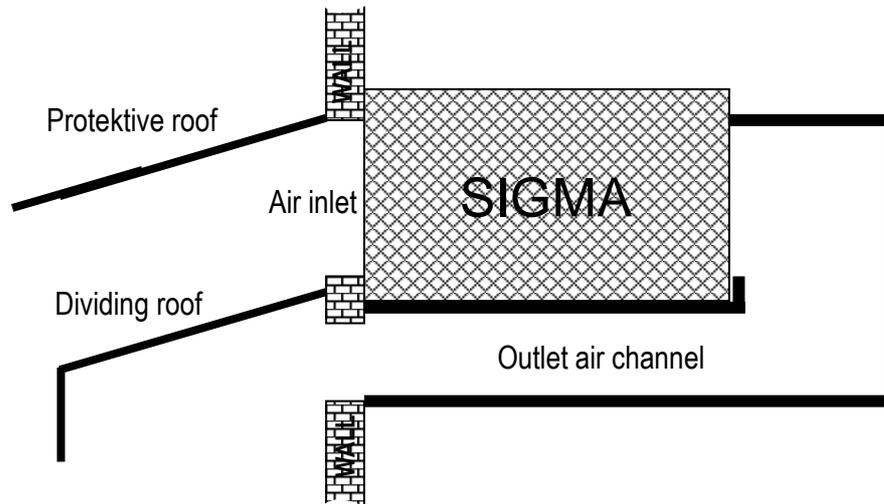


Figure 2.4. Installation of the SIGMA insensitive to wind.

The main unit of the SIGMA is shown in Figure 2.3. The normal position of the instrument is with the horizontal airflow. On this occasion, the height of the instrument with handles is 350 mm, the width is 280 mm and the length along the airflow is 550 mm. The air is sucked in through a vertical opening of 240 (height) \times 90 (width) mm in the front end of the instrument (see Figure). The air flows out from a similar opening in the rear end. The pressure drop generated by the internal air blower is about 200 Pa. This is not a high value and the instrument is slightly sensitive to wind. The external wind velocity difference of 5 m/s between the inlet and the outlet of the instrument will generate an extra pressure of about 15 Pa that results in a 4% change of the internal air flow velocity. It is recommended to install the instrument for stationary measurements in such a way that the air will flow out from the same side of the building as the inlet (see draft in Figure 2.4). The cross section of the external air channel should be at least 4 dm².

The independence of the measurement of wind is conventional. Installation according Figure 2.4 avoids a systematic shift of mobility and concentration. However, if the instrument is not protected of wind, then the concentration of large dust particles near the inlet can be high during windy weather and these particles would generate a strong noise signal in the measurements.

It is not recommended to install the instrument inlet through a wall that is exposed to the direct sunlight. The convection of heated air and temperature fluctuations near the wall can disturb the measurement, resulting in increased fluctuation noise in the measurements.

An alternative possibility is to install the instrument in a vertical position so that the inlet is below and the air will flow vertically upwards.

The most frequent service operation is the cleaning of the electrometric collectors, which requires opening of the short upper panel of the fan section, as well as the long left or right side panel. Thus it is strongly recommended to keep the access to these panels free and leave at least 300 mm free space above the fan section of the instrument.

After the instrument has been positioned, the electrical connections have to be made:

- A ground wire must be connected to the ground terminal or to a cover screw of the SIGMA.
- After the ground wire is connected, the computer should be connected with the SIGMA by means of an USB cable.
- A 24 V DC power supply (Mascot-2020 or accumulator battery) should be connected to the SIGMA via the power inlet. Make sure that the center electrode of the power plug is positive (warning: the polarity of the Mascot-2020 output plug is convertible).

- NB: when using the mains, the ground wire must be connected with the Protective Earth of the mains (green-yellow cable wire, abbreviation PE) and a mains surge voltage protector is recommended especially during the thunderstorm period.

The ground connection is obligatory because the electric field around the case of the instrument can disturb the admission of ions and charged particles into the inlet of the instrument. However, the SIGMA is relatively insensitive to weak external electric fields due to its high ventilation rates.

A desktop computer should be powered via UPS during routine measurements. A laptop computer with a good internal battery can be used without UPS. The control program SIGMA1A will restart the measurements after a power blackout when the computer is still running.

The SIGMA has an additional socket, which enables the switching of an external device (see Table 4.2 and Appendix 8). The standard control program SIGMA1A.EXE supports only the manual control of the external device. Automatic control can be included when modifying the program code; see Appendix A4.4 of the manual.

It is strongly recommended to learn the instructions in the next chapter before starting the measurements.

3. OPERATION OF THE INSTRUMENT

(excluded from the reduced manual)

4. MAINTENANCE OF THE INSTRUMENT

(excluded from the reduced manual)

5. OUTPUT FILES

5.1. Nomenclature of data files

Output of the SIGMA is compiled by the control program and may be different when using different control programs. The nomenclature and structures explained below correspond to the software, which is distributed together with the instrument.

The name of a data file is composed of the prefix, date and extension. The date is written with 6 digits, which are replaced by letters YYMMDD (year, month, day) in the examples below. If a file corresponds to several days then the combination DD is replaced by 00. If a file contains data for several months, then MM is replaced by 00. Example: filename S1A090700.XL contains the prefix S1A and extension XL, and the file contains data for several days in September of 2009.

The extension of the filenames in the examples is XL, but it can be changed by the user when editing the control file SIGMA1A.INI.

All files contain tab-delimited text tables and can be read using MS Notepad or Excel.

The output data files are:

- The *scan data* `_S1AYYMMDD.XL` is saved online by the control program into the subfolder SCANDETAILS of the SIGMA folder. It contains full information, which can later be converted offline into the basic data or standard data using the converter SIGMA1C. The conversion allows flexible setting of structure parameters independent of the cycle length in the display and in the immediately saved standard data. Typical size of a diurnal file of scan data is about 1.7 MB.
- The *basic data* `~S1AYYMMDD.XL` can be created only offline, converting the scan data by means of the program SIGMA1C.EXE. The basic data contains less information than the scan data but more than the standard data. It is suitable for a user, who is processing the data using self-written programs.
- The diurnal *standard data* `S1AYYMMDD.XL` is saved online by the control program into the subfolder DAYS and the monthly standard data `S1AYYMM00.XL` is saved online into the subfolder MONTHS. The diurnal and monthly files contain the same data and can be considered as a backup for each other. The standard data contain significantly processed information, which is easy to use by means of universal data analysis programs, e.g. MS Excel and modify by means of the preprocessor SIGMA1P.EXE. The size of a standard data file is inversely proportional to the duration of the measuring cycle. Typical size of a standard data file is about 80 kB per day in case of 5 minute cycles. Different versions of standard data corresponding to different structure parameters can be created later offline when converting the scan data with the converter SIGMA1C.EXE.
- The diurnal *diagram table* `dYYMMDD.XL` is saved online by the control program into the subfolder DIATABLES. It is prepared for the MATLAB program SIGMA1D, see Section 6.4 and Appendix 5. Typical size of a diurnal diagram table file is about 25 kB in case of 5 minute cycles. The diagram tables corresponding to different processing parameters can be created later offline when converting standard data by means of the preprocessor SIGMA1P.EXE.

In case of a serious technical failure, SIGMA1A will save into the SIGMA folder a log file named `SIGMA1A_failure.txt`, which includes the values of the diagnostic parameters in the moment of detecting the failure.

Additionally, the *normal tables* and *fraction tables* can be compiled afterwards by means of the postprocessor programs. Postprocessing of data and the structures of the resulting files are explained in Chapters 6 and 5.

Warning: Usage of MS Excel for file inspection requires special care. If an output file is opened and afterwards saved by MS Excel as a workbook, then the structure of the file is changed and may be unacceptable for the postprocessing program SIGMA1P and the diagram function SIGMA1D.

5.2. Standard headers

A file of the scan data, basic data, or standard data begins with three header lines. The following lines of the file present the rows of the data table. The header lines are:

- 1) calibration header, consisting of tab-delimited words explaining the calibration constants presented in the next line (see Table 5.1),
- 2) calibration line composed of tab-delimited numerical values: program date, calibration date, and calibration constants acquired from the current calibration file (see Table 5.1. and Appendix 4),
- 3) data header, consisting of tab-delimited words explaining the contents of the data columns depending on the specific data (see Sections 5.3–4 and Tables 5.2–3).

Table 5.1. Columns of the calibration header and the calibration line
(explanations of the parameters see Appendix 4)

No	Header	Corresponding parameters in the calibration line and SIGMA1A
1	SIGMA1A	date of the control program
2	CALIBR	date of the calibration file
3	V-fctr	voltagefactor for voltage-mobility conversion
4	C-fctr+	concentrationfactor for +ADC to dn/dlogZ conversion
5	C-fctr-	concentrationfactor for -ADC to dn/dlogZ conversion
6	st-ads	standardadsorption
7	colctr-Z	collectormobility
8	c-sup-V	c_supplyvolt
9	c-filt-V	c_filtervolt
10	c-bat-V	c_batteryvolt
11	c_bias	c_bias
12	c-pres-a	cpressurea
13	c-pres-b	cpressureb
14	c-temp-a	ctemperaturea
15	c-temp-b	ctemperatureb
16	c-hum-a	chumiditya
17	c-hum-b	chumidityb
18	e-delay	electrometerdelay
19	charging-t	chargingtime
20	timeout	timeout
21	c-inv-n2	c_inv_n2, parameter of inversion matrix
22	c-inv-n1	c_inv_n1, parameter of inversion matrix
23	c-inv-p1	c_inv_p1, parameter of inversion matrix
24	c-inv-p2	c_inv_p2, parameter of inversion matrix
25	z-limit	z_limit, threshold mobility of inversion matrix

5.3. Scan data

A table of scan data begins with three standard header lines where the third line consists of 87 words corresponding to the columns of the data table. Every data line corresponds to one 20-second scan. The 87 tab-delimited columns contain integer numbers:

- 1) time HHMMSS (midpoint between the start and finish of the scan),
- 2) regime: 0 = inlet gate closed, 1 = inlet gate open,
- 3) temperature, 0.01 C,
- 4) RH, 0.1%,
- 5) atmospheric pressure, 0.1 mb,
- 6–40) ADC counts for positive ion extended mobility distribution, 0.1 counts,
- 41–75) ADC counts for negative ion extended mobility distribution, 0.1 counts,
- 76) supply voltage, 0.1 V,
- 77) filter voltage +, V,
- 78) filter voltage –, V,
- 79) battery voltage +, V,
- 80) battery voltage –, V,
- 81) electrometer bias +, 0.01 mV,
- 82) electrometer bias –, 0.01 mV,
- 83) pretime, % (see Section 4.1),
- 84) analyzer voltage RC relaxation time constant, ms,
- 85) analyzer deflection voltage polarity asymmetry, promille,
- 86) number of electrometer overloads in the scan, 100×positive ions + negative ions,
- 87) regime, a complex index equal to the variable 78 of the standard data
(see Section 5.5).

The values of ADC counts are averages over the extended mobility distribution fractions, whose center mobilities are shown in the header.

Extended mobility distribution contains the concentrations of 35 narrow mobility fractions of positive ions and 35 narrow mobility fractions of negative ions. It is available only in the scan data and the basic data. A decade of mobility is logarithmically uniformly divided into 16 fractions in the extended distribution.

If the fractions are numbered with index $i = 1, 2, \dots, 35$ then the

- lower borders are $Z = 10^{(i-26)/16} \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ (0.0274–3.65),
- fraction centers are $Z = 10^{(i-25.5)/16} \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ (0.0249–3.92),
- upper borders are $Z = 10^{(i-25)/16} \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ (0.0316–4.22).

Prefix of the scan data filename is `_S1A`.

5.4. Basic data

Prefix of the basic data filename is `~S1A`. The structure is the same as of the scan data; only the content of columns 1, 2, 6–75, and 86 is different. The rows can be written for every open-gate scan or as averages for uniformly distributed time intervals up to 60 minutes. One file can include data for several days and the date is indicated in the first column of every row. The ADC readings of electrometers, which were presented in the scan data, are replaced by the values of the mobility distributions.

The 87 tab-delimited columns of the table contain integer values:

- 1) date YYMMDD,
- 2) time HHMMSS (if the time interval is one minute or more, then the midpoint between the start and finish of the interval),
- 3) temperature, 0.01 C,
- 4) relative humidity, 0.1%,
- 5) atmospheric pressure, 0.1 mb,
- 6–40) positive ion extended mobility distribution $dn / d \log Z$, cm^{-3} ,
- 41–75) negative ion extended mobility distribution $dn / d \log Z$, cm^{-3} ,
- 76) supply voltage, 0.1 V,
- 77) filter voltage +, V,
- 78) filter voltage –, V,
- 79) battery voltage +, V,
- 80) battery voltage –, V,
- 81) electrometer bias +, 0.01 mV,
- 82) electrometer bias –, 0.01 mV,
- 83) pretime, % (see Section 4.1),
- 84) analyzer voltage RC relaxation time constant, ms,
- 85) analyzer deflection voltage polarity asymmetry, promille,
- 86) `ovl&sc`, a complex index described below,
- 87) `regime`, a complex index described below.

The index `ovl&sc` is composed as

$$10000 \times (\text{number of positive electrometer overloads in the cycle}) + \\ 100 \times (\text{number of negative electrometer overloads in the cycle}) + \\ 1 \times (\text{number of scans in the cycle}).$$

The index `regime` is composed as:

$$100000 \times (\text{structure, 0, 1 or 2}) + \\ 10000 \times (\text{simulated measurement, 0 or 1}) + \\ 1000 \times (\text{noise regime, 0 or 1}) + \\ 100 \times (\text{extracorreption 0 = no, 1 = dust pulses, 2 = transfer, 3 = both}) + \\ 10 \times (\text{external meteodata used, 0 or 1}) + \\ 1 \times (\text{mark, which was manually set using keys 0...9 during measurements})$$

The subindex *structure* of the regime is defined as:

0 = 16-fraction full range mobility distribution,

1 = 16-fraction cluster range mobility distribution,

2 = 35-fraction extended mobility distribution

Thus the value of *subindex structure* is always 2 in the basic data; the alternative values of 0 and 1 are reserved for the standard data.

5.5. Standard data

Prefix of a standard data filename is S1A. The file is composed of three standard header lines and following data lines. The structure of the third header line is explained in Tables 5.2 and 5.3. The data line consists of 78 numeric values. Most of these are presented as whole numbers and a few contain a point-separated decimal part. The data columns are explained in Tables 5.2 and 5.3. The content of the columns 29...60 depends on the parameter *clusterregime* of the control file SIGMA1A.INI. *Clusterregime* = 0 initiates the full mobility range and *clusterregime* = 1 initiates the cluster ion regime. Columns 1...28 and 61-78 are universal and do not depend on the parameter *clusterregime*. A decade of mobility is logarithmically uniformly divided into 8 standard fractions in the regime of full mobility range and into 16 narrow fractions in the cluster ion regime.

Table 5.2. Columns of the standard data table in case of full mobility range

No	Excel	Header	Value	Sign	Range, nm or $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$
1	A	YYMMDD	Date YYMMDD		
2	B	HHMM	Time HHMM (center of the interval)		
3	C	DAY	Day of year (4 decimal places)		
4	D	T:C	Temperature, °C (1 decimal place)		
5	E	RH:%	Rel. humidity, % (1 decimal place)		
6	F	p:mb	Air pressure, mb (2 decimal places)		
7	G	noise+	Index of zero level noise, cm^{-3}		
8	H	noise-	Index of zero level noise, cm^{-3}		
9	I	D+0.487	$f_{\log d} = dn / d \log d, \text{cm}^{-3}$	+	0.42–0.56
10	J	D+0.649	$f_{\log d} = dn / d \log d, \text{cm}^{-3}$	+	0.56–0.75
11	K	D+0.866	$f_{\log d} = dn / d \log d, \text{cm}^{-3}$	+	0.75–1.00
12	L	D+1.155	$f_{\log d} = dn / d \log d, \text{cm}^{-3}$	+	1.00–1.33
13	M	D+1.540	$f_{\log d} = dn / d \log d, \text{cm}^{-3}$	+	1.33–1.78
14	N	D+2.054	$f_{\log d} = dn / d \log d, \text{cm}^{-3}$	+	1.78–2.37
15	O	D+2.738	$f_{\log d} = dn / d \log d, \text{cm}^{-3}$	+	2.37–3.16
16	P	D+3.652	$f_{\log d} = dn / d \log d, \text{cm}^{-3}$	+	3.16–4.22
17	Q	D+4.870	$f_{\log d} = dn / d \log d, \text{cm}^{-3}$	+	4.22–5.62
18	R	D+6.494	$f_{\log d} = dn / d \log d, \text{cm}^{-3}$	+	5.62–7.50
19	S	D–0.487	$f_{\log d} = dn / d \log d, \text{cm}^{-3}$	–	0.42–0.56
20	T	D–0.649	$f_{\log d} = dn / d \log d, \text{cm}^{-3}$	–	0.56–0.75
21	U	D–0.866	$f_{\log d} = dn / d \log d, \text{cm}^{-3}$	–	0.75–1.00
22	V	D–1.155	$f_{\log d} = dn / d \log d, \text{cm}^{-3}$	–	1.00–1.33
23	W	D–1.540	$f_{\log d} = dn / d \log d, \text{cm}^{-3}$	–	1.33–1.78
24	X	D–2.054	$f_{\log d} = dn / d \log d, \text{cm}^{-3}$	–	1.78–2.37
25	Y	D–2.738	$f_{\log d} = dn / d \log d, \text{cm}^{-3}$	–	2.37–3.16
26	Z	D–3.652	$f_{\log d} = dn / d \log d, \text{cm}^{-3}$	–	3.16–4.22
27	AA	D–4.870	$f_{\log d} = dn / d \log d, \text{cm}^{-3}$	–	4.22–5.62
28	AB	D–6.494	$f_{\log d} = dn / d \log d, \text{cm}^{-3}$	–	5.62–7.50
29	AC	Z+0.037	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	0.032–0.042
30	AD	Z+0.049	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	0.042–0.056
31	AE	Z+0.065	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	0.056–0.075
32	AF	Z+0.087	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	0.075–0.100
33	AG	Z+0.115	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	0.100–0.133
34	AH	Z+0.154	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	0.133–0.178
35	AI	Z+0.205	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	0.178–0.237
36	AJ	Z+0.274	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	0.237–0.316

No	Excel	Header	Value	Sign	Range, nm or $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$
37	AK	Z+0.365	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	0.316–0.422
38	AL	Z+0.487	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	0.422–0.562
39	AM	Z+0.649	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	0.562–0.750
40	AN	Z+0.866	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	0.75–1.00
41	AO	Z+1.155	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	1.00–1.33
42	AP	Z+1.540	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	1.33–1.78
43	AQ	Z+2.054	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	1.78–2.37
44	AR	Z+2.738	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	2.37–3.16
45	AS	Z–0.037	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	0.032–0.042
46	AT	Z–0.049	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	0.042–0.056
47	AU	Z–0.065	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	0.056–0.075
48	AV	Z–0.087	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	0.075–0.100
49	AW	Z–0.115	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	0.100–0.133
50	AX	Z–0.154	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	0.133–0.178
51	AY	Z–0.205	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	0.178–0.237
52	AZ	Z–0.274	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	0.237–0.316
53	BA	Z–0.365	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	0.316–0.422
54	BB	Z–0.487	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	0.422–0.562
55	BC	Z–0.649	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	0.562–0.750
56	BD	Z–0.866	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	0.75–1.00
57	BE	Z–1.155	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	1.00–1.33
58	BF	Z–1.540	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	1.33–1.78
59	BG	Z–2.054	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	1.78–2.37
60	BH	Z–2.738	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	2.37–3.16
61	BI	supply	Supply voltage, V		
62	BJ	filt+	Filter voltage +, V	+	
63	BK	filt–	Filter voltage –, V	–	
64	BL	batt+	Battery voltage +, V	+	
65	BM	batt–	Battery voltage –, V	–	
66	BN	bias+	Electrometer bias +, mV	+	
67	BO	bias–	Electrometer bias –, mV	–	
68	BP	pre%	$100 \times \text{frt [1]} / (\text{frt [2]} - \text{frt [1]})$		
69	BQ	tau	HV relaxation time constant, ms		
70	BR	asym	HV asymmetry, promille		
71	BS	N+	Concentration of aerosol ions +, cm^{-3}	+	0.032–0.49
72	BT	N–	Concentration of aerosol ions –, cm^{-3}	–	0.032–0.49
73	BU	n+	Concentration of cluster ions +, cm^{-3}	+	0.49–3.16
74	BV	n–	Concentration of cluster ions –, cm^{-3}	–	0.49–3.16
75	BW	Z+	Z_{average} of cluster ions +, $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$	+	0.49–3.16
76	BX	Z–	Z_{average} of cluster ions –, $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$	–	0.49–3.16
77	BY	ovl&sc	Complex index (explanation see 5.4)		
78	BZ	regime	Complex index (explanation see 5.4)		

Table 5.3. Columns 29...60 of the standard data table in case of *clusterregime* = 1.

