Milestone M3 (M1.3)

NRT aerosol number size distribution ST for RI-URBANS

RI-URBANS

Research Infrastructures Services Reinforcing Air Quality Monitoring Capacities in European Urban & Industrial AreaS (GA n. 101036245)

By

TROPOS

TROPOS
Leibniz-Institut für Troposphärenforschung

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RI-URBANS (www.RIURBANS.eu) is supported by the European Commission under the Horizon 2020 – Research and Innovation Framework Programme, H2020-GD-2020, Grant Agreement number: 101036245

Milestone M3 (M1.3): NRT aerosol number size distribution ST for RI-URBANS

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Work package (WP)	WP1 Novel AQ metrics and advanced source apportionment STs for PM, and nanoparticles
Milestone	M3 (M1.3)
Lead beneficiary	TROPOS
Means of verification	Near real time aerosol number size distribution service tool for RI-URBANS adaptation available. This has to be used by pilots.
Estimated delivery deadline	M5 (28/02/2022)
Actual delivery deadline	10/02/2022
Version	Final
Reviewed by	WP1 leaders
Accepted by	RI-URBANS Project Coordination Team
Comments	Report summarising the process for NRT particle size distribution Service Tool for RI-URBANS

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1. About this document

This document summarises the process for NRT aerosol size distribution Service Tool for RI-URBANS (Research Infrastructures Services Reinforcing Air Quality Monitoring Capacities in European Urban & Industrial AreaS, Horizon-2020 GD project #101036245). This is a public document, available at the RI-URBANS website, https://riurbans.eu/work-package-1/#milestones-wp1, and distributed to all RI-URBANS partners for their use as well as submitted to European Commission as a RI-URBANS milestone 3 (M3).

2. Guidelines

2.1 Setup and operation of MPSS

Aerosol particle number size distribution measurements in the submicrometer size range are done by a Mobility Particle Size Spectrometers. The CEN/TS 17437 is the standard for ambient aerosol measurements, being the basis for ACTRIS and RI-URBANS compatible measurements. The guidelines are available (https://www.actris-ecac.eu/measurement-guidelines.html). The guidance documents are attached as an annex at the end of this milestone (Annex 1).

The data should be provided according to the level-0 ACTRIS data protocol that the data processing can be done at the ACTRIS data Center.

2.2 NRT Software

The software for generating an ACTRIS compliant data stream is readily available. Adaptations and configuration will take place during the implementation phase. Evaluation of the station, including quality assurance, is carried out by means of a test phase, after which the data stream is redirected to the ACTRIS data centre.