No	Excel	Header	Value	Sign	Range, $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$
29	AC	Z+0.45	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	0.42–0.49
30	AD	Z+0.52	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	0.49–0.56
31	AE	Z+0.60	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	0.56–0.65
32	AF	Z+0.70	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	0.65–0.75
33	AG	Z+0.81	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	0.75–0.87
34	AH	Z+0.93	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	0.87–1.00
35	AI	Z+1.07	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	1.00–1.15
36	AJ	Z+1.24	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	1.15–1.33
37	AK	Z+1.43	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	1.33–1.54
38	AL	Z+1.65	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	1.54–1.78
39	AM	Z+1.91	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	1.78–2.05
40	AN	Z+2.21	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	2.05–2.37
41	AO	Z+2.55	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	2.37–2.74
42	AP	Z+2.94	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	2.74–3.16
43	AQ	Z+3.40	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	3.16–3.65
44	AR	Z+3.92	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	+	3.65–4.22
45	AS	Z–0.45	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	0.42–0.49
46	AT	Z–0.52	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	0.49–0.56
47	AU	Z–0.60	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	0.56–0.65
48	AV	Z–0.70	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	0.65–0.75
49	AW	Z–0.81	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	0.75–0.87
50	AX	Z–0.93	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	0.87–1.00
51	AY	Z–1.07	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	1.00–1.15
52	AZ	Z–1.24	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	1.15–1.33
53	BA	Z–1.43	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	1.33–1.54
54	BB	Z–1.65	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	1.54–1.78
55	BC	Z–1.91	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	1.78–2.05
56	BD	Z–2.21	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	2.05–2.37
57	BE	Z–2.55	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	2.37–2.74
58	BF	Z–2.94	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	2.74–3.16
59	BG	Z–3.40	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	3.16–3.65
60	BH	Z–3.92	$f_{\log Z} = dn / d \log Z, \text{cm}^{-3}$	–	3.65–4.22

The concentrations of aerosol ions (variables 71 and 72) and cluster ions (variables 73 and 74) are calculated according to the measured mobility distribution, where ions of the mobility of less than $0.5 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ are considered as aerosol ions and ions of the mobility of above this threshold are considered as cluster ions. Explanations of the complex index variables 77 and 78 are presented at the end of Section 5.4.

5.6. Diagram table

A diagram table dYYMMDD.XL saved by SIGMA1A corresponds to one day. The table has no header and contains 21 columns of tab-delimited numerical values. The number of rows is $1 + 1440 / \text{cycleminutes}$ (289 rows in case of standard value *cycleminutes* = 5) and they correspond to the values of minute in the day 0...1440. The first column contains the minute in the day and the columns 2...21 the same data as in the columns 9...28 of the standard table (values of $dn / d(\log d)$ for positive and negative particles). The missing values and the negative values are replaced by zeroes. The tables, which are saved immediately by the control program, are couple smoothed. The smoothing methods are explained in Section 5.7.4.

If the control program SIGMA1A was temporarily closed for some time during the day, then only the data collected after the restart are available and presented in the immediately saved diagram table.

The online saved diagram tables are intended for a preliminary inspection of the data. The diagram tables of the same structure can be generated afterwards using the postprocessor program SIGMA1P, which acquires information from the scan details or standard data and makes the flexible control of data conditioning available. See Chapter 6.

5.7. Comments about the output data

5.7.1. Recurrent files

A diagram table dYYMMDD.XL is created and saved once at midnight and will never be changed afterwards. This is the only non-recurrent output file of the control program.

Other online output files of the control program are recurrent. The diurnal file of scan data *_S1AYYMMDD.XL* is completed after every scan and the diurnal file of standard data *S1AYYMMDD.XL* is completed after every cycle. If the data saving is temporarily interrupted by a control key or a pause in measurements then the completing process will be restarted after restoration of the data saving regime and the beginnings of the files are preserved.

The monthly file of standard data *S1AYYMM00.XL* is completed every midnight and the saving regime set by the controls M and D is checked just before saving. If the control program SIGMA1A was temporarily closed for some time during the day then only the data collected after the restart are available in the computer memory. Thus the completeness of *S1AYYMM00.XL* depends on the way the program SIGMA1A was closed before the interruption. In case of a normal XZ procedure the computer saves the data of the unfinished day into the monthly file and no data are lost. In case of an irregular closing of the program, the beginning of the day may be absent in the monthly file. However, the lost data can be found and restored using the parallel diurnal file *S1AYYMMDD.XL*.

The log of failures *SIGMA1A_failure.txt* is always appended and contains data since it was first created.

Warning: The headers of the files contain information read from the calibration file and the control file. The headers of a recurrent file are written once when the file is first created. If the measurements are interrupted and restarted with new settings, then the header lines are not changed and will not correspond to the new settings. This may be followed by misinterpretation of the saved data in the future. Thus an **urgent recommendation:**

rename data files S1AYYMMDD.XL and _S1AYYMMDD.XL if the measurements will be restarted during the day of changes and rename S1AYYMM00.XL if the measurements will be restarted during the month of changes in the calibration file. Do not change the date included into the filename and use only an additional letter after the date.

Example: *S1A091218.XL* can be renamed as *S1A091218a.XL*.

Another way is to move the existing recurrent files away from the standard folders. In both cases the control program will create new data files including up-to-date header lines.

5.7.2. Treating of negative values

The true values of concentrations and distribution functions cannot be negative. However, the recorded values are sometimes negative due to the random noise. When the concentration of intermediate ions is much less than the instrumental noise, then about half of the measured values of distribution functions can be negative in the intermediate ion range. The negative values may be treated in different ways.

The control program SIGMA1A always records the scan data without any corrections. The standard tables can be recorded without corrections or corrected with the aim of dust pulse

suppression. A cloud of charged dust generates a negative pulse in one polarity channel and a positive pulse in the other polarity channel. If an internal narrow fraction concentration comes out negative, then a compensating positive value can be added to this fraction concentration and simultaneously subtracted from the fraction concentration of the opposite polarity. The probability of such an event is nearly equal for both polarities and the correction does not produce a systematic shift in the results. The dust pulse correction can be switched on or off by editing the control file SIGMA1A.INI.

All negative values are replaced with zeroes only in the diagram tables. Replacement of negative values with zeroes produces a shift in the averages calculated for longer time intervals or wider mobility ranges. Thus the diagram tables should not be used as a source for further calculations.

A recommended method for suppressing the negative values is the smoothing of the data in time during the offline postprocessing, see the next section and Chapter 6.

5.7.3. Extra corrections

Extra corrections are options in the SIGMA standard software. They can be applied or not according to the choice by the user. In SIGMA1A.INI the choice is expressed by the value of the parameter *extracorrect*.

Pulses induced by clouds of charged dust passing the analyzer are the essential elements of noise in the electrometric signal. These pulses have different polarity in records of positive and negative air ions and can be identified when the real value of the distribution function is near to zero. Then a negative value in the distribution function for one polarity and a positive value in the opposite polarity is an indication of the dust pulse. In this case the disturbance can be compensated by a transfer of some part of the negative signal from one polarity to another. The dust pulse correction algorithm in the SIGMA standard software is applied to the array of 35 narrow fraction signals, which are denoted below pos_i and neg_i :

```
for i := 1 to 35 do begin
  p := posi; q := negi;
  if (p < 0) and ((p + q) ≥ 0) then begin posi := 0; negi := p + q; end;
  if (q < 0) and ((p + q) ≥ 0) then begin negi := 0; posi := p + q; end;
end;
```

Another extra correction available in SIGMA1A is a partial inversion of the transfer function with the aim of improving the mobility resolution in the cluster ions subrange. Let x_i be the true distribution and y_j be the broadened record of the spectrometer. Then

$$y_j = \sum_i G_{ji} \times x_i \text{ and } x_i = \sum_j H_{ij} \times y_j$$

where G is the transfer matrix and $H = G^{-1}$ is the inverter matrix. Conservation of the air ion concentration requires $\sum_j H_{ij} = 1$. The elements of H tend to be alternatively negative and positive and the coefficient of noise amplification $K = \sqrt{\sum_j H_{ij}^2}$ may come out intolerably large. As a rule, the full inversion appears inappropriate and the matrix for partial inversion is to be chosen as a compromise between mobility resolution and noise. The SIGMA standard software allows using only pseudo-five-diagonal inverter matrices, whose elements are calculated according to the last five calibration coefficients (see Table 5.1) as follows:

```
for i := 1 to 35 do begin
  if Zi < z_limit then c := Zi / z_limit else c := 1;
  sum := 0;
  for j := 1 to 35 do begin
    Hij := 0
    if j = i - 2 then Hij := c × c_inv_n2;
    if j = i - 1 then Hij := c × c_inv_n1;
    if j = i then Hij := 1;
    if j = i + 1 then Hij := c × c_inv_p1;
    if j = i + 2 then Hij := c × c_inv_p2;
    sum := sum + Hij;
  end;
  for j := 1 to 35 do Hij := Hij / sum;
```

end ;

5.7.4. Smoothing of data in time

Smoothing of time series can be used to suppress the occasional negative values of the distribution functions and to reduce the random noise. The control program SIGMA1A applies smoothing only in diagram tables. Deeper smoothing is available in the postprocessor SIGMA1P. Two methods are used:

- couple smoothing replaces each member of time series x_i with $(x_{i-1} + x_i) / 2$,
- triplet smoothing replaces each member of time series x_i with $(x_{i-1} + 2x_i + x_{i+1}) / 4$.

The couple smoothing involves a half-step time shift. This is just welcome when compiling the diagram tables because the standard data in case of *cycleminutes* = 5 correspond to the minutes 2.5, 7.5, etc but the values in a diagram table should correspond to the minutes 0, 5, 10, etc. Thus the couple smoothed diagram tables saved by the control program are free of the time shift.

The postprocessor can apply triplet smoothing when compiling normal tables and both triplet and couple smoothing when compiling the diagram tables. The triplet smoothing can be applied repeatedly and the number of reiterations is called the *smoothing level*. Reduction of the time resolution by triplet smoothing is visible when comparing Figures 6.3 and 6.3.

The standard tables saved online by SIGMA1A are not smoothed, which means the smoothing level is zero.

5.7.5. Mobility distribution or size distribution?

The control program SIGMA1A records simultaneously the distribution of ions according to the particle diameter and the distribution according to electric mobility. The two distributions are compiled using different partitions of the series of electrometer signals according to the voltage of the aspiration condenser recorded during a scan. However, the distribution according to mobility is a more fundamental result, because it presents a better size/mobility resolution and does not depend on:

- the specific mobility-size conversion algorithm,
- the assumptions about the particle density and charge,
- the measurements of atmospheric pressure and air temperature,

The control program SIGMA1A uses the algorithm by Tammet (1995) assuming the particle density of 2.08 g cm^{-3} and the particle charge of 1 elementary charge. The results do not fit well the assumptions because particle density affects the conversion only for the finest particles of the diameter of below 2 nm and the probability of double charges on particles below 7.5 nm is usually negligible (warning: an exception may occur when measuring electrospray ions).

The most fundamental result is the extended mobility distribution saved in scan data as a raw table of the ADC counts and in the basic data as a table of $dn / d \log Z$. These tables are accompanied by the values of air temperature and pressure and allow a later compilation of size distributions on the arbitrary diameter grids as specified by the user.

6. DATA PROCESSING

6.1. Patterns of data processing

As a rule, the control computer is not used for data processing. Thus the data are to be transferred into another computer at first. The program *data_transfer.exe* supports convenient transfer of all new data files from the control computer to a memory stick or SD card.

Three typical methods of data processing are:

Introductory level

- use online-saved standard data and diagram tables,
- process the diagram tables with SIGMA1D and inspect the results,
- analyze the standard data by means of MS Excel.

Medium level

- use online-saved scan data and diagram tables,
- process the diagram tables with SIGMA1D for a preliminary inspection of the results,
- convert the scan data into the standard data using the converter SIGMA1C,
- convert the data into an appropriate structure using the postprocessor SIGMA1P,
- visualize the data (use SIGMA1D) and analyze it by means of MS Excel or packages for statistical analysis,
- repeat the last operations until obtaining required results.

Advanced level

- use online-saved scan data,
- convert the scan data into the basic data using the converter SIGMA1C,
- analyze the data by means of advanced statistical packages and user-written programs.

6.2. Converter SIGMA1C

SIGMA1C.EXE converts the raw scan data into the standard data or the basic data. It makes available the repeated low level data processing using different values of the parameters. The user is free to choose the duration of the cycle and the period of the checking of the zero level independent of the parameters of the online-saved standard data.

The program is to be launched dragging a file or a folder onto the converter icon. The dragged item should be:

- a file of scan data, whose name begins with *_S1A* or
- a folder containing files of scan data (maximum 1000000 scans \approx 7.5 months).

The dragged folder or the folder, where the dragged file is located, is called the *working folder*. All files in the dragged folder are expected to be recorded with the same version of SIGMA1A.EXE and the same version of SIGMA1A.CAL in the regular situation. Otherwise, the file headers can be imported only from the last file. The output file will be saved into the working folder. If all records belong to the same day, the name of the standard data will be S1AYYMMDD, otherwise S1AYYMM00, S1AYY0000, or S1A000000. Corresponding names of the basic data have additional prefix *~*. The extension will be set according to the demand.

The converter asks for five control parameters, whose values can be entered from the keyboard or written as a plain text into the control file SIGMA1C.INI. This file is compiled according to the same rules as SIGMA1A.INI and should be saved into the working folder. If the control file is found, then the parameters will be imported from this file. If a line of the

control file is absent or marked as a comment (the first symbol is space), then the converter asks for the missing value from the keyboard. An example of the content of the control file is:

```
SIGMA1C.INI 20101027
outputstyle=0      0 = full range (8 fr/dec) standard data,
                   1 = clusterregime (16 fr/dec) standard data,
                   2 = basic data
extracorrect=3     0 = no, 1 = dust pulses, 2 = resolution, 3 = both
cycleminutes=2    0..60, 0 means one cycle per scan, otherwise
                   60/cycleminutes must be a whole number
zerominutes=20    3..600 but not less than cycleminutes
extension=x1      extension of the output file name
```

If the parameter *cycleminutes* is indicated as 0, then the output will be compiled for all individual open-gate scans that have an interval of about 20 s or 40 s.

Extracorrect has the same function as in the control program SIGMA1A. The five inverter parameters are imported from the header of the file of scan details. However, there is a possibility to use different parameter values, which should be added into the control file SIGMA1C.INI as optional lines, e.g.:

```
c_inv_n2=-0.16    inverter parameter
c_inv_n1=-0.22    inverter parameter
c_inv_p1=-0.10    inverter parameter
c_inv_p2=-0       inverter parameter
z_limit=1         inverter parameter
```

In this case, the second header of the output files includes the new parameter values.

The control program SIGMA1A begins to issue the standard data only when the full symmetric zero period, containing $(\text{zerominutes} - \text{cycleminutes}) / 2$ minutes before as well as after the cycle, is available. Different from this, the converter begins to issue the results straightaway when the reference cycle comes available, at first using only one wing of the full zero period.

6.3. Postprocessor SIGMA1P

The postprocessor allows modifying the structure of the standard data, which was saved by the control program SIGMA1A or the converter SIGMA1C. It is used for merging the files, extracting the excerpts with limited time range and/or limited set of variables, calculating the size distribution on an user-defined diameter grid, calculating the averages for hours or different special time periods, smoothing the time series, correcting the outliers, and compiling the diagram tables according to the standard data. *Technical restriction:* the number of lines in the input tables as well as in the output table should not exceed the number of minutes in one year ($366 \times 24 \times 60$).

The program is to be launched by dragging a file or a folder onto the postprocessor icon. The dragged item should be a standard file, whose name begins with S1A, or a folder containing standard files. The dragged folder or the folder, where the dragged file is located, is called the *working folder*.

An exceptional usage: if two files are dragged together then one file should be a standard file and the second could be any file. The sole result of the second file is a special regime: output variable 61 will be calculated independent of the variable list as the ratio of the output time step to the input time step and marked with n in the data header. NB: the number of output variables must be at least 61 in this case.

A normal file created by SIGMA1P can be processed like a standard file if it consists of the same 78 variables as a standard file and has a name S1A###x00.xl (## marks any digit and x any character permitted by Windows in a filename).

If the user is going to calculate values of the size distribution at extra diameters, then a special file *diameters.txt* must be prepared and saved into the working folder beforehand.

If the dragged item is a folder, then a situation when some files are saved in the cluster regime and some in the normal regime is possible. In this case the postprocessor identifies the regime according to the first file and skips all the data recorded in the conflicting regime.

The data in the input folder can be duplicated e.g. when a monthly file and some diurnal files are simultaneously included into the working folder. If two input rows appear equal, then only one of them is accepted as the actual input data.

After the program is launched, the user is asked several questions. The sequence of questions can be interrupted any time pressing immediate ENTER without entering any answer. An exception is stating the name of the output subfolder for diagram tables: here the immediate ENTER means that the tables should be written directly into the working folder. When all questions are answered, the postprocessor analyzes the dragged files, compiles an output file or files, and saves them into the working folder. The questions are:

- if the input data is presented for several days, then the first day and the last day to be processed (6-digit format yymmdd),
- correction method (0...3),
- multiplier of standard deviation (3...9)
is asked only when the correction method is 1 or 3,
- smoothing level for negatives n (0...9),
- fill single gaps in time series (1) or not (0),
- total smoothing level k (-1...9),
- time step in the output tables expressed in full minutes (1...1440).

The ratio (1440 / time step) should be a whole number. Time step in the output table should be equal or larger than the time step in the input tables. If the time step is larger, then a row of the output table will present an average of several rows of the input standard table. If the time step is 3 minutes or more then the next question is:

- trimming degree g .

The following question will be:

- compile Normal table, Fraction table, or Diagram tables (N, F, or D).

If the required output is diagram tables then the program will ask for the

- name of the output subfolder.

If the folder with the chosen name does not exist, it will be automatically created as a subfolder of the working folder.

If the required output is a normal table or a fraction table, then three questions follow:

- format of the time presentation,
- list of variables,
- name of the output file.

6.4. Explanations of the parameters of the postprocessor

Three cycles are called *close neighbor cycles* if both the time steps before and after the central cycle do not exceed 10 minutes and the ratio of these time steps does not exceed 1.5.

► **Corrections** will be made *only in the range of the intermediate ions* of $d = 2.05\text{--}4.87$ nm and $Z = 0.049\text{--}0.365$ cm²V⁻¹s⁻¹. **Method 0** marks no data correction. **Method 1** is a formal outlier analysis and replacement. At first the close neighbors (neighbor size or mobility and close neighbor cycle) of the value of the distribution function are collected. A correction can be made when at least 6 close neighbors of possible 8 are present. The minimum and the maximum values are trimmed away from the sample of neighbors. A value is qualified as outlier when the deviation from the average of neighbors exceeds at least *multiplier* times the standard deviation inside the sample of neighbors. An outlier is replaced by the trimmed mean of the neighbors. *Correction method 2* is based on the strong negative correlation of errors created by charged dust clouds in the positive and negative ion channels. When a result of measuring the concentration of a distribution fraction is negative in one polarity and positive

in the opposite polarity, then the event is identified as the dust cloud effect and the negative value is fully or partially moved into the opposite polarity. Correction **method 3** includes both the methods 1 and 2. NB: the integral values Z , n and N are not subjects of correction and could be extra recalculated according to the corrected distributions when needed.

Correction and smoothing operations will be carried out when analyzing the input data before converting the time series to the output time step.

► Triplet *smoothing* (see Section 5.7.4) of close neighbors in time is performed at first n times for negative values and, after that, k times for all values.

An exceptional value $k = -1$ means no smoothing, as does $k = 0$. The difference appears only when the output is diagram tables. Normally, the diagram tables are additionally once *couple smoothed* after k repeated procedures of triplet smoothing are finished. The couple smoothing is performed as well at $k = 0$. The exceptional value $k = -1$ prevents the couple smoothing. This ensures the best time resolution but produces a half-cycle time shift. E.g. at the 5-minute cycles the diagram table lines marked with 0, 5,... minutes will correspond to the time of 2.5, 7.5,... minutes etc. Such a time shift is peculiar only to the SIGMA1P-produced diagram tables in case of $k = -1$. Otherwise, the couple smoothing ensures exact synchronization of time in diagrams.

The smoothing is performed on the time grid of input files. If the output time step largely exceeds the input time step, then the calculating of averages may outperform the smoothing, which will have only a minor effect in this case.

Variables 1–4, 66, 67, 77 and 78 of Table 5.2 are excluded from processing during the smoothing of negatives and variables 1–3, 77, and 78 are excluded during total smoothing.

► If the *time step* in the output table equals the time step of the input table, then every row of the input standard table produces one row in the output normal table of the fraction table. A larger number requires averaging of the data in time. The maximum allowed output time step of 1440 minutes leads to a table of diurnal averages as the output. The averages are calculated as g -trimmed means in the output time intervals. As an exception, the columns 1, 77, and 78 (YYMMDD, ovl&sc and regime) of the standard table are processed in a different way and presented by the input values in the center of the output time intervals independent of the trimming degree. Variable 2 (HHMM) is exceptionally presented independent of the input data by the calculated central time of the interval.

Warnings:

- the output time step must be chosen so that a full day will consist of a whole number of the steps,
- the output time step must be equal to or larger than the time step in the input tables.

► *Trimming degree* g shows how many of the smallest and the largest entries are excluded while calculating the arithmetic average of $n - 2g$ middle entries of the n input values (n is the number of input time steps in an output time interval). Trimming can be prevented by choosing $g = 0$.

The computer has no information about the actual time steps in the input tables when asking for the trimming degree. Thus the answer to the question about the trimming degree is limited to assuming that an input table can contain measurements with one minute time step. Too high trimming degree will be automatically reduced. The maximum trimming will result in the median mean.

► A *Normal table* includes three header lines and the following data lines like a standard data table (see section 5.5). The differences are:

- the first line is the header explaining the process parameters asked by SIGMA1P,
- the second line contains the values of the process parameters,
- the third line contains column headers for the output variables, specified by the entered list of variables,

- the data lines contain the values of variables selected according to the entered list,
- time can be presented in a flexible format explained below.

► *Fraction tables* differ from normal tables only in the presentation of the distributions. In a fraction table, the values of $dn / d \log d$ or $dn / d \log Z$ are replaced by the fraction concentrations, which are, in the decade-to-eight structure, just 8 times less than the values of the distribution functions : $dn / d \log d = 8 n (d_1, d_2)$ and $dn / d \log Z = 8 n (Z_1, Z_2)$.

► *Diagram tables* have the same structure as produced online by SIGMA1A, see Section 5.6. The smoothing level is flexible (level 0...3 is recommended), the gaps up to one hour are filled with interpolated data and tables with less than one hour of measurements are skipped in the output. For comparison: SIGMA1A always applies couple smoothing (like in case of smoothing level 0), does not fill the gaps and delivers a table independent of the number of actually recorded cycles.

► The *filename* should not coincide with a name of any pre-existing file in the working folder.

► If the *format of time presentation* differs from "x", then a row of the output table will proceed from the time presented according to the chosen format. The time compiled according to the specified format corresponds to the center of the time interval in the normal table. The format of time presentation can be:

- x – no extra time column in the output,
- j – day of year (integer 1...366),
- j.f – day of year with the 4-digit decimal fraction,
- t.f – MS Windows DateTime with the 4-digit time fraction,
- custom format.

Table 6.1. Examples of time presentation

Format	Example
yyyy-mm-dd hh:nn	2009-08-09 15:52
dd.mm.yyyy @ hh	09.08.2009 @ 15
j	221
j.f	221.661

A *custom format* of time presentation can be compiled using following notation:

- yyyy – year, yy – two last digits of the year,
- mm – month, dd – day, hh – hour,
- nn – minute (NB: mm would be month),
- any separator different of the following letters: y m d h n j f t x .

Some examples are given in Table 6.1.

If the user is satisfied with the time presentation style of the input standard table then the description of the time format can be written with one letter x (no extra time column). The time columns (1, 2 and/or 3, see Table 5.2) of the standard table will be carried forward into the output table when the corresponding column numbers are included into the list of variables.

► *Extra diameters* (an option) should be specified in a special file *diameters.txt* saved into the working folder. This file should contain just as many lines as the number of extra diameters. Each line should contain one diameter expressed in nanometers. The number of diameters should not exceed 20 and the diameters must lie in the range of 0.5–7 nm. Calculation of the distribution function on an extra diameter begins with a search for three logarithmically closest diameters in the standard table diameter set, which contains 10 diameters from 0.49 to 6.5 nm. Next, the value of the distribution function is calculated using the method of square parable interpolation on the logarithmic scale of the diameters.

► *List of variables* for the output table is to be compiled from the numbers of the variables shown in the first column of Table 5.2 or 5.3 using delimiters "," and "-". Spaces in the list are ignored. An example: the variable list 6,4,29-60 requires the including of 34 variables – the atmospheric pressure, temperature, and mobility distributions for positive and negative air

ions. The legal entries of the list of variables are limited with intervals $1...78$, $81...80 + n$, and $101...100 + n$, where n is the number of extra diameters specified in the file *diameters.txt*.

Values of $dn/dlog(d)$ for *extra diameters* can be indicated in the list by numbers exceeding 80. The value of $dn/dlog(d)$ should be indicated in the list as the variable number $80 + k$ for positive and $100 + k$ for negative ions, where k is the number of the line in the file *diameters.txt*.

An example of the computer display during data postprocessing is presented in Figure 6.1.

```

Postprocessor SIGMA1P version 20111218
Date 2011-12-18, time 22:09:08
dragged item is folder C:\temp\test
found 3 SIA-files:
C:\temp\test\SIA100419.xl
C:\temp\test\SIA100420.xl
C:\temp\test\SIA100421.xl
found a file of 7 extra diameters:
1.00 2.00 3.00 4.00 5.00 6.00 7.00

Please answer some questions below. Asked parameters are explained
in the manual of SIGMA. NB: the input procedure can be interrupted
pressing ENTER without entering any answer.

Choose the days to be processed:
first day (use 6-digit format YYYYMMDD) = 100419
last day (use 6-digit format YYYYMMDD) = 100420

Correction methods (see SIGMA manual for detailed explanations):
0 = no correction,
1 = elimination of outliers
2 = suppression of dust-induced errors,
3 = combination of the methods 2 and 3.
Please choose the method (0..3) = 3
A value is qualified as outlier when the deviation from the average
of neighbors at least multiplier times exceeds the standard deviation.
Choose multiplier of std (3..9) = 5

Some random errors can be suppressed repeating n times the smoothing,
which replaces only negative values of distribution functions with
(1+2+1)/4-triplet averages over the neighborhoods of these values,
please choose smoothing level for negatives n (0..9) = 5

The data can be totally smoothed repeating k times the replacing
of values with (1+2+1)/4-triplet averages over the neighbors.
Diagram tables will be additionally couple-smoothed by default.
Extra choice k = -1 is equivalent of k = 0 for triplet smoothing,
but cancels default couple smoothing of diagram tables.
Please choose total smoothing level k (-1..9) = 0

Single gaps in time series can be filled with interpolated values.
fill single gaps (1) or not (0), write 0 or 1 : 0

Write time step for output table in minutes (1..1440) = 60

g lowest and g highest entries can be trimmed away
when calculating averages in the enhanced time intervals.
Please choose trimming degree g (0..20) = 3

Compile Normal table, Fraction table, or Diagram tables (N, F, or D) = n

See rules for time formatting in the manual
(examples: x = no extra column, yyyy-mm-dd hh:nn = ISO standard)
and choose format for time presentation = x

Example of a list: 6,4,29-60 (see rules in the manual)
Write list of variable numbers = 1-28,81-87,101-107

Please choose name for the output file = example.xl
Method 1: outliers are corrected in 1.240 % of available cells
Method 2: dust pulses are corrected in 0.548 % of available cells
Done

Finished 22:11:25, press ENTER!

```

Figure 6.1. Window of the postprocessor after answering all questions.

The DataDiurna data manager (Tammert, 2009) makes available different possibilities for data reorganization and, especially, for compiling of a data archive.

6.5. Diagram function SIGMA1D

Two kinds of time-size distribution diagrams that can easily be created by means of MATLAB are surface plots and contour plots. A good MATLAB function written by Kaupo Komsaare allows creating surface plots using the monthly standard files. This function is available in the SIGMA website <http://ael.physic.ut.ee/tammet/sigma/>. Contour plots can be created by means of a MATLAB function SIGMA1D.m, which is presented in Appendix 5. The function processes automatically all diagram table files from the folder indicated by the *filepath*, which correspond to the indicated year *yy*. The results are saved in the same folder as the diagram tables and named *prefixYYMMDD.PNG*, where *prefix* is an arbitrary text defined in SIGMA1D. One image contains diagrams for both polarities. The size of an image file is usually in the range of 60–100 kilobytes in case of standard 5-minute cycles.

It is recommended to use a special working folder, which contains only the file SIGMA1D.m and the necessary diagram table files. After processing, the diagram table files and the resulting png-files can be moved away from the working folder.

Before launching the function, SIGMA1D.m should be edited because the information about the data location and process control parameters are written immediately into the function code. The full text of the function is presented in Appendix 5. The text can be copied into the clipboard and then pasted into the Notepad, M-editor, or immediately into the MATLAB command window.

Control lines are explained by comments in the program code:

```
% The control lines to be modified by the user (the permanent sample is preserved above):
prefix = 'SIGMA'; % for output file name
filepath = 'C:\_A\Aparaat\SIGMA\SIGMA1D\test\'; % where the files are located
yy = '09'; % year
months = 6:10; % interval of months, can be restricted
days = 1:31; % interval of days, can be restricted
first_hour = 0; % integer 0...23
hours_in_plot = 24; % integer 1...24
plots_in_day = 1; % (first_hour + plots_in_day * hours_in_plot) should not exceed 24!
first_mark = 0; % number of extra letter, 0 = nothing, 1 = a, 2 = b, ...
levels = [100 200 300 500 700 1000 2000 3000]; % for dn/dlogd
% The controls, which could be modified only in special situations:
pos_axes = [50 320 700 200]; % left, bottom, width, height
neg_axes = [50 60 700 200]; % width (standard is 700) may be sometimes modified
y_tickpositions = [1 2.18 3.43 4.85 6.65 8.3 10]; % vertical positions
y_ticklabels = {'0.5';'0.7';'1.0';'1.5';'2.5';'4.0';'6.5'}; % nanometers
```

The typical settings of the time parameters are written in the example above. It follows one full day diagram per day like presented in Figures 6.2 and 6.3. The name of the output file in Figure 6.2 is SIGMA090806.png and the file occupies 56 kB on the computer disk. Figure 6.3 is compiled for the same day and thus it is recommended to set a different value of *first_mark*. If *first_mark* = 1 then the diagram file is named SIGMA090806a.png.

Figures 6.4 and 6.5 are generated using different settings of parameters *first_hour*, *hours_in_plot*, and *plots_in_day*, see explanations at the end of Appendix 5.

6.6. Examples of processed measurements

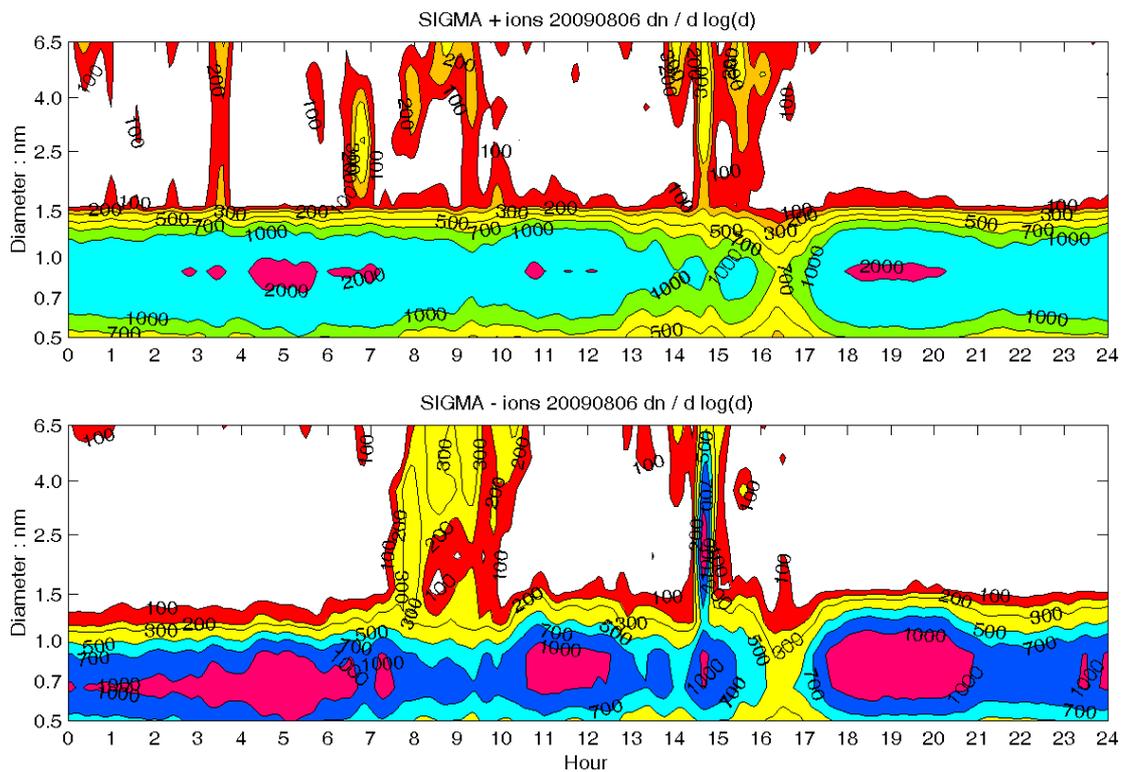


Figure 6.2. Diagram, corresponding to the total smoothing level 3 (see Sections 6.1–2). Measurements at Tammemäe, see A6.6. The fine details are lost due to the smoothing.

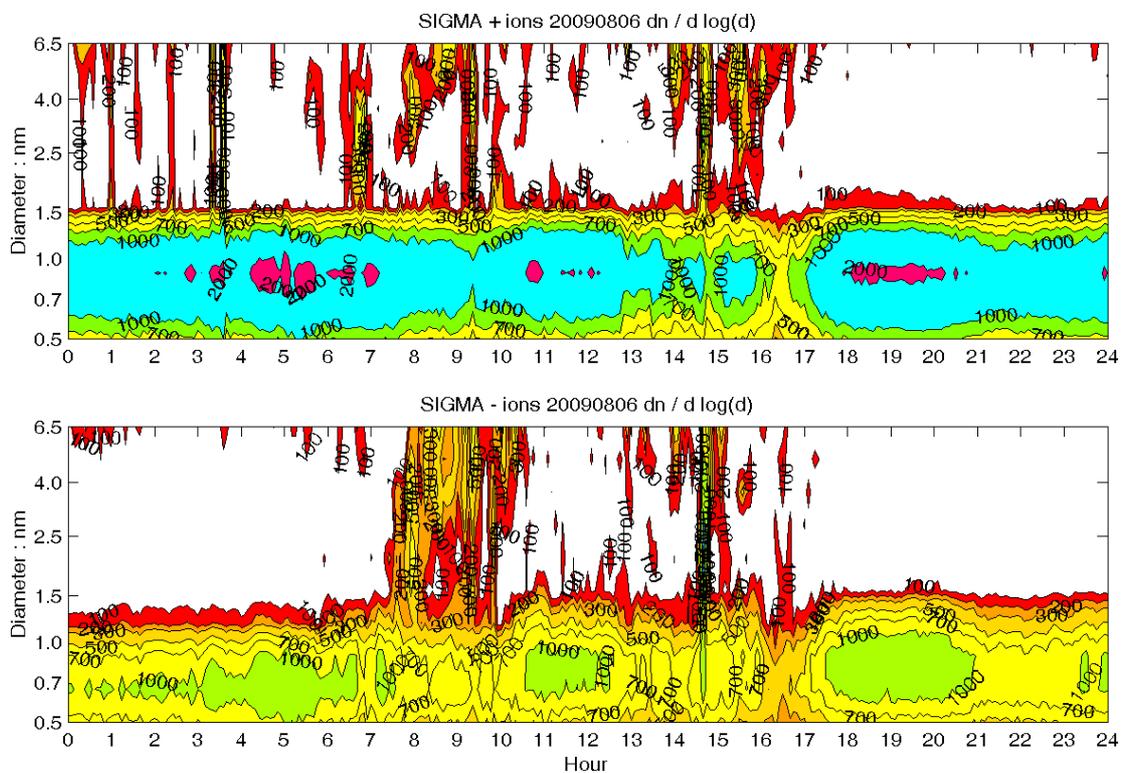


Figure 6.3. Diagram, corresponding to the total smoothing level –1. The fine details are formally preserved and the size of the file on the disk is 82 kB versus 56 kB of Figure 6.2. However, the details are barely visible due to the long time interval.