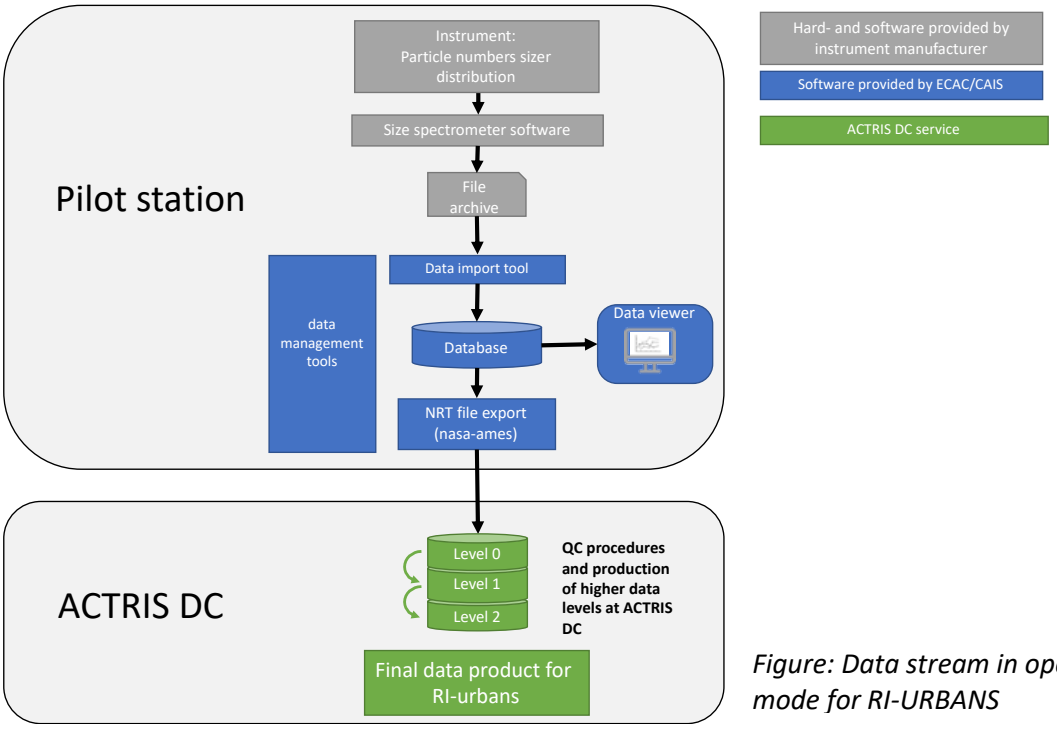

Figure: Data stream in operational mode for RI-URBANS

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3. Process for implementing NRT aerosol size distribution Service Tool for RI-URBANS

The implementation of the service tool for measuring the particle number size distribution and deployment near real time requires a number of steps. The procedure aims to generate ACTRIS compatible data for provision in the data centre.

1. Aerosol particle number size distribution measurement in an urban environment: The pilot stations Barcelona, Helsinki and Birmingham are operational.
2. Contact was made by the ACTRIS Central Facility CAIS-ECAC to ensure that the measurement guidelines are followed.
3. Collection of information about instrument and measurement setup: in progress
4. Timeline for software implementation: Software is ready and specific adaptions will be implemented in M6 to start producing test data sets
5. Suggestion for modification based and test data sets analysed by CAIS
6. Preparation at ACTRIS DC to receive NRT data
7. Opening the data stream between the measurement site and ACTRIS DC: M7

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Annex 1

1. ACTRIS Recommendation for mobility particle size spectrometer measurements: Part I recommended instrument set-up
2. ACTRIS Recommendation for measurements with mobility particle size spectrometers - Part II recommended particle loss correction
3. ACTRIS Recommendation for measurements with mobility particle size spectrometers - Part III Standard Operation Procedure
4. ACTRIS Recommendation for measurements with mobility particle size spectrometers - Part IV Constants and Relevant Equations
5. ACTRIS In Situ Aerosol: Guidelines for Manual QC of MPSS Data [MF1]

RI-URBANS (www.RIURBANS.eu) is supported by the European Commission under the Horizon 2020 – Research and Innovation Framework Programme, H2020-GD-2020, Grant Agreement number: 101036245

ACTRIS Recommendation for mobility particle size spectrometer measurements: Part I recommended instrument set-up

Alfred Wiedensohler and Wolfram Birmili

Leibniz Institute for Tropospheric Research

ACTRIS Technical Standard

This recommendation is based on the article of Wiedensohler et al. (2012).

Within the EUSAAR and ACTRIS projects, we have developed technical standards for mobility particle size spectrometers. Parts of these standards have resulted from the desire to harmonize aspects of hardware, and enhance the accuracy and definition of the measurement. Others were conceived to enhance the data formatting and evaluation procedure of the measurements. The recommended standards have been clearly motivated by the needs of long-term field experiments, nurtured by a multi-annual practice of field observations and laboratory intercomparisons of mobility particle size spectrometers. The general spirit of these recommendations is to improve the accuracy and world-wide comparability of such measurements. We encourage operators of atmospheric measurements of particle number size distributions to adhere to these standards as far as possible.