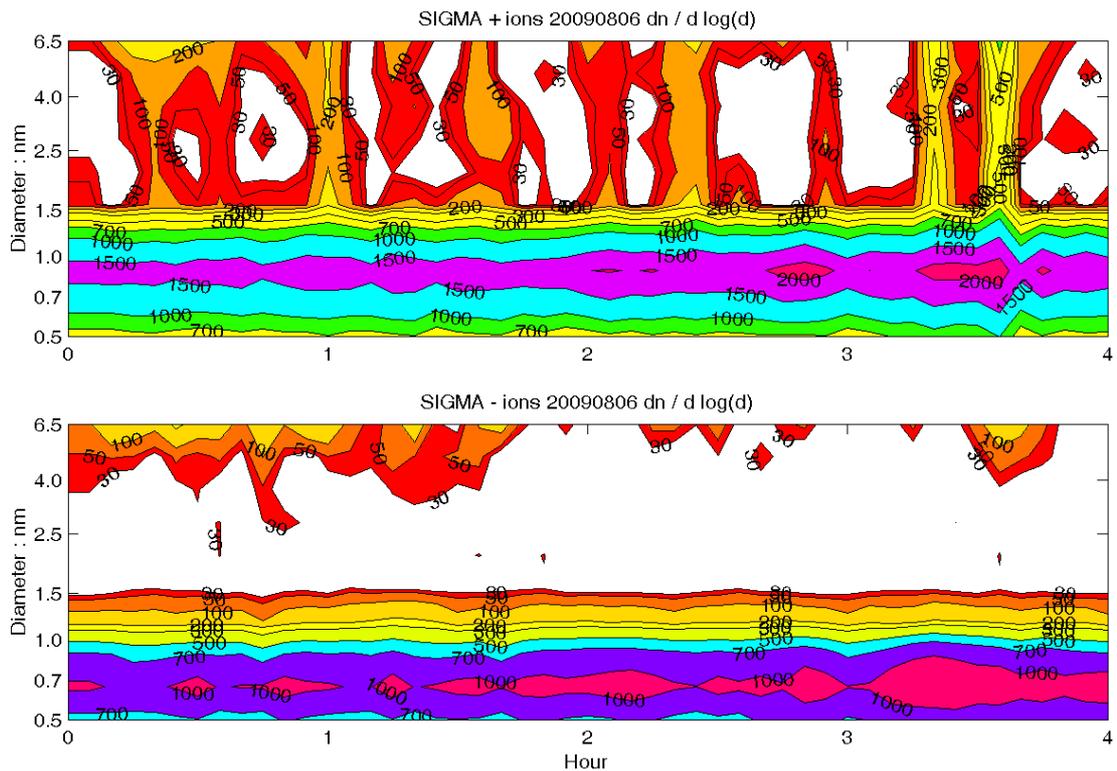


Figure 6.4. Diagram covering the first 4 hours. The levels were modified to make details visible: levels = [30 50 100 200 300 500 700 1000 2000 3000]. The night-time bursts of nanometer particles (see Junninen et al, 2008) caused by the positive ion-induced nucleation are much better visible when compared with the results obtained by the BSMA.

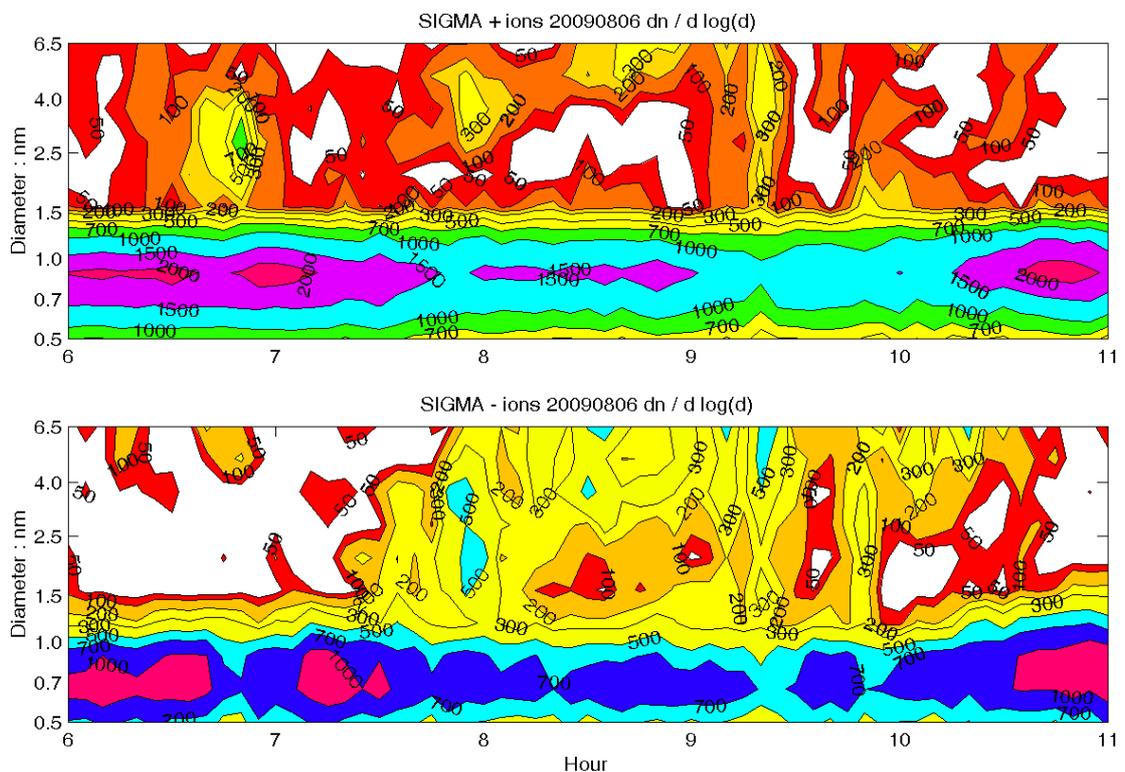


Figure 6.5. Diagram covering five morning hours. Note a weak extraordinary burst of positive intermediate ions of the size of about 2.5 nm during 06:30-07:00. The following burst of mostly negative intermediate ions 07:15-10:00 is a typical weak nucleation event.

APPENDICES

Appendix 1. Mobility versus size, temperature and pressure

Mobilities of 2 g/cm ³ particles at 950 mb							
d:nm	t=-20	t=-10	t=0	t=+10	t=+20	t=+30	t=+40
0.398	2.7573	2.8464	2.9357	3.0235	3.1098	3.1946	3.2781
0.501	2.2232	2.2878	2.3511	2.4132	2.4742	2.5342	2.5931
0.631	1.7725	1.8188	1.8643	1.9089	1.9527	1.9957	2.0379
0.794	1.3841	1.4177	1.4506	1.4829	1.5146	1.5457	1.5763
1.000	1.0545	1.0787	1.1025	1.1258	1.1487	1.1711	1.1932
1.259	0.7768	0.7925	0.8076	0.8222	0.8362	0.8497	0.8627
1.585	0.5176	0.5249	0.5320	0.5390	0.5457	0.5524	0.5589
1.995	0.3282	0.3336	0.3389	0.3442	0.3495	0.3547	0.3598
2.512	0.2174	0.2215	0.2256	0.2297	0.2336	0.2376	0.2414
3.162	0.1468	0.1497	0.1526	0.1554	0.1582	0.1609	0.1636
3.981	0.0989	0.1009	0.1028	0.1047	0.1065	0.1084	0.1102
5.012	0.0660	0.0673	0.0686	0.0699	0.0711	0.0723	0.0735
6.310	0.0437	0.0446	0.0454	0.0462	0.0470	0.0478	0.0486
7.943	0.0287	0.0293	0.0298	0.0303	0.0309	0.0314	0.0319
10.000	0.0188	0.0191	0.0195	0.0198	0.0201	0.0205	0.0208

Mobilities of 1 g/cm ³ particles at 1000 mb							
d:nm	t=-20	t=-10	t=0	t=+10	t=+20	t=+30	t=+40
0.398	3.1232	3.2241	3.3252	3.4246	3.5224	3.6185	3.7130
0.501	2.3779	2.4470	2.5147	2.5811	2.6464	2.7105	2.7735
0.631	1.8096	1.8569	1.9033	1.9488	1.9935	2.0374	2.0805
0.794	1.3692	1.4024	1.4350	1.4669	1.4982	1.5290	1.5593
1.000	1.0237	1.0473	1.0703	1.0929	1.1151	1.1369	1.1583
1.259	0.7464	0.7614	0.7760	0.7900	0.8034	0.8164	0.8289
1.585	0.4946	0.5016	0.5084	0.5151	0.5215	0.5279	0.5341
1.995	0.3128	0.3179	0.3230	0.3281	0.3331	0.3380	0.3429
2.512	0.2069	0.2108	0.2147	0.2186	0.2224	0.2261	0.2298
3.162	0.1396	0.1424	0.1451	0.1478	0.1504	0.1530	0.1556
3.981	0.0940	0.0959	0.0977	0.0995	0.1013	0.1030	0.1048
5.012	0.0628	0.0640	0.0652	0.0664	0.0676	0.0687	0.0699
6.310	0.0416	0.0424	0.0432	0.0439	0.0447	0.0455	0.0462
7.943	0.0273	0.0279	0.0284	0.0289	0.0293	0.0298	0.0303
10.000	0.0179	0.0182	0.0185	0.0188	0.0192	0.0195	0.0198

Mobilities of 2 g/cm ³ particles at 1000 mb							
d:nm	t=-20	t=-10	t=0	t=+10	t=+20	t=+30	t=+40
0.398	2.6198	2.7044	2.7892	2.8726	2.9546	3.0352	3.1145
0.501	2.1123	2.1737	2.2338	2.2928	2.3508	2.4078	2.4637
0.631	1.6841	1.7282	1.7713	1.8137	1.8553	1.8961	1.9363
0.794	1.3152	1.3470	1.3783	1.4089	1.4391	1.4686	1.4977
1.000	1.0020	1.0250	1.0476	1.0697	1.0914	1.1128	1.1337
1.259	0.7381	0.7530	0.7674	0.7812	0.7945	0.8074	0.8197
1.585	0.4918	0.4988	0.5056	0.5122	0.5186	0.5249	0.5310
1.995	0.3119	0.3170	0.3221	0.3271	0.3321	0.3371	0.3419
2.512	0.2066	0.2105	0.2144	0.2183	0.2220	0.2258	0.2294
3.162	0.1395	0.1423	0.1450	0.1477	0.1503	0.1529	0.1555
3.981	0.0940	0.0959	0.0977	0.0995	0.1013	0.1030	0.1047
5.012	0.0628	0.0640	0.0652	0.0664	0.0676	0.0687	0.0699
6.310	0.0416	0.0424	0.0432	0.0439	0.0447	0.0454	0.0462
7.943	0.0273	0.0278	0.0284	0.0289	0.0293	0.0298	0.0303
10.000	0.0179	0.0182	0.0185	0.0188	0.0192	0.0195	0.0198

Mobilities of 2 g/cm ³ particles at 1050 mb							
d:nm	t=-20	t=-10	t=0	t=+10	t=+20	t=+30	t=+40
0.398	2.4954	2.5759	2.6567	2.7361	2.8142	2.8910	2.9665
0.501	2.0121	2.0704	2.1277	2.1839	2.2391	2.2934	2.3467
0.631	1.6042	1.6461	1.6872	1.7276	1.7672	1.8061	1.8443
0.794	1.2528	1.2831	1.3129	1.3421	1.3707	1.3989	1.4266
1.000	0.9545	0.9764	0.9979	1.0190	1.0396	1.0599	1.0799
1.259	0.7032	0.7173	0.7310	0.7442	0.7569	0.7691	0.7808
1.585	0.4685	0.4752	0.4816	0.4879	0.4940	0.5000	0.5059
1.995	0.2971	0.3020	0.3069	0.3117	0.3164	0.3211	0.3257
2.512	0.1968	0.2006	0.2043	0.2079	0.2115	0.2151	0.2186
3.162	0.1330	0.1356	0.1382	0.1407	0.1432	0.1457	0.1481
3.981	0.0896	0.0914	0.0931	0.0948	0.0965	0.0981	0.0998
5.012	0.0598	0.0610	0.0622	0.0633	0.0644	0.0655	0.0666
6.310	0.0396	0.0404	0.0411	0.0419	0.0426	0.0433	0.0440
7.943	0.0261	0.0266	0.0270	0.0275	0.0280	0.0284	0.0289
10.000	0.0170	0.0174	0.0177	0.0180	0.0183	0.0186	0.0189

Appendix 2. Presentation of the mobility and size distributions

The concentration of ions in a narrow interval of the ion electric mobility dZ around the mobility Z is $dn = f_Z(Z) dZ$, where $f_Z(Z)$ is the distribution function of ions according to the mobility. The geometric size of an ion can be described by the ion mass diameter d (Tammet, 1995). The electric mobility depends on the mass diameter as $Z = \text{mob}_{\text{air}}(d, T, p)$, where mob_{air} is a specific function, T is the air temperature and p is the pressure. An approximation of the function mob_{air} proposed by Tammet (1995) is used in the SIGMA software. The size distribution of ions is described by the function $f_d(d) = dn / dd$, where dn is the concentration of ions in a narrow interval of the ion size dd . $dZ = (d \text{mob}_{\text{air}}(d) / dd) dd$ and

$$f_d(d) = \frac{dn}{dd} = \frac{d \text{mob}_{\text{air}}(d)}{dd} f_Z(Z). \quad (\text{A2.1})$$

An alternative is the presenting of distributions according to the decimal logarithms of the size and mobility. In this case $dn = f_{\log Z}(Z) d(\log Z) = f_{\log d}(d) d(\log d)$ and

$$f_{\log d}(d) = \frac{dn}{d(\log d)} = \frac{d \log(\text{mob}_{\text{air}}(d))}{d(\log d)} f_{\log z}(Z) = \frac{d}{Z} \frac{d \text{mob}_{\text{air}}(d)}{dd} f_{\log z}(Z). \quad (\text{A2.2})$$

All equations containing $\log d$ or $\log Z$ are conventional abbreviations, because a logarithm can be calculated only from dimensionless quantities. The strict way is to write $\log(d / d_a)$ instead of $\log d$, where d_a is an arbitrary fixed size. However, the differentials are independent of the choice of d_a , and an inconvenient full expression $d(\log(d / d_a))$ is abbreviated as $d(\log d)$.

Table A2.1. Units and conversion coefficients

Quantity	SI unit	Practical unit	Convert to SI
ion concentration n	m^{-3}	cm^{-3}	$\times 10^{-6}$
ion diameter d	m	nm	$\times 10^{-9}$
ion mobility Z	$\text{m}^2 \text{V}^{-1} \text{s}^{-1}$	$\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$	$\times 10^{-4}$
$f_d(d)$	m^{-4}	$\text{cm}^{-3} \text{nm}^{-1}$	$\times 10^{-15}$
$f_{\log d}(d)$	m^{-3}	cm^{-3}	$\times 10^{-6}$
$f_Z(Z)$	$\text{m}^{-1} \text{Vs}$	$\text{cm}^{-1} \text{Vs}$	$\times 10^{-1}$
$f_{\log Z}(Z)$	m^{-3}	cm^{-3}	$\times 10^{-6}$

Units of the considered quantities and the conversion coefficients are shown in Table A2.1.

The control program SIGMA1A records the electrometric current as an average through the mobility and size fractions, see Figure A2.1. The geometric centers are the reference values of mobility or size when presenting a distribution function. The "decade to 16" and "decade to 8" logarithmic fraction schemes are used where the fraction borders are set according to a geometric sequence with the factor of $10^{1/16} \approx 1.15478$ or $10^{1/8} \approx 1.33352$.

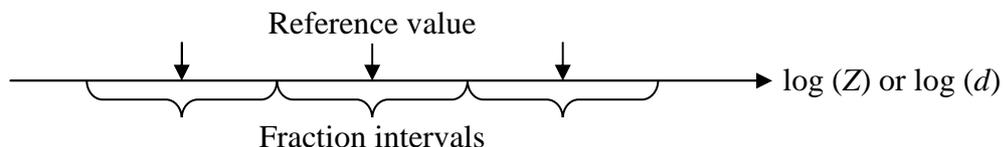


Figure A2.1. Fraction intervals and reference values of mobility or size.

The SIGMA output files include the distributions that are always presented with the values of the logarithmic distribution. The display in the computer screen is flexible as the control file SIGMA1A.INI allows to choose the display of the values of the distribution functions or the display of fraction concentrations. A mobility fraction concentration in an interval (Z_1, Z_2) is

$$n(Z_1, Z_2) = \int_{Z_1}^{Z_2} f_z(Z) dZ = \int_{\log Z_1}^{\log Z_2} f_{\log z}(Z) d(\log Z) \quad (\text{A2.3})$$

The size fraction concentrations can be expressed in similar way via the functions of the size distribution. A fraction concentration is estimated as the product of the logarithmic distribution function in the center of the fraction and the logarithmic width of the fraction, which is usually 1/8. In this case the conversion from the fraction presentation to the distribution function presentation is made according to the approximate equation

$$f_{\log Z}(\sqrt{Z_1 Z_2}) \approx 8n(Z_1, Z_2) \quad \text{and} \quad f_{\log d}(\sqrt{d_1 d_2}) \approx 8n(d_1, d_2) \quad (\text{A2.4})$$

An exception is the mobility distribution in the cluster ion regime where the logarithmic width of the fraction is 1/16.

Table A2.2. Size fractions and the corresponding electric mobilities in typical and extreme conditions

d : nm		Z : $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$		Z : $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$	
border	center	-20 C & 1050 mb		10 C & 1000 mb	
0.422		2.3646		2.7158	3.0916
	0.487		2.0676		2.6706
0.562		1.8005		2.0429	2.3022
	0.649		1.5573		1.7593
0.750		1.3356		1.5038	1.6840
	0.866		1.1347		1.2743
1.000		0.9545		1.0697	1.1932
	1.155		0.7937		0.8865
1.334		0.6432		0.7110	0.7814
	1.540		0.4960		0.5426
1.778		0.3717		0.4071	0.4456
	2.054		0.2816		0.3104
2.371		0.2175		0.2409	0.2662
	2.738		0.1698		0.1885
3.162		0.1330		0.1477	0.1636
	3.652		0.1040		0.1155
4.217		0.0811		0.0900	0.0997
	4.870		0.0630		0.0699
5.623		0.0488		0.0541	0.0598
	6.494		0.0376		0.0417
7.499		0.0290		0.0321	0.0354

The control program SIGMA1A is designed with the aim of measuring concentrations of 10 size fractions, the borders and centers of which are shown in Table A2.2. The boundaries of the mobility fractions are arranged according to the "decade to 8" or "decade to 16" scheme shown in Tables 5.2 and 5.3. The boundary electric mobilities in Table A2.2 are not arranged according to the "decade to 8" scheme and depend on the air temperature and atmospheric pressure.

Appendix 3. Inlet loss of ions

A3.1. General considerations

Some amount of ions is lost in the inlet tract before entering the mobility classifying zone (von der Weiden et al., 2009). The effect of electrostatic image forces on the deposition of ions is negligible when compared with the diffusion (Tammert, 1970), and the inertial deposition is not important because the size range of the SIGMA is limited to fine nanometer particles. The ion concentration is limited by technical constraints of the instrument in the range, where the space charge effects can be neglected. Simple geometry of the inlet enables estimating the diffusion losses directly using the well-proven semiempirical methods of the heat transfer theory. In case of the ion and nanometer aerosol particles the preconditions required for the full analogy of heat and mass transfer (Kays et al., 2005, see Chapter 18) are satisfied and the equations of the heat transfer to the surface of constant temperature can be used when replacing the Nusselt number Nu with the Sherwood number Sh and the Prandtl number Pr with the Schmidt number Sc (Incropera and Dewitt, 2002).

The diffusion deposition flux of ions onto a flat surface is $q_{dep} = WLn u_{dep}$, where W is the width, L is the length of the surface along the air flow, n is the undisturbed concentration of ions and u_{dep} is the mean deposition velocity along the length L . The mean deposition velocity is

$$u_{dep} = (D/L)Sh_{mL}, \quad (A3.1)$$

where Sh_{mL} is the mean Sherwood number related to the length L , and D is the ion diffusion coefficient. The air enters as a plug flow and the boundary layer over the surface will develop along the channel. The displacement thickness of the boundary layer is estimated as

$$\delta_{disp} = 1.72 \sqrt{\frac{\nu L}{u_{flow}}}, \quad (A3.2)$$

where ν is the kinematic viscosity of the air and u_{flow} is the free flow velocity (Incropera and Dewitt, 2002). A numeric calculation shows that the displacement thickness in the SIGMA inlet grid and filter is much less than the distance between the surfaces. Thus a semiempirical equation obtained for the boundary layer over a flat plate (Incropera and Dewitt, 2002) can be adopted in our problem:

$$Sh_{mL} = 0.664 Re_L^{1/2} Sc^{1/3}. \quad (A3.3)$$

The equation above takes into account the entrance profiles for both the ion concentration and for the air flow velocity. The Reynolds number related to the same length L and the velocity u_{flow} as the Sherwood number is

$$Re = \frac{u_{flow} L}{\nu}. \quad (A3.4)$$

Equation (A3.3) is valid when $Sc > 0.6$ (Incropera and Dewitt, 2002). The Schmidt number $Sc = \nu / D$ has the lowest value of about 3 in the case of cluster ions. Thus the concentration boundary layer is always thinner than the velocity boundary layer and Equation (A3.3) can be used for the evaluation of the ion deposition without restrictions so far as the boundary layer remains thin enough. The diffusion coefficient of single charged ions of the electric mobility Z is

$$D = \frac{kTZ}{e}, \quad (A3.5)$$

where k is the Boltzmann constant, T is the absolute temperature, and e is the elementary charge. The composite equation is

$$u_{dep} = \frac{kTZ}{Le} 0.664 \left(\frac{u_{flow} L}{\nu} \right)^{1/2} \left(\frac{\nu e}{kTZ} \right)^{1/3} = 0.664 (k/e)^{2/3} L^{-1/2} u_{flow}^{1/2} \nu^{-1/6} T^{2/3} Z^{2/3}. \quad (A3.6)$$

A3.2. Deposition of ions onto the inlet grid and the parallel plate filter

The inlet grid is made of a perforated sheet and the air enters through cylindrical channels. The inlet flux of the air is $\pi R^2 u_{flow}$ and the loss flux is $2\pi R L u_{griddep}$. Thus the relative loss is

$$A_{grid} = \frac{L_{grid} u_{griddep}}{2R u_{gridflow}} = 0.332(k/e)^{2/3} L_{grid}^{1/2} R^{-1} u_{gridflow}^{-1/2} v^{-1/6} T^{2/3} Z^{2/3}. \quad (A3.7)$$

The gate filter is a flat channel between two parallel plates separated with a distance h . The inlet flux of the air is Whu_{flow} and the flux of the loss onto both surfaces is $2WL u_{gatedep}$. The relative loss is

$$A_{gate} = \frac{2WL_{gate} u_{gatedep}}{Whu_{gateflow}} = 1.33(k/e)^{2/3} L_{gate}^{1/2} h^{-1} u_{gateflow}^{-1/2} v^{-1/6} T^{2/3} Z^{2/3}. \quad (A3.8)$$

A3.3. Correction of concentration according to the inlet loss

In the control program of the BSMA, the values of the distribution function or the fraction concentration f were corrected assuming a very low adsorption (Tammet, 2006):

$$f_{corrected} = \frac{f_{uncorrected}}{(1 - A_{grid})(1 - A_{gate})}, \quad (A3.9)$$

In the control program of the SIGMA the next approximation is used, where $1-A$ is replaced by $\exp(-A)$. The result is

$$f_{corrected} = f_{uncorrected} \exp(A_{grid} + A_{gate}) \quad (A3.10)$$

Let the index 0 mark the standard values of the parameters $T_0 = 273$ K, $p_0 = 1013$ mb, $Z_0 = 1$ cm²V⁻¹s⁻¹, and $v_0 = 1.33 \times 10^{-5}$ m²/s. The dependence of the deposition velocity on the air viscosity is weak and a rough approximation is sufficient:

$$v = v_0 \left(\frac{T}{T_0} \right)^{5/3} \left(\frac{p}{p_0} \right)^{-1}, \quad (A3.11)$$

If the deposition velocity at standard conditions is u_{dep0} , then

$$u_{dep} = u_{dep0} (T/T_0)^{7/18} (p/p_0)^{1/6} (Z/Z_0)^{2/3}. \quad (A3.12)$$

The estimate of the total relative adsorption $A = A_{grid} + A_{gate}$ is now calculated as:

$$A = A_0 (T/T_0)^{7/18} (p/p_0)^{1/6} (Z/Z_0)^{2/3}, \quad (A3.13)$$

where $A_0 = c(0.332L_{grid}^{1/2}R^{-1}u_{gridflow}^{-1/2} + 1.33L_{gate}^{1/2}h^{-1}u_{gateflow}^{-1/2})$, (A3.14)

and $c = (k/e)^{2/3} v_0^{-1/6} T_0^{2/3} Z_0^{2/3} = 0.0114$. (A3.15)

The air flow rate in SIGMA is about 34000 cm³/s. The parameters of the grid are $L = 0.7$ mm, $R = 0.5$ mm, full area 216 cm², and the area of holes 99 cm². It follows the air speed about 3.4 m/s. The parameters of the gate are $L = 123$ mm, $h = 10$ mm. The area of the free section between the inlet filter plates is 177 cm² and the corresponding average linear velocity is 1.92 m/s. The distance between the plates of the gate exceeds the distance between filter plates (which is 4 mm) and the air speed is increased in the gate up to the estimated value of 2 m/s. Calculation yields the estimates for SIGMA:

$$A_{0grid} = 0.011, \quad A_{0gate} = 0.038, \quad A_0 = 0.049. \quad (A3.16)$$

The relative uncertainty of the fraction concentrations induced by the uncertainty of the 5% correction is about 20 times less than the uncertainty of the estimate of the relative adsorption.

Appendix 4. Control program SIGMA1A

A4.1. Outline of the program

The control program SIGMA1A.EXE is compiled from the PASCAL-source SIGMA1A.DPR using DELPHI6 and it works under MS Windows (version at least XP SP2) in the console regime. The listing of the source code is available as a separate document.

It is expected below that the active folder is the SIGMA folder, which contains the file SIGMA1A.EXE. When SIGMA1A is launched, it looks in the SIGMA folder for three files: SIGMA1A_simulator.txt, SIGMA1.CAL, and SIGMA1A.INI. The first file is optional and should not be present during real measurements, see explanations below in Section A4.4. The calibration file SIGMA1.CAL and the control file SIGMA1A.INI are obligatory. If any of these two files is not available, the program delivers a corresponding message and stops.

After successful reading of the calibration file and the control file the program displays the start menu, shown in Figure 3.1. The instruction of the practical usage of the control program during routine measurements is presented in Chapter 3 and the test operations are described in Section 4.6 of the manual. The present appendix consists of the explanation of the principles of the measuring algorithm. The details of the algorithm are available in the source code of the control program.

When SIGMA1A encounters a failure, it creates an additional file SIGMA1A_failure.txt in the SIGMA folder, recording the time and a brief explanation of the failure. As a rule, the program will be stopped for 10 minutes after a failure and it will try to continue afterwards. If the file SIGMA1A_failure.txt already exists, then a new failure message will be appended to the existing file.

A4.2. Calibration file and control file

The calibration constants are presented in the plain text file SIGMA1A.CAL. When making any changes, the *date in the heading of the file must be corrected*. Otherwise the control program interprets the change as a corruption of the file. The content of the calibration file version 20101026 is:

```
SIGMA1A.CAL 20101026
  Includes calibration coefficients.
  Any line beginning with a space is a comment.
  An assignment should be written without spaces.
  A space after the assignment starts a comment until the end of the line.
  Initial text of SIGMA1A.CAL is attached to the end of SIGMA1A.DPR
volt_factor=885           for voltage-mobility conversion (Q = 34 l/s)
conc_factor_pos=11.5     for ADC-dn/dlogZ conversion
conc_factor_neg=11.5     for ADC-dn/dlogZ conversion
standardadsorption=0.0505 for Z = 1 cm2V-1cm-1, 0 C, 1013 mb
collectormobility=0.04   cm2V-1cm-1, effective value for corrections
c_supplyvolt=0.002167    supplyvoltage / ADC_counts
c_filtervolt=-0.089      filter voltage / ADC_counts
c_batteryvolt=0.0093     electrometer battery voltage / ADC_counts
c_bias=0.003052          electrometer inlet mV at 1 ADC, 0.1526 / 50
c_pressurea=0.03368      pressure (mb) = c_pressurea * ADCcounts + c_pressureb
c_pressureb=113
c_temperaturea=0.01083   Celsius = c_temperaturea * ADCcounts + c_temperatureb
c_temperatureb=-290     factory value is -273
c_humiditya=0.00607     humidity (%) = c_humiditya * ADCcounts + c_humidityb
c_humidityb=-35         required correction RH = RH (1.0546 - 0.0216 T:C)
electrometerdelay=385   ms, incl. airflow + electrometer - HV signal
chargingtime=1500      ms
timeout=30000          ms
c_inv_n2=-0.16          inverter parameter
c_inv_n1=-0.22          inverter parameter
c_inv_p1=-0.10          inverter parameter
c_inv_p2=-0             inverter parameter
z_limit=1              inverter parameter
```

If any of the inverter parameters is not shown in the file then the default values are used: `c_inv_n2=0`, `c_inv_n1=0`, `c_inv_p1=0`, `c_inv_p2=0`, `z_limit=1`. If another parameter is missing in the calibration file, then SIGMA1A issues a failure message and stops.

The current values of calibration constants will be saved in the headers of the output files of the SIGMA.

The user-editable control constants are presented in the plain text file SIGMA1A.INI. When making any changes, the date in the heading of the file must be corrected. The version 20101022 of SIGMA1A.INI is as follows:

```
SIGMA1A.INI 20101022
Includes custom controls.
Any line beginning with a space is a comment.
An assignment should be written without spaces.
A space after the assignment starts a comment until the end of the line.
Initial text of SIGMA1A.INI is attached into the end of SIGMA1A.DPR
cboard=0          defined during the USB1608 initialization
clusterregime=0   0 = dn/dlogZ full range 8 fr/dec
                  1 = dn/dlogZ only clusters 16 fr/dec,
extracorrect=2    correct: 0 = no, 1 = dust pulses, 2 = transfer, 3 = both
cycleminutes=5    1, 2, 3, 4, 5, 6, or 10, standard value is 5
timezone=2        for time presentation
showfractions=1  0 = show dn/dlog, 1 - show fraction n (save still dn/dlog)
extension=x1      extension of the output file name, usually txt or xl
extrapath=***     for extracopy of midnight tables, *** means no extracopy
controls=1011    four symbols 0 or 1 setting initial values of saving
                  controls: internal, external, diatable, scandetails
extrapath can be written with or without final \
a sample: extrapath=J:\SIGMA
```

Clusterregime is explained in Section 5.5.

Extra corrections are explained in Section 5.7.3.

Extrapath is essential when the *extrafolder* regime is turned on. The regime can be turned on and off during the measuring process by means of control keys, indicated on the computer screen, see Section 3.2. *Extrapath* can be directed to a shared folder or a removable media.

WARNING: if the measurements are restarted after making changes in the calibration file or the control file, the headers of the recurrent output files remain unchanged. Follow recommendations presented in Section 5.7.1.

A4.3. Structure of the measuring process

During the measuring process, scans of the mobility distribution are performed in a cycle, which can be interrupted only pressing the key combination XZ. Simplified structure of the measurements is:

```
repeat scan
  if voltageperiod full then measure voltages
  if cycleperiod full then process and save cycle
  if new day then append monthly file and save diagram table
until XZ pressed;
```

In the beginning of every scan the HV capacitors are charged and the voltage is increased up to about 3000 V for each polarity during about 1.5 s. Next, the voltage will exponentially decline in the relaxation process of a RC circuit with a time constant of about 3.8 s down to about 20 V, which covers the full size and mobility range of ions. Measurements are made during the period of the voltage decrease. Usually, three scans are made during every minute. Some of these scans are performed with an open air ion inlet gate and some with a closed air ion inlet gate. The gate (see Figure 2.1 and Appendix 7) is controlled by switching the voltage between the gate plates between zero and 520 V. The period of switching depends on the parameter *zeroperiod*, which is initiated as an internal constant in the program. In case of the

standard value $zeroperiod = 3$, the sequence of closed gate (0) and open gate (1) scans is 011011011...

The full mobility range of $0.0274...4.217 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ is divided into 35 fine mobility fractions. Before every scan, the air temperature and pressure are measured and the voltages corresponding to the boundaries between fine mobility fractions are calculated. During a scan, the voltages of electrodes and electrometer outputs are measured about 100 times per second until the voltage reaches the value corresponding to the highest mobility of $4.22 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$. Next, the collected data is distributed into 35 fine mobility fractions and the averages are calculated for every fraction. When calculating the averages, the weak trimming is applied discarding the smallest and the largest readings of every sample. The results of a scan are collected into an array called *scanline*. Composition of this array corresponds to the composition of a line of the scan data file explained in Section 5.3. Additionally, the zero element is included: *scanline* [0] equals to the wintertime in MS Windows format referring to the midpoint between the start and finish of the scan. The following elements are the same as listed in Section 5.3.

At the end of a scan, the scanline is written into a circular buffer *scanbuffer* and saved (except the zero element) on demand into the file of scan details as a string of integer numbers. The pointer of *scanbuffer* is denoted as *scanpoint* and it is incremented up and down as $scanpoint := (scanpoint + 1) \bmod scanmax$ and $scanpoint := (scanpoint + scanmax - 1) \bmod scanmax$.

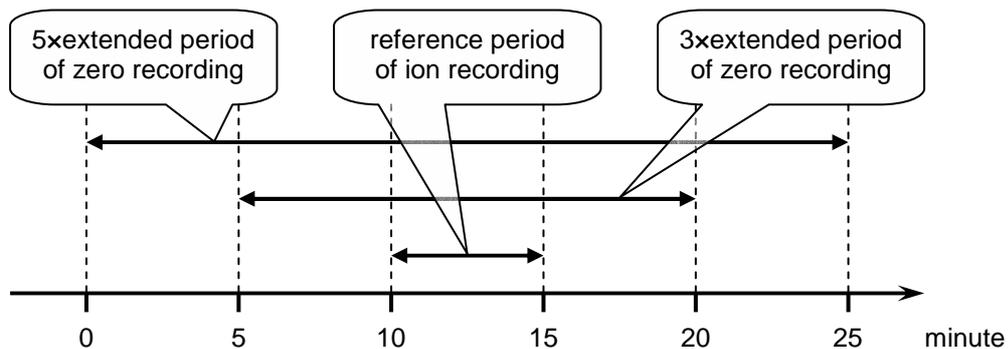


Figure A4.1. The periods of estimating the signal and zero in case of 5-minute cycles. The recorded ion measuring interval is marked with minute = 12 in the output files.

When the cycle is full, the buffered data are processed. The signal of an electrometer is composed of the signal of ions to be measured, the zero level of the electrometer, and the signal of the large ions passed through the sheath air filter (see Figure 2.1). Switching off the inlet gate affects only the signal of ions to be measured. When processing the cycle data, the average closed gate signal is subtracted from the open-gate signal. The drift of the closed-gate signal is usually slow compared with the variation of the open-gate signal. Thus the period of averaging the closed-gate signal is elongated with aim of suppressing the random noise as explained in Figure A4.1.

The statistical distribution of the noise of the electrometric signal is not Gaussian. The heavy-tailed shape of the distribution is caused by several factors, especially by the outliers generated by coarse aerosol particles. Thus the method of interquartile trimmed mean is applied when processing the buffered data. When calculating the trimmed mean of n repeated scans, the g smallest and g largest entries are discarded and the mean is estimated as the arithmetic average of $n - 2g$ middle entries of the sample. SIGMA1A applies the trimming degree $g = (n + 1) \text{ div } 4$.

The noise level is estimated simultaneously for two ion polarities when analyzing the results of zero scans. The method is based on calculating the deviations from the cubic interpolation of the zero signals of the 16 mobility fractions. The deviations are calculated as

$$\Delta x_i = x_i - (x_{i-2} - 4x_{i-1} - 4x_{i+1} + x_{i+2})/6, \quad i = 3 \dots 14. \quad (\text{A4.1})$$

A4.4. Comments for the software developer

The program SIGMA1A is written as a console application, which helps to keep the well-accustomed BSMA-style user interface. The console output is localized into particular procedures, whose names begin from the word "show". This makes a future reorganization of the program into a GUI application relatively easy.

The program includes a simple mechanism to help development and testing. The user interface, internal data processing and data output can be quickly tested without a connection to a real instrument. This requires including an extra file SIGMA1A_simulator.txt into the SIGMA folder. An example of the content of this file is:

```

100  simufactor, how many times the clock is accelerated
2220 start of the simulated clock, hhmm
1    spectrumstyle, 0 = flat dn/dlogZ, 1 = like natural
30   simunoisesigma, noise sigma expressed in ADC counts
0.01 simuoutlierprobability
300  simuoutliersigma in ADC counts

```

The six lines of this file must be written strictly in the same order as in the example; the explanations after the spaces are for humans and will not be analyzed by the computer.

If the extra file SIGMA1A_simulator is found, then a logical variable *simulator* is assigned *true* and the real measurements are replaced by the output of the function *ADCsimulator*. Search the program code for the phrase "**if simulator**" to see the differences compared with the normal regime.

Warning: too high value of *simufactor* may lead to a situation, when the computer is not able to complete some procedures in time. In case of a 2 GHz processor the troubles are probable when *simufactor* > 100. It is recommended to close resource-consuming side processes when working with high value of *simufactor*.

The logical constant *developer* defined in line 9 of the program code is an additional tool. If this constant is assigned *true*, then three additional commands will be available. In the main menu, an additional command *Z* appears, which launches a user-written procedure *Z_test*. Two additional commands *A* and *B* appear in the test menu, which allow launching the user-written procedures. In the present version of SIGMA1A, two developer procedures *electrometertransition1* and *electrometertransition3* are retained as examples.