Technical Features of the Mobility Particle Size Spectrometers

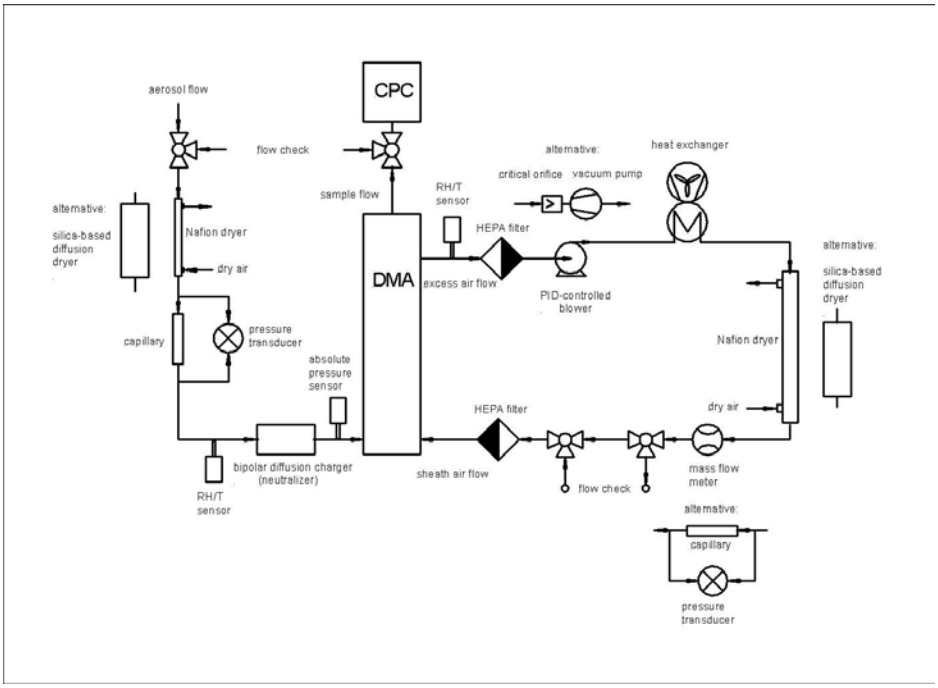

Figure 1: Recommended set-up of a mobility particle size spectrometer for long-term measurements of the ambient aerosol

The schematic of our recommended mobility particle size spectrometer is shown in Figure 1. Here, the sheath air flow is circulated in a closed loop, a principle implemented in most commercial and custom-made mobility particle size spectrometers. The recommended set-up includes dryers to reduce RH in the aerosol sample and sheath air flows. The dryer in the sheath air flow helps to avoid measurements with moist air somewhere in the DMA and to achieve a stable relative humidity in the system. Furthermore, it reduces the time lag to dry all flows and HEPA (High Efficiency Particle) filters. The sheath air loop contains a heat exchanger and HEPA filters. Sensors continuously record the aerosol and sheath air flow rates, relative humidity and temperature in both flows, and absolute pressure in the aerosol flow entering the DMA.

For scanning mobility particle size spectrometers, we recommend using a minimum scanning time (up or down scan) of 2 min to avoid smearing effects in the particle counters with a relatively slow response time. These smearing effects can cause e.g. significant false measurements at the slope towards larger particles in the accumulation mode range.

Relative humidity (RH) Control and Measurement

Due to the hygroscopic growth of atmospheric aerosol particles at RH well below supersaturation, it is essential to control or limit RH in mobility particle size spectrometers. The philosophy is to obtain comparable data sets and, therefore, to measure the “dry” particle number size distribution. When working in a warm and moist atmospheric environment, the dew point temperature can reach the standard temperature of a measurement laboratory (20-25°C). This requires that the aerosol sample flow has to be dried, either directly in the main sampling line or at the instrument. A dry aerosol sample is needed to ensure the correct bipolar charge equilibrium and, thus, sizing downstream of the bipolar diffusion charger in the DMA. A dry sheath air is needed to ensure particle sizing inside the DMA with a minimum fluctuation in RH. The recommendation is to limit RH inside an instrument to below 40%. In this regime, changes in particle diameter as a result of RH are expected to be below 5%.