Appendix 5. MATLAB function SIGMA1D.m

The text of the function can be copied into the clipboard and pasted into the Notepad, M-editor, or immediately into the MATLAB command window. The user should arrange the files and edit the control lines before launching the function as explained in Section 6.5.

```

% SIGMA1D - creates contour plots using SIGMA diatable files - HT20111222
%=====
% Permanent sample of control lines, please keep unmodified:
% prefix = 'SIGMA'; % for output file name
% filepath = 'C:\SIGMA\diatables!'; % where the files are located
% yy = '11'; % year
% months = 1:12; % interval of months, can be restricted
% days = 1:31; % interval of days, can be restricted
% first_hour = 0; % integer 0...23
% hours_in_plot = 8; % integer 1...24
% plots_in_day = 3; % (first_hour + plots_in_day * hours_in_plot) should not exceed 24!
% first_mark = 1; % number of letter in the filename, 0 = nothing, 1 = a, 2 = b, ...
% levels = [50 100 200 300 500 700 1000 1500 2000 3000 5000]; % for dn/dlogd
% pos_axes = [50 320 700 200]; % left, bottom, width, height

```

```

% neg_axes = [50 60 700 200];
% y_ticknumbers = [1 2.18 3.43 4.85 6.65 8.3 10];
% y_ticklabels = {'0.5';'0.7';'1.0';'1.5';'2.5';'4.0';'6.5'};
% resolution = '-r150'; % pixels per inch, screen 100, report 200, publishers 600
% brightness = 0.5; % -1..+1, -1 = dark, 0 = normal, +1 = bright
%=====
% Examples of controls for covering full day with diagrams:
% first_hour = 0; % in all three examples
% hours_in_plot = 24; % one plot per day
% plots_in_day = 1;
% hours_in_plot = 8; % three plots per day
% plots_in_day = 3;
% hours_in_plot = 3; % eight plots per day
% plots_in_day = 8;
%=====
% The control lines to be modified by the user (the permanent sample is preserved above):
prefix = 'SIGMA'; % for output file name
filepath = 'C:\test\'; % where the files are located
yy = '11'; % year
months = 1:11; % interval of months, can be restricted
days = 1:31; % interval of days, can be restricted
first_hour = 0; % integer 0...23
hours_in_plot = 24; % integer 1...24
plots_in_day = 1; % (first_hour + plots_in_day * hours_in_plot) should not exceed 24!
first_mark = 0; % number of extra letter, 0 = nothing, 1 = a, 2 = b, ...
levels = [50 100 200 300 500 700 1000 1500 2000 3000 5000]; % for dn/dlogd
% The controls, which could be modified only in special situations:
pos_axes = [50 320 700 200]; % left, bottom, width, height
neg_axes = [50 60 700 200]; % width (standard is 700) may be sometimes modified
y_tickpositions = [1 2.18 3.43 4.85 6.65 8.3 10]; % vertical positions
y_ticklabels = {'0.5';'0.7';'1.0';'1.5';'2.5';'4.0';'6.5'}; % nanometers
resolution = '-r150'; % pixels per inch, from 100 (screen) to 600 (publishers)
brightness = 0.8; % -1..+1, -1 = dark, 0 = normal, +1 = bright
%=====
% Modification of the text below of this line is not recommended
% set(0,'DefaultAxesColorOrder',[0 0 0]);
for month = months;
    mm = num2str(month);
    if month < 10
        mm = ['0' mm];
    end;
    for day = days;
        dd = num2str(day);
        if day < 10
            dd = ['0' dd];
        end;
        if exist([filepath 'd' yy mm dd '.xl'], 'file')
            xx = load([filepath 'd' yy mm dd '.xl']);
            [rows, nd] = size(xx);
            cycles = (rows - 1) / 24;
            for turn = 0:(plots_in_day - 1); % day sectors a, b, c, ...
                a = 1 + cycles * (first_hour + turn * hours_in_plot); % first row in data table
                b = first_hour + turn * hours_in_plot; % first hour of the plot
                mark = char(96 + first_mark + turn); % extra letter to distinguish the files
                if mark == char(96)
                    mark = ''; % filename will be without an extra letter
                end;
                x = xx(a : a + cycles * hours_in_plot, 2:11); % sector of positive ions
            end;
        end;
    end;
end;

```

```

if sum (sum (x)) > 9999 % empty diagram is skipped
  axes ('units', 'pixels', 'position', pos_axes);
  [c,h] = contourf (x', levels);
  clabel (c, h);
  set (gca, 'xtick', [1:cycles:rows], 'xticklabel', [b:1:b+hours_in_plot]);
  set (gca, 'ytick', y_tickpositions);
  set (gca, 'yticklabel', y_ticklabels);
  ylabel('Diameter : nm');
  title(['SIGMA + ions 20' yy mm dd ' dn / d log(d)']);
  colormap(hsv);
  brighten (brightness);
  print (resolution, '-dpng', [filepath prefix yy mm dd mark '.png']);
end; % if sum > 9999
x = xx (a : a + cycles * hours_in_plot, 12:21); % sector of negative ions
if sum (sum (x)) > 9999 % empty diagram is skipped
  axes ('units', 'pixels', 'position', neg_axes);
  [c,h] = contourf (x', levels);
  clabel (c, h);
  set (gca, 'xtick', [1:cycles:rows], 'xticklabel', [b:1:b+hours_in_plot]);
  set (gca, 'ytick', y_tickpositions);
  set (gca, 'yticklabel', y_ticklabels);
  xlabel('Hour');
  ylabel('Diameter : nm');
  title(['SIGMA - ions 20' yy mm dd ' dn / d log(d)']);
  colormap(hsv);
  brighten (brightness);
  print (resolution, '-dpng', [filepath prefix yy mm dd mark '.png']);
end; % if sum > 9999
close all;
end; % of turn
end; % of if exist
end; % of days
end; % of months and finish of processing

```

Some explanations:

The typical settings of the time parameters are written above. It follows one full day diagram per day like presented in Figures 6.2 and 6.3. The name of the output file in Figure 6.2 is SIGMA090806.png and the file occupies 56 kB on the computer disk. Figure 6.3 is compiled for the same day and thus it is recommended to set a different value of *first_mark*. If *first_mark* = 1 then the diagram file is named SIGMA090806a.png.

The settings:

```

first_hour = 0; % integer 0...23
hours_in_plot = 8; % integer 1...24
plots_in_day = 3; % (first_hour + plots_in_day * hours_in_plot) should not exceed 24!
first_mark = 2; % number of extra letter, 0 = nothing, 1 = a, 2 = b, ...

```

generate three diagrams per day covering the hour intervals 0–8, 8–16, and 16–24 and named SIGMA090806b.png, SIGMA090806c.png, and SIGMA090806d.png.

Covering of a full day in a diagram is not a must. Example: if you will set *first_hour* = 4, *hours_in_plot* = 3, and *plots_in_day* = 2 then you will get two diagrams per day covering hours 4–7 and 7–10.

Appendix 6. Calibration and testing the instrument

A6.1. Reference mobility and analyzer voltage

The characteristic mobility of an aspiration condenser is proportional to the air flow rate and inversely proportional to the voltage between the attracting and repelling electrodes:

$$Z = \text{const} \times Q / V. \quad (\text{A6.1})$$

The voltage V is proportional to the measured high voltage signal ADC_{HV} (ADC counts) and the flow rate in the SIGMA is proportional to the power supply voltage U . Thus the required value of HV signal is

$$\text{ADC}_{\text{HV}0} = \text{VC} \times (U/U_0) \times Q_0 / Z_0, \quad (\text{A6.2})$$

where Z_0 is the required reference mobility used as the argument of the distribution function, Q_0 is the standard flow rate at the standard power supply voltage U_0 and VC is the voltage calibration constant. The mobility calibration of the instrument requires determining of the calibration constant VC and the standard flow rate Q_0 .

The calibration constant VC is calculated solving the Laplace equation for the electric field in the condenser and computing the ion trajectories when passing the condenser (see Figure 2.1 and Appendix 7). The result is

$$\text{VC} = 0.02855.$$

The air flow rate is measured using the rotary gas flow meter PC-400, the measuring range of which is up to $111 \text{ dm}^3 \text{ s}^{-1}$. The flow meter counts the fixed-volume air parcels passed through the instrument and its calibration is expected to change only as a result of a mechanical damage. The increased friction of the rotor is usually the first indication of wear. The time of free inertial rotating of about 100 s validates the good condition of the instrument. The SIGMA air flow is measured using the compensation method: the air is blown into a large box through the flowmeter by means of an extra fan, the flow is controlled by a mechanical slide, and sucked out from the box by the SIGMA. One side of the box is made of thin plastic film, which helps precisely to balance the pressure in the box by adjusting the slide. The result at the voltage $U = 23.4 \text{ V}$ is 34.4 liters per second. Former experiments with the same flowmeter have shown that the readings exceed the actual flowrate by about 2%. Thus the value of the flowrate at the standard supply voltage 23.5 V is estimated in the calibration of 20100323:

$$Q_0 = 34000 \text{ cm}^3/\text{s}.$$

The calibration constants were verified by means of the iodine test. The admixture of iodine vapor in the clean air forms negative ions of the mobility about $1.85\text{--}1.90 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ (Tammet, 1975). The control experiments in iodine containing laboratory air were in satisfactory accordance with this value.

The calibration constant VC and the standard flow rate Q_0 are never independently used in the control program, where only the product $\text{VF} = \text{VC} \times Q_0 = 985$ is necessary. This product is called the voltage factor and included into the calibration file. The ADC counts corresponding to the mobility values are calculated in the program as

$$\text{ADC}_{\text{HV}0} = (U/U_0) \times \text{VF} / Z_0. \quad (\text{A6.3})$$

A6.2. Air ion concentration: theoretical approach

The collector current $I(Z_0)$ of monomobile ions of mobility Z and concentration n at the reference mobility of the analyzer Z_0 is $I(Z_0) = eQ_c g(Z_0, Z)n$, where e is the elementary charge, Q_c is the air flow rate through the collector and $g(Z_0, Z)$ is the normalized transfer function, which values are in the range of (0...1). If the ions are distributed according to the mobility as $f(Z) = dn/dZ$, then $I(Z_0) = eQ_c \int g(Z_0, Z) f(Z) dZ$. Let us consider the ions be distributed according to a fictive flat distribution and $f(Z) = c$. In this case the collector current is $I(Z_0) = ceQ_c \int g(Z_0, Z) dZ$. In a special situation of rectangular transfer function when $g = 1$ in a range ($Z_a \dots Z_b$) of mobility and $g = 0$ elsewhere, the integral $\int g(Z_0, Z) dZ = Z_b - Z_a$. In the general

situation, the integral $\int g(Z_0, Z)dZ$ is called the effective width of the transfer function and denoted ΔZ_g . In case of a triangular transfer function the effective width equals to the half-height width of the triangle. As a conclusion we get in case of the flat distribution $f(Z) = c$ the flat value of the collector current $I = ceQ_c\Delta Z_g$ and the distribution function can be calculated according to the measured current as $c = I/eQ_c\Delta Z_g$. A fraction concentration of ions in a range of $(Z_1...Z_2)$ is $n(Z_1, Z_2) = c\Delta Z_{fr}$, where $\Delta Z_{fr} = Z_2 - Z_1$. Thus the fraction concentrations can be calculated according to the equation

$$n(Z_1, Z_2) = \frac{I\Delta Z_{fr}}{eQ_c\Delta Z_G}, \quad (\text{A6.4})$$

where the reference mobility Z_0 is expected to be in middle of the interval $(Z_1...Z_2)$ and the distribution function is expected to be nearly flat around the reference mobility. In sake of brevity a complex instrument calibration coefficient is introduced

$$w = \left(\frac{Q_c\Delta Z_G}{Q\Delta Z_{fr}} \right), \quad (\text{A6.5})$$

where Q is full air flow rate through the instrument. Now the fraction concentrations are calculated as

$$n(Z_1, Z_2) = \frac{I}{ewQ}. \quad (\text{A6.6})$$

The control program SIGMA1A additionally considers the penetration of the inlet grid and inlet gate P (see Appendix 3) and the working formula is

$$n(Z_1, Z_2) = (CC_n / (wQ)) \times (U_0 / U) \times ADC_E / P, \quad (\text{A6.7})$$

where ADC_E is the output of the electrometric channel and CC_n is the calibration constant, which includes the transformations of measuring units. The calibration constant can be roughly estimated according to the following considerations. Let $n(Z_1, Z_2) = 1 \text{ e/cm}^3$, $wQ = 1 \text{ cm}^{-3}$, $U = U_0$, and $P = 1$. In this case the current of the collector is $1 \text{ e/s} = 1.602 \times 10^{-19} \text{ A}$. The voltage in the electrometer $5 \text{ G}\Omega$ input resistor is $8.01 \times 10^{-10} \text{ V}$ and the $50\times$ amplified signal is $4.005 \times 10^{-8} \text{ V}$. The corresponding count rate of ADC_E is 2.625×10^4 . When all these numbers are entered into the working formula above, we get $CC_n = 3810$. Let us consider $Q = (U/U_0) \times Q_0$ and transformation from the fraction concentration presentation to the distribution function presentation

$$f_{\log Z}(\log Z) = \frac{dn}{d\log Z} = \frac{n(Z_1, Z_2)}{\log(Z_2/Z_1)}, \quad (\text{A6.8})$$

In the "decade to eight" system

$$f_{\log Z} = 8 \times n(Z_1, Z_2). \quad (\text{A6.9})$$

The working formula can be written immediately for the distribution function

$$f_{\log Z} = (CC_f / (wQ)) \times (U_0 / U) \times ADC_E / P, \quad (\text{A6.10})$$

The estimate of the calibration constant according to the considerations above is $CC_f = 8CC_n = 30476$.

A6.3. Air ion concentration: empiric approach

There are no known methods for exact estimation of the air flow coefficient w , which is necessary for the application of the theoretical model. Thus the value of this coefficient is a subject of experimental calibration, where the SIGMA is compared with a precise integral air ion counter. The integral air ion counters can be exactly calibrated using absolute methods. Then the coefficient w should be determined so that the integral concentrations measured using the two instruments are equal.

In case of the experimental calibration, the internal structure of the term $CC_f / (wQ)$ in Equation (A6.10) is of no importance and this term is considered as a single concentration calibration factor CF. The values of the logarithmic distribution function are calculated in the control program as:

$$f_{\log Z} = CF \times (U_0 / U) \times ADC_E / P, \quad (\text{A6.11})$$

Calibration is performed in the conditions of a relatively high concentration of cluster ions of about 26000 cm^{-3} . The air ion concentration was measured simultaneously with the SIGMA (concentration is the sum of fraction concentrations in the cluster ion range) and the integral air ion counter of absolute calibration. The air was prepared with a special air ion generator, providing stable uniform ionization in the air flow that at least twice exceeds the air consumption by the instruments. The measurements were made individually for positive and negative air ions and the concentration factors proved to be slightly different:

$$CF_+ = 6.7,$$

$$CF_- = 6.6.$$

The results are included into the calibration file SIGMA1A.CAL.

A6.4. Calibration of the meteosensors

Atmospheric pressure, air temperature and relative humidity are calculated according to linear transformations of the ADC counts: $result = a \times ADC + b$

Atmospheric pressure is measured by the sensor MPX5100 designed in Motorola and manufactured by Freescale. According to the company information, the coefficient b should be negative. The same sensor was used in the BSMA and calibration according to a mercury barometer results in rather positive values of b . According to the experimental calibration in case of the SIGMA the coefficients are $a = 0.03368$ and $b = 113$.

Temperature and humidity sensors are calibrated in comparison with an Assmann psychrometer. Temperature is measured by the sensor AD590 manufactured by Analog Devices. The experimental calibration $a = 0.01083$ and $b = -290$ differs a little from the factory data. Relative humidity is measured by Honeywell sensor HIH-4031-001. The empiric coefficients $a = 0.00607$ and $b = -35$ differ from the factory data. Additionally, humidity is corrected according to temperature using the factory-recommended transformation $RH = RH_0 / (1.0546 - 0.0215 T:C)$.

The long-term stability of meteosensors is not good and the readings of sensors should be compared with readings of exact instruments every few months. The calibration constants should typically be corrected about once a year.

A6.5. Test measurements

The first field test of the instrument was carried out at a rural site in Abissaare village (cottage Tammemäe, $58^{\circ}05'39'' \text{ N}$ & $26^{\circ}43'09'' \text{ E}$), located about 30 km south of Tartu, Estonia. The installation of the instrument in the attic of the cottage according to the Figure 2.4 is shown in Figure A6.1. Air is sampled through an opening in the gable facing the east. The gable is open to direct sunlight, which results in the enhanced noise in the measurements until the midday. The air inlet and landscape near the cottage are shown in Figure A6.2.



Figure A6.1. The installation of the SIGMA in the attic.

The main purpose of the test measurements was the identification of instrument response to different meteorological conditions, which enabled to elaborate the control program and include proper reactions to extraordinary situations, e.g. distortions emerging in case of contamination of the aspiration condenser and electrometric filters. About 10000 five-minute cycles were recorded during 2009 July-October. The acquired experience is basis for many improvements in the initial version of the software.



Figure A6.2. Cottage Tammemäe and the SIGMA air inlet in the center of the gable.

A6.6. Sensitivity

Some series of measurements were performed especially for estimating the instrumental noise. About 500 cycles of noise were recorded during September 26–28 during strong wind, RH between 80 and 100% and drizzling rain from time to time. The only difference from the standard measurement was permanently closed inlet gate. The results are shown in Figures A6.3–5.

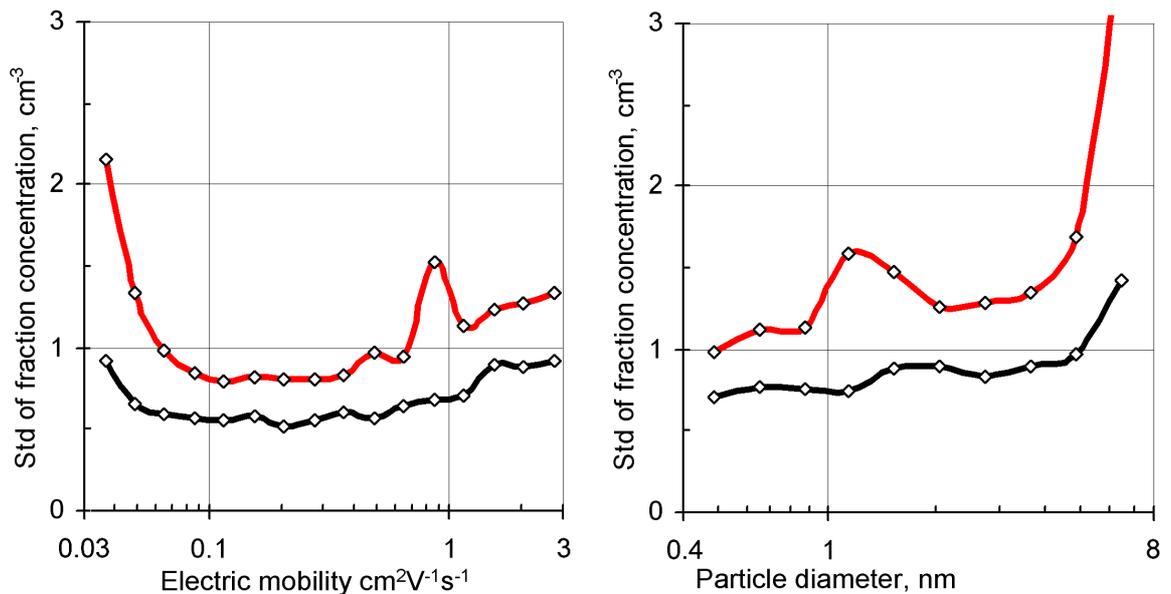


Figure A6.3. Standard deviations of noise signal in 10-minute averages.
Green line for below-median and red line for above-median noise.