To limit RH in the aerosol sample flow (see also the recommendation for the drying), we concretely recommend using a membrane dryer (made from materials such as Nafion™), or a silica-based aerosol diffusion dryer. Operation of a membrane dryer will require a continuous supply of dry air in the laboratory, while a silica-based dryer will require regular regeneration. Utmost care should be taken to select or design dryers that feature minimum particle losses, such as due to Brownian diffusion. Ideally, particle losses across the dryer are characterized and accounted for in the data processing as an equivalent pipe length (see below).

In complete analogy, the sheath air flow rate should be dried below 40% RH as well. Both membrane and diffusion dryers can be used. RH in the sheath air flow should be monitored continuously by a calibrated humidity sensor as well. The sheath air RH sensor should be installed as close as possible to the DMA at the excess air outlet. The objective is to measure RH at a temperature and pressure that best represent the conditions inside the DMA. As a guideline, the temperature of the sheath air RH sensor should not differ more than 1 K from the temperature in the DMA.

RH in both the aerosol and sheath air flows should be monitored continuously by calibrated humidity sensors with a maximum uncertainty of maximum 5% RH across the range of 10-90%. These data should be recorded and stored with at least the same time resolution as the electrical particle mobility distributions. When dual mobility particle size spectrometers (systems with two parallel DMAs) such as a TDMPS (Twin Differential Mobility Particle Sizer) are used to cover a wider particle size range (e.g. below 10 nm), the RH parameters should be separately reported for each DMA.

Sheath Air Flow Circuit Specifications

In the case of a closed-loop sheath air flow, a heat exchanger is needed to remove the excess heat generated by the pump or blower. An ideal instrument employs two HEPA filters to provide particle-free sheath air at the exit from and entrance to the DMA. The pressure drop across the HEPA filters should be minimal to ensure a correct measurement in the closed loop of the sheath air flow. For a critical orifice/pump set-up, the absolute pressure downstream of the critical orifice should be monitored to ensure critical flow conditions (pressure downstream less than half of the upstream pressure).

Aerosol and Sheath Air Flow Measurement

One of the important but sometimes apparently underestimated issues in particle electrical mobility measurements is the correct determination of the instrumental air flows. Errors in the experimental aerosol and sheath flow rates will propagate immediately into the derived particle number concentrations and/or particle sizes. Our general advice is to combine continuous and automated flow measurements inside the instrument with the manual precision measurements that are typically part of regular maintenance. To ensure continuous observations of the aerosol and sheath air flow, our recommended set-up includes the use of calibrated flow meters in the respective positions.

For the aerosol flow, we recommend using a calibrated differential pressure transducer measuring the pressure drop across a laminar flow element (capillary). While such a capillary can be manufactured from widely available plumbing elements, care should be taken to warrant an undisturbed laminar flow across the device. It is particularly not recommended to use mass flow meters for the aerosol flow, because of particle losses. The measured flow values should be recorded and stored with at least the same time resolution as the measured electrical particle mobility distributions. As a guideline for quality control, the continuously recorded aerosol flow should not deviate more than 5% from the set-point. Besides the continuous measurement, the aerosol flow needs to be checked manually using a precision volumetric flow meter (e.g. an electrical bubble flow meter). This manual measurement should take place as often as possible, but at least at each service occasion (every month at least). The quality of the continuous flow measurement will be improved if the differential pressure transducer is recalibrated regularly.

For the sheath air flow measurement, two options are possible: Either a differential pressure flow meter as described above, or a mass flow meter – because particle losses do not matter inside the sheath air flow. To capture the flow rate under conditions as close to the conditions (pressure, temperature) inside the DMA, the flow meter should be installed near the sheath air inlet (but upstream of the HEPA filter). For differential pressure flow meters, the sensor voltage is typically

calibrated against a reference volumetric flow. Any mass flow meter should also be calibrated for volumetric flow using a reference volumetric flow meter, thereby accounting for air pressure and temperature in the laboratory. As a guideline, the sheath air flow should be kept as constant as possible, with a maximum deviation of its floating average of 2% around the set-point value. The required temporal stability can be accomplished either by a critical orifice/pump set-up or by an air blower that is controlled by software or hardware.

Temperature and Pressure

To ensure the highest quality and traceability of mobility particle size spectrometer measurements, temperature and absolute air pressure should be monitored in the instrument. The objective is to determine the conditions given at any time inside the DMA, because these are needed to ascertain the correct sizing of the particles and to adjust the final particle number size distributions to standard conditions (273.15 K, 1013.25 hPa). The preferred option is to monitor temperature and absolute air pressure near the aerosol inlet of the DMA, however, without disturbing the laminar flow profile. Since RH sensors are usually capable of recording temperature as well, it is useful to store the temperatures values from those positions as well. As mentioned before, all parameters should be stored with at least the time resolution of the measured electrical mobility distribution. In the case of dual mobility particle size spectrometer, it is obligatory to report the recorded parameters separately in conjunction with each DMA.

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ACTRIS Recommendation for measurements with mobility particle size spectrometers - Part II recommended particle loss correction

Alfred Wiedensohler and Wolfram Birmili

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Correction of Particle Losses

This recommendation is based on the article of Wiedensohler et al. (2012).

Particle losses may practically occur in any part of a mobility particle size spectrometer. An important mechanism is particle diffusion to walls e.g. inside of pipes, the DMA, aerosol dryer and bipolar charger, especially for particles smaller than 100 nm in size. If particle losses in a particularly device are known as a function of particle size, they can be corrected during the data post-processing. A useful parameter to describe particle losses in any component of the mobility particle size spectrometer is the method of “equivalent pipe length”. Particle losses by diffusion of different components of the mobility particle size spectrometers are described by a straight pipe, which has the same particle penetration (equivalent pipe length, see Table 1). The losses can thus be easily computed for any particle size and flow rate from such an equivalent pipe length. Equivalent pipe lengths of different devices and plumbing elements aligned in sequence can be simply added if they are traversed by the same rate of aerosol flow. To ensure traceability of the data, any such corrections need to be documented when submitting data to a data base.

Plumbing

Particle losses by diffusion in a straight pipe can be described by analytical formulas derived for the laminar flow regime. For a developed laminar flow, these losses depend only on the pipe length, the flow rate through the pipe, and the particle size. When designing a mobility particle size spectrometer, it is advisable to use connecting pipes as short as possible, and as straight as possible. Enhanced diffusional particle losses may occur in sampling pipes containing bends or elbows. These enhanced particle losses increase with a decreasing radius of the bend or elbow. We estimated the equivalent pipe length of a 90° bend based on the investigation of Wang et al. (2002). Using curves with smooth radii instead of elbow joints will also reduce the opportunity for particle losses. It is very essential that the plumbing consists of electrical conducting material, preferably stainless steel. Experience has shown that non-conductive tubing (e.g. plastics) may remove a considerable fraction of any charged particles by electrostatic forces.

Bipolar Diffusion Charger

Particle losses also occur inside bipolar diffusion chargers. The loss correction can be directly applied based on the experimentally determined penetration efficiency. Alternatively, any experimental penetration efficiency under a specific flow can be converted to an equivalent pipe length using the

diffusional deposition formula for laminar flow. Particle losses for sub-10 nm particles across 85Kr bipolar diffusion chargers were measured for a TROPOS custom-made bipolar charger.

Differential Mobility Analyzer

Different DMA types exhibit different particle losses due to Brownian diffusion. The probability of a particle penetrating through a DMA depends on the losses in the DMA inlet and outlet region as well as on the transfer function in the DMA classification region. Short column lengths and high aerosol and sheath air flows are general design features that minimize particle losses. Particle losses can be either simulated by diffusional deposition models, or estimated experimentally. As with the bipolar diffusion charger, the diffusional losses across different DMAs have been simulated by an equivalent pipe length as given in Table 1.