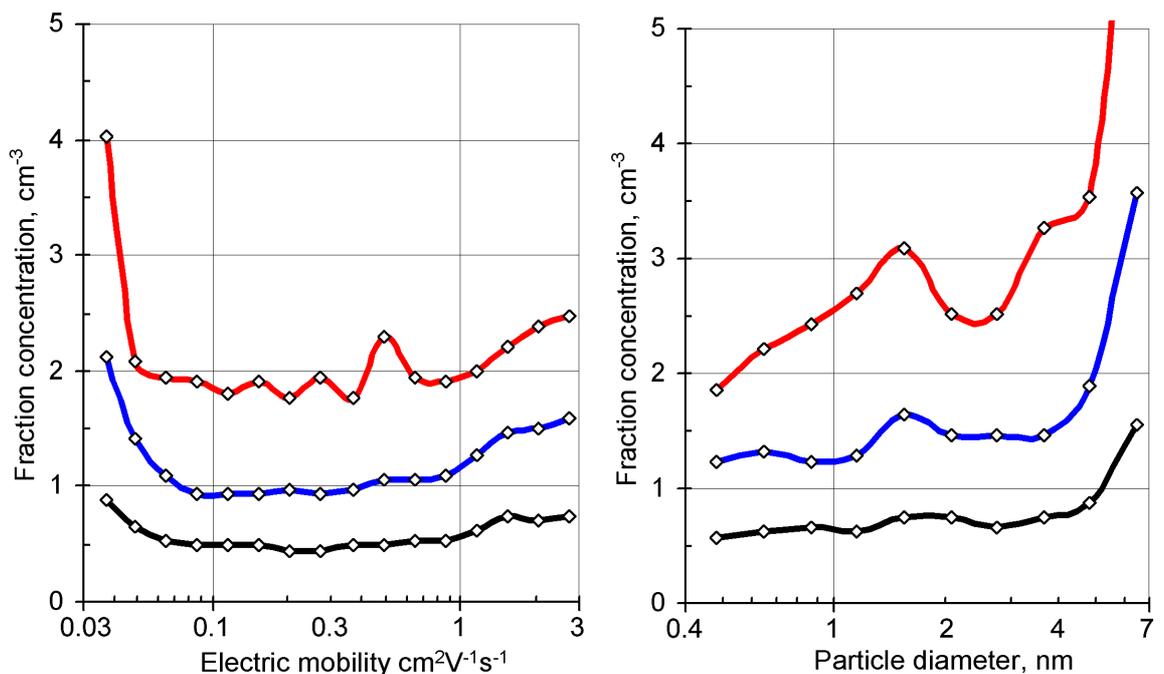


Figure A6.4. 50%, 90% and 99% quantiles of fraction concentration absolute values at zero level of the real concentration.

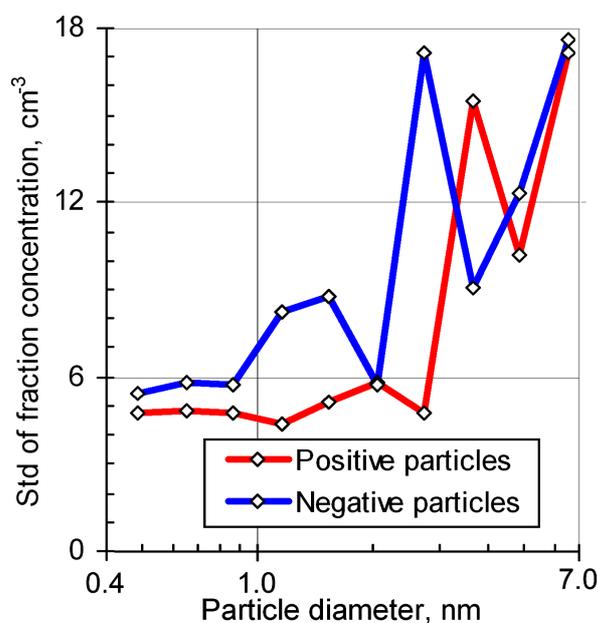


Figure A6.5. Standard deviation of noise signal in size fraction concentrations in case of 20 s time resolution (4470 scans during field measurements Sept 27, 2009).

A6.7. Mobility resolution

Research of nucleating nanoparticles in atmospheric air brings up specific requirements, which radically differ from the requirements for a laboratory mobility analyzer. The signal to noise ratio turns up to be critical due to the extremely low concentration of nanometer ions in atmospheric air, see Figure 1.1. Theoretical models of atmospheric aerosol nucleation include continuous growth of nanometer particles and the mobility distribution is expected to be relatively smooth. Thus the factors of sensitivity will have priority before the factors of resolution. Request of high sensitivity requires increasing the collected ion current. The inlet slit and the collector of the analyzer cannot be narrow and the geometric resolving power is limited. The air is sucked into the instrument immediately from the turbulent atmosphere and the flow rate should be large. Some turbulence inside the analyzer is inevitable in these conditions. As a result, the mobility resolution of instruments for atmospheric measurements cannot be high.

Traditional parameters of the DMA resolution are the relative standard deviation of measured mobilities in case of actually monomobile ions of mobility Z , and the ratio of this mobility to the width of the transfer function at the half height $\Delta Z_{1/2}$. These parameters are denoted below $RSTD = \sigma / Z$ and $RES = Z / \Delta Z_{1/2}$. In case of a Gaussian transfer function $RES = 0.425 / RSTD$.

The resolution depends on several factors:

- width of the inlet slit and width of the electrometric collector,
- representing the distribution with averages over mobility fractions of definite width,
- smoothing due to the response time of the electrometer,
- thermal diffusion,
- turbulent diffusion.

The resulting composite transfer function is approximated by a Gaussian curve and the relative standard deviation of this curve is considered as the measure of the resolution. The geometric transfer function is determined by the widths of the inlet slit and the electrometric collector. According to the numerical model, the geometric transfer function of the SIGMA is very similar to the earlier discussed triangular transfer function of the BSMA (Tammets, 2006). The RSTD of this component of the transfer function proved to be 0.115. Presenting the spectra by fraction averages is necessary for increasing the signal to noise ratio. This

procedure generates a rectangular smoothing function with the relative width of 1.33 and the corresponding component $RSTD = 0.081$. The time constant of the electrometric amplifier is 0.32 s and the smoothed response creates the component $RSTD$ of about 0.027. The effect of thermal diffusion in the SIGMA is low and noteworthy only in case of the smallest ions. The estimate for the ions of the mobility of $1.7 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ is a component $RSTD = 0.026$.

Exact estimating of the mobility resolution of the SIGMA was embarrassed by lack of tools for correct measurement of the turbulence inside the analyzer and lack of the sources of monomobile ions with a large air flow rate. Thus the following estimates are rough. The turbulence in the analyzer was visualized using a thin soft thread vibrating in the air flow in the mobility classification zone. The position of the free tip of the thread was measured from 130 photos and the standard deviation proved to be 1.2 mm. The vibration of the thread is damped by inertia and the standard deviation of air parcels exceeds this value. Thus the value of 1.2 mm is considered only as a lower margin of the turbulent pulsations. The lower margin of the corresponding turbulent dispersion of the mobility was estimated by a component $RSTD = 0.04$ when considering the dimensions of the classification zone. The composite $RSTD$ is a square root of the squares of the five components (0.115, 0.081, 0.027, 0.028, and 0.04), the result is the $RSTD = 0.15$. This value is considered as the lower margin of the estimate of the resolution of the SIGMA.

An estimate of the upper margin of the resolution can be found analyzing the measured spectra of small ions. A spectrum of small ions contains several components and it is always wider than the transfer function. The effect of side components of the ions near the main component is minimized when the spectrum is approximated by a Gaussian line using values of the measured distribution function only near the spectrum peak. The sharpest peak with a mobility of $1.7 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ was found in the spectrum of negative small ions in the artificially ionized laboratory air, where the concentration of ions was about 20000 cm^{-3} . The $RSTD = 0.20$ of this peak is considered as the upper margin of the resolution.

The two margins allow estimating the mobility resolution parameter $RSTD$ to be between 0.15 and 0.2 that corresponds to the values of the RES between 2.1 and 2.8.

A6.8. Numerical improvement of mobility resolution

The mobility distribution of atmospheric intermediate ions is pretty smooth as a rule. The low mobility resolution of the SIGMA may appear unwelcome mainly in the mobility subrange of cluster air ions. A spectral instrument transforms the physical high-resolution distribution into the broadened record of the spectrometer. The mobility resolution can be improved when inverting this transformation. The record of the spectrometer y is a discrete vector that contains up to 35 scalar elements in case of the SIGMA. Thus a determined solution of the algebraic inverse problem can contain no more than 35 unknown scalar elements. In a simplest linear model the instrument noise is neglected, the unbroadened mobility distribution is described by a 35-dimensional discrete vector v , and the instrument transformation is described by the equation

$$y_j = \sum_i G_{ji} v_i \quad (\text{A6.12})$$

where G is the transfer matrix including 35×35 elements. A formal solution of the apparatus equation is

$$v_i = \sum_j G^{-1}_{ij} y_j \quad (\text{A6.13})$$

where G^{-1} is the inverse matrix.

Two facts raise overwhelming difficulties in the application of the simplest model:

- the transfer matrix is not exactly known and minor errors in the matrix elements may cause huge errors in the inverted distribution v ,

- the measured record y contains random errors and minor errors in the elements of y can cause huge errors in the inverted distribution v .

A known technique to outperform the intolerable error amplification is called the regularization. In this case the solution is a partially broadened distribution

$$x_i = \sum_j H_{ij} y_j, \quad (\text{A6.14})$$

where the inverter matrix H is compiled so that the product GH is close to the unit matrix but the transformation A6.14 does not cause too large amplification or measurement errors.

In case of the SIGMA both vectors y and x are composed of fraction concentrations, whose sum should be invariant of the transformation procedure. Thus

$$\sum_j H_{ij} = 1 \quad (\text{A6.15})$$

and the coefficient of amplification the measurement errors in distribution fractions is

$$K_i = \sqrt{\sum_j H_{ij}^2}. \quad (\text{A6.16})$$

The practical problem is to compile H in this way that the vector x can be considered a better representation of the mobility distribution when compared with y , and the elements of K will be low in the range of low-concentration intermediate ions and may be slightly larger in the range of high-concentration cluster ions. An additional test requirement is that the application of inverter to the exactly measured narrow mobility distributions of high-concentration cluster ions should not generate negative elements in the vector x . A practical solution is always a compromise between different requirements and there is no universal solution best in all diverse measuring situations.

During the calibration experiments of the SIGMA, the distributions of artificially generated cluster ions were used as a test subject when compiling the inverter for the decade-to-16 mobility distribution. The possible inverters were discriminated with pseudo-five-diagonal matrices compiled according to the rule described in Section 5.7.3. The choice of generating elements for the standard inverter for SIGMA No.1 is:

$$c_inv_n2 = -0.16, c_inv_n1 = -0.22, c_inv_p1 = -0.10, c_inv_p2 = 0, z_limit = 1.$$

This choice is followed by an error amplification $K_i = 2$ in range of $Z > 1 \text{ cm}^{-2}\text{V}^{-1}\text{s}^{-1}$ and a quick decrease in error amplification when the mobility is decreasing. The near-diagonal elements of a row of the matrix H after normalizing according to (A6.15) are

$$\dots -0.31 \quad -0.42 \quad 1.92 \quad -0.19 \quad 0 \dots$$

and the near-diagonal elements of a correspondingly regularized transfer matrix H^{-1} are:

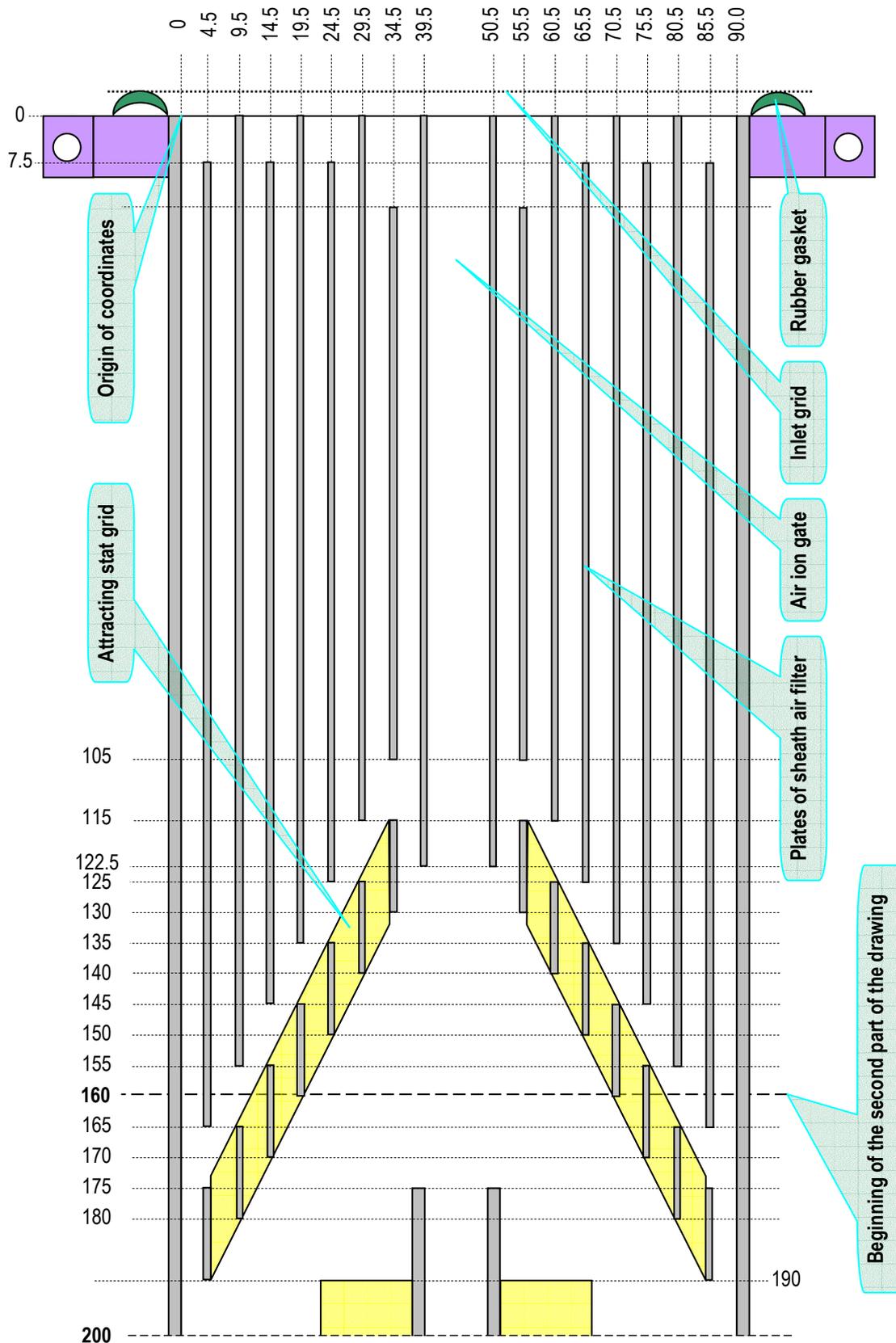
$$\dots 0.05 \quad 0.12 \quad 0.14 \quad 0.55 \quad 0.06 \quad 0.01 \quad 0 \dots$$

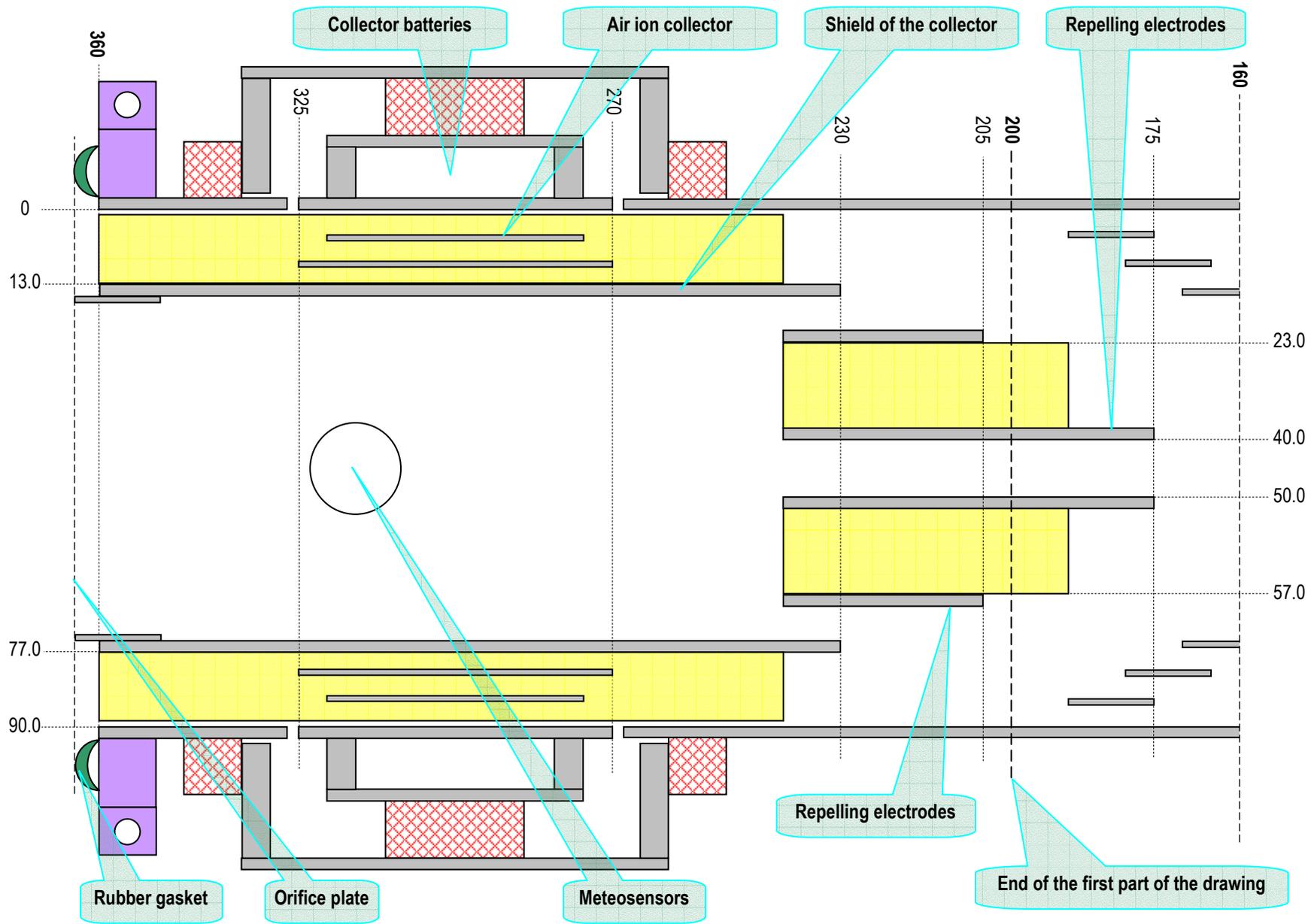
The matrix H and the corresponding regularized transfer matrix appear essentially asymmetric. The explanation is the distortion of air flow in the analyzer: the air is slightly slowed down near the side insulators and in the wakes of sleeves, which are fastening the slats in the center of the attracting grids. The ions passing these regions are recorded with enhanced mobility, which is to be compensated by the stretched left wing of the rows of the inverter matrix.

The standard software of the SIGMA enables to apply or not apply the inversion and to change the values of generating elements of the inverter when the improved calibration data will be available.

Appendix 7. Aspiration condenser

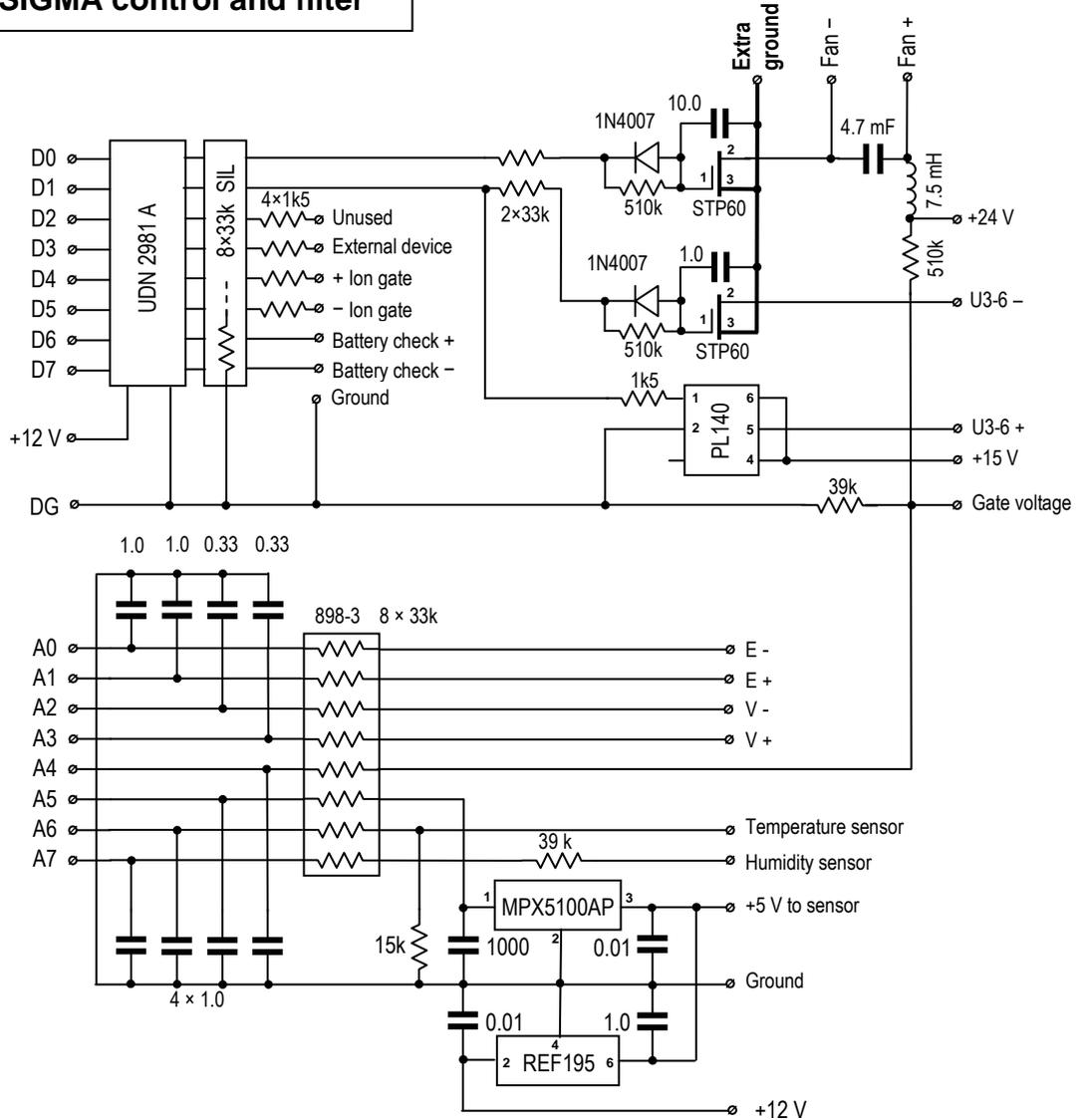
A drawing of the cross-section of the aspiration condenser is presented below in its original size on two pages, at first the outlet part and next, the inlet part. The internal height of the condenser is 240 mm and the height of the ion collector is 190 mm. All plates are made of aluminum sheets of the thickness of 1.0 and 2.0 mm.



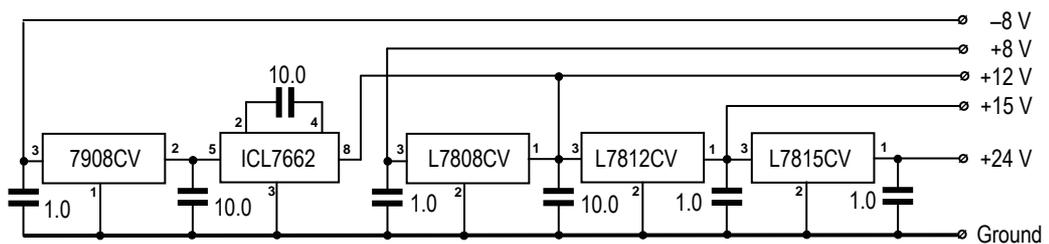


Appendix 8. Electric diagrams and PC boards

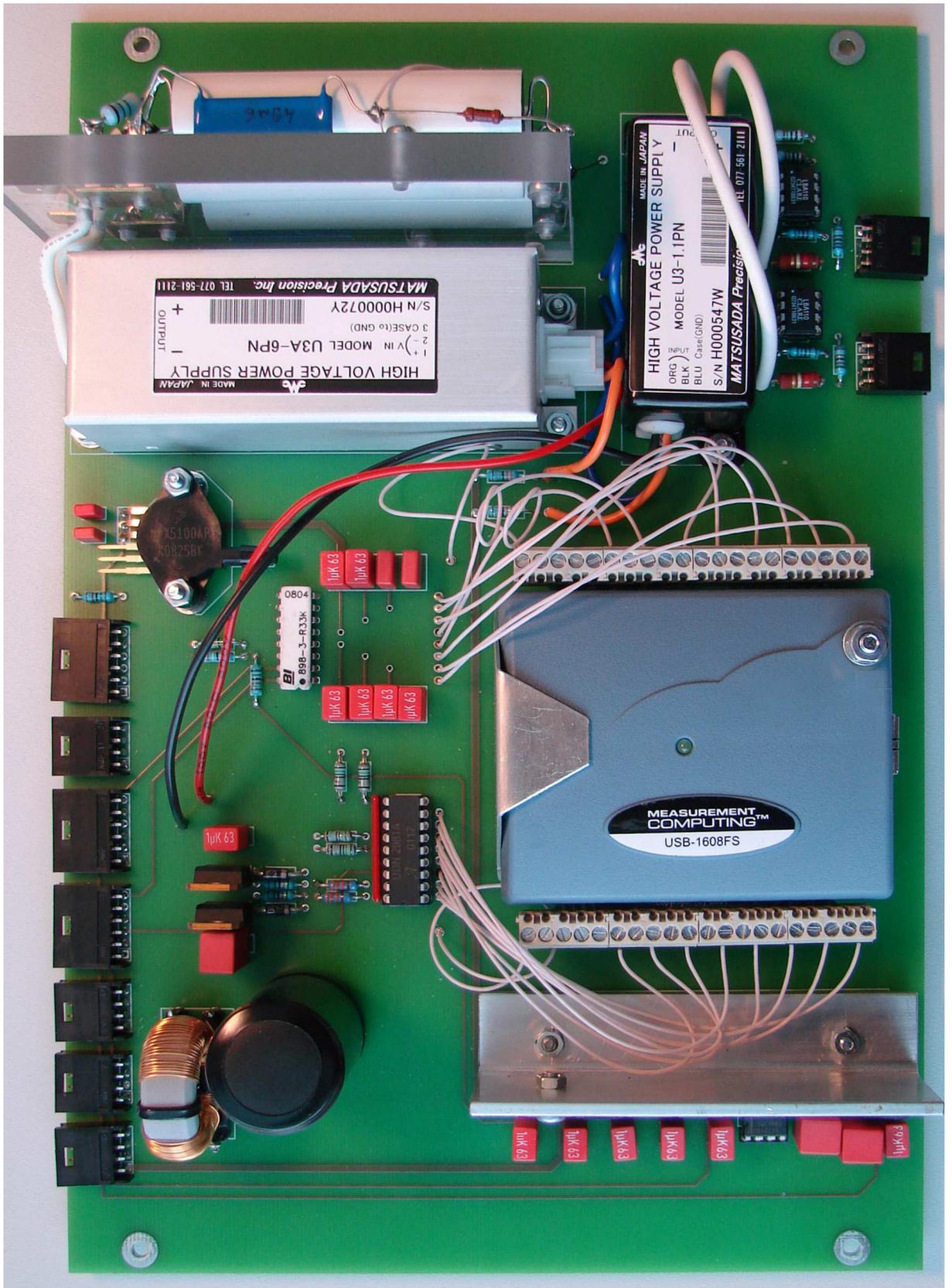
SIGMA control and filter



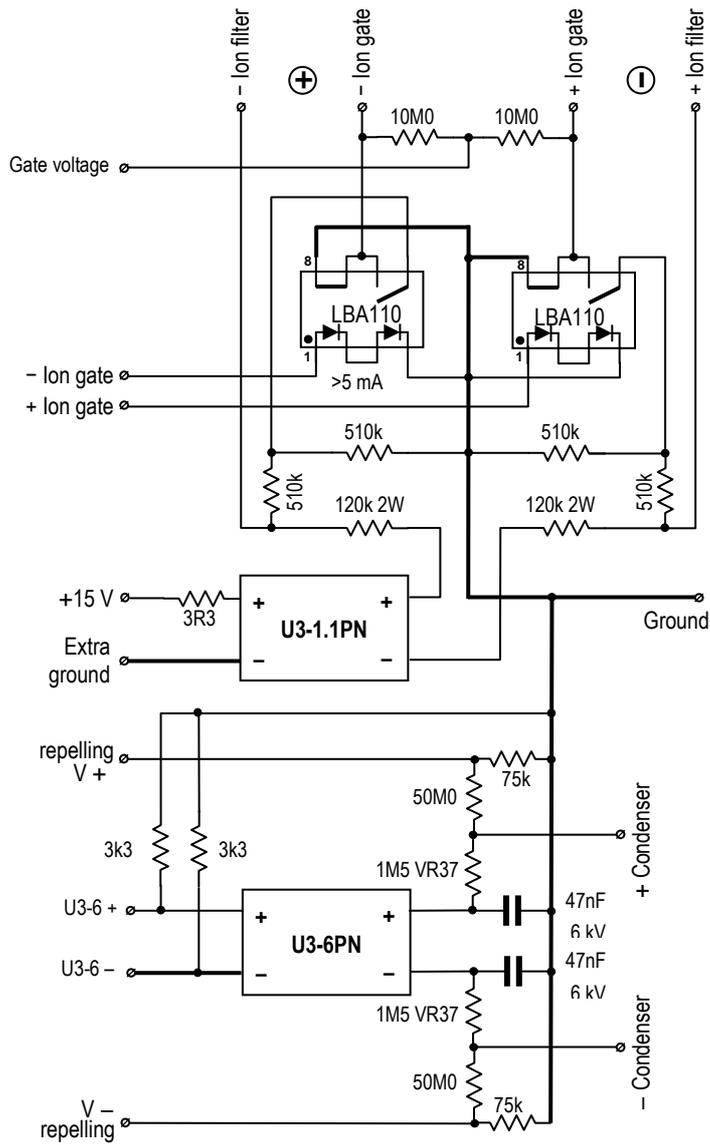
SIGMA voltage regulator



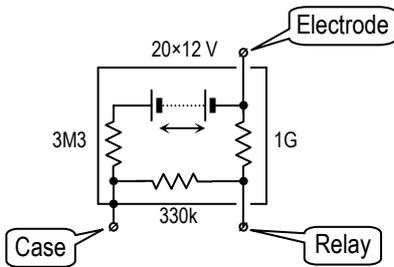
SIGMA control PCB, view from inside the instrument



SIGMA HV

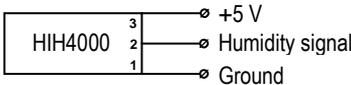
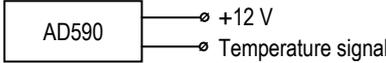


SIGMA filter battery

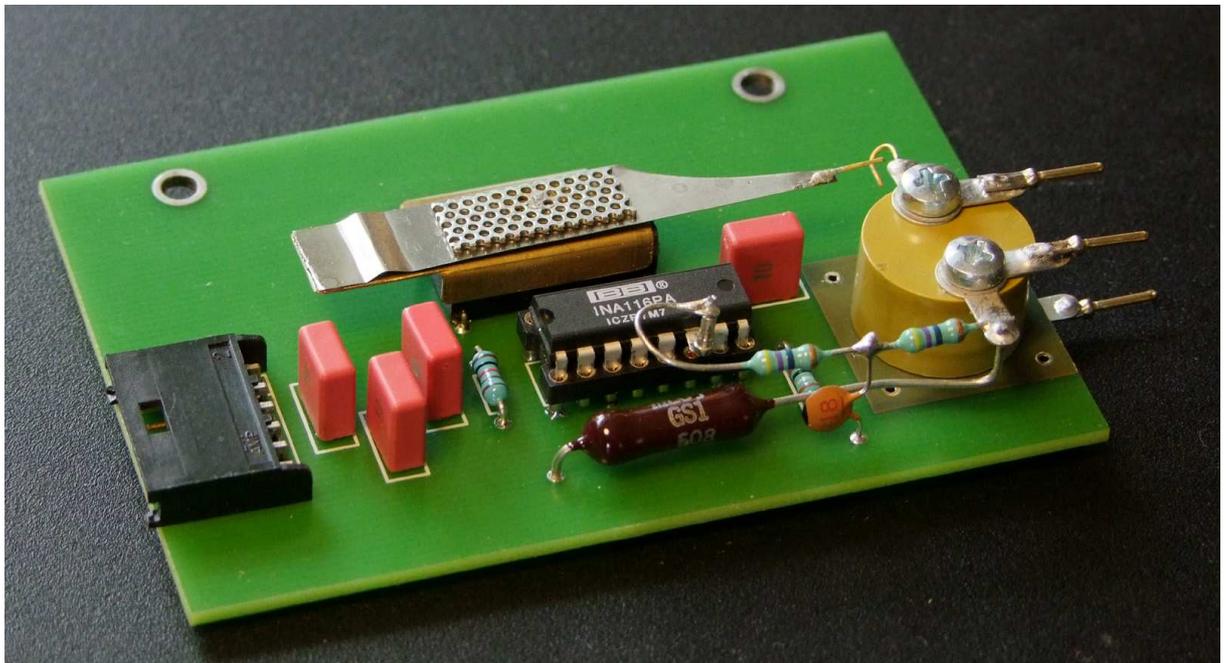
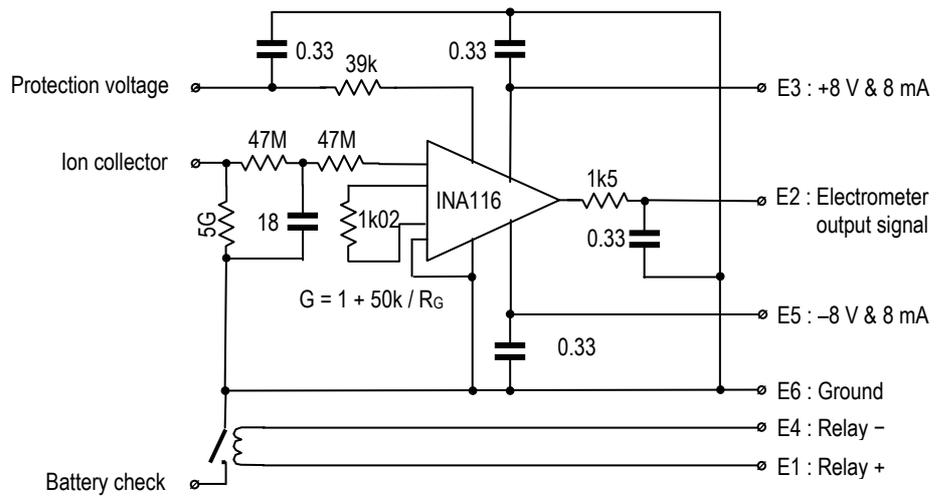


Jagaja 1 GΩ / 327 kΩ
Juhe 37 mm

SIGMA sensors



SIGMA electrometer



Appendix 9. Specifications of SIGMA No. 1

Size: length 505 mm, width 280 mm, height 350 mm.

Mass: 17.2 kg.

Power: AC 47–63 Hz, 90–260 V, 70 W,
or DC 23-25 V (as two car batteries), 60 W.

Suitable AC power units: Mascot 2020 or ZVC65SG24.

Collector batteries: 40 batteries GP27A (expected life time exceeds one year)

Air flow rate with standard orifice plate: $34 \text{ dm}^3 \text{ s}^{-1}$.

Passage time of air from the inlet grid to the ion collector: 0.15 s.

Heat emission of the electronics inside of the analyzer section: 20 W.

Increase in the air temperature during measurement due to the heat emission: $< 0.5 \text{ K}$.

Humidity of analyzed air in case of unpolluted insulators: up to 99%.

Electrometric amplifiers: INA116.

Thermal insulation of the analyzer side panels: foam polystyrene 6 mm.

Mobility range: $0.032\text{--}3.2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ or $0.42\text{--}4.2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$.

Mobility resolution: about 2.5, 16 logarithmically distributed mobility fractions are presented in two decades of mobility.

Diameter range: 0.42–7.5 nm.

Diameter resolution: 10 logarithmically distributed size fractions.

Standard range of fraction concentrations: $0\text{--}40000 \text{ cm}^{-3}$.

Standard deviation of a fraction concentration in the conditions of simultaneous measurement of two polarities, 5-minute averaging, clean insulators, low radon concentration, and low dust concentration about 1 cm^{-3} .

Calibration data from SIGMA1A.CAL 20101026

```

volt_factor=885           for voltage-mobility conversion (Q = 34 l/s)
conc_factor_pos=11.5      for ADC-dn/dlogZ conversion
conc_factor_neg=11.5      for ADC-dn/dlogZ conversion
standardadsorption=0.0505 for Z = 1 cm2V-1cm-1, 0 C, 1013 mb
collectormobility=0.04    cm2V-1cm-1, effective value for corrections
c_supplyvolt=0.002167     supplyvoltage / ADC_counts
c_filtervolt=-0.089       filter voltage / ADC_counts
c_batteryvolt=0.0093      electrometer battery voltage / ADC_counts
c_bias=0.003052           electrometer inlet mV at 1 ADC, 0.1526 / 50
c_pressurea=0.03368       pressure (mb) = c_pressurea * ADCcounts + c_pressureb
c_pressureb=113
c_temperaturea=0.01083    Celsius = c_temperaturea * ADCcounts + c_temperatureb
c_temperatureb=-290       factory value is -273
c_humiditya=0.00607       humidity (%) = c_humiditya * ADCcounts + c_humidityb
c_humidityb=-35           required correction RH = RH (1.0546 - 0.0216 T:C)
electrometerdelay=385     ms, incl. airflow + electrometer - HV signal
chargingtime=1500        ms
timeout=30000            ms
c_inv_n2=-0.16            inverter parameter
c_inv_n1=-0.22            inverter parameter
c_inv_p1=-0.10            inverter parameter
c_inv_p2=-0                inverter parameter
z_limit=1                inverter parameter

```

Included inverter parameters (see Sections 5.7.3 and A6.8) tolerate noise amplification in cluster ion subrange up to 2 times.

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<http://dx.doi.org/doi:10.1016/j.atmosres.2006.02.009>.

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