Condensation Particle Counter

Each CPC may have a rather individual particle counting efficiency, which can be determined experimentally. The size-dependent counting efficiency of an individual CPC depends on many specific factors, such as CPC geometry, or the actual supersaturation profile inside the condenser. If experimental data on the counting efficiency of a particular CPC are not available, the manufacturer's calibration curve can be applied with caution. Our recommendation is, however, to calibrate CPCs individually against a reference instrument. Experience suggests that the performance of CPC degrades typically after one year of continuous ambient measurements due to laser power deterioration or contamination of the optics. When calibrating a CPC, particle losses inside the CPCs are implicitly included in the measured counting efficiency.

Device	**Equivalent pipe length**	
Hauke-type medium-DMA (28 cm effective length)	4.6m	Karlsson and Martinsson (2003)
Hauke-type short-DMA (11 cm effective length)	4.6m	IFT internal calibration
TSI long-DMA (444mm effective length)	7.1m	Karlsson and Martinsson(2003)
TSI nano-DMA (49.9mm effective length)	3.64m	Jiang et al. (2011)
Permapure Nafion dryer SS2400	2.5m	Dick et al. (1995)
Permapure Nafion dryer SS1200	1.25m	Dick et al. (1995)
Diffusion dryer (e.g. TOPAS)	5m	estimated from Tuch et al. (2009)
90 bend (less than 5 cm radius)	0.15m	estimated from Wang et al. (2002)
Bipolar diffusion charger (IFT custom-made)	1m	Covert et al. (1997)

Table 1: Recommended equivalent lengths taken from Wiedensohler et al. 2012

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ACTRIS Recommendation for measurements with mobility particle size spectrometers - Part III Standard Operation Procedure

Alfred Wiedensohler and Wolfram Birmili

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Standard Operation Procedure and System Checks

This recommendation is based on the article of Wiedensohler et al. (2012).

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

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

- In case of a closed-loop instrument, the pump/blower must be sealed and leak testing should be part of the regular maintenance schedule for the instrument.

- Humidity and temperature sensors for the aerosol and sheath air flow have to be checked prior to their deployment and afterwards at least once per year.

- The response function of the high voltage (HV) supply should be calibrated. This should include the analogue output module if high voltage supply is controlled through an analogue voltage. The calibration function of the high voltage should be implemented into the scanning software or the data analysis. Correct sizing of small particles is highly sensitive to accurate knowledge of the applied HV. Particular care is, hence, required in the low voltage range. A HV-probe with ultralow impedance should be used here. The HV power supply has to be checked monthly.

- Furthermore, CPCs have to be calibrated regularly at least once per year to detect malfunctions such as degradation of the laser diode, temperature instabilities, or internal pollution. CPCs should be only used after determining the flow rate and after a calibration of the detection efficiency curve (see also Wiedensohler et al., 1997) and the plateau detection efficiency. Often, the CPC flow rate is controlled by a critical orifice. It should not differ more than few percent from the nominal value. The deviation of the flow rate from the nominal value should then be taken into account in the calculation of the particle number size distribution. The volume flow rate should be checked on a monthly basis.

- The sizing accuracy of mobility particle size spectrometers have to be verified using 200 nm PSL spheres frequently. The use of 200 nm PSL particles is a compromise obtaining a

sufficient particle number concentration and a minimum of residual material on the particles. The measured peak diameter should be within the nominal uncertainties of the PSL spheres (+/-2.5%) and the sheath air flow rate (+/-1%). Due to a pressure drop over the external volumetric flow meter, it is often difficult to precisely measure the actual flow rate of the sheath air. In this case, the sheath air flow rate might be slightly adjusted by few percent to match the nominal PSL sphere size.

- For scanning mobility particle size spectrometers, an incorrect pluming delay time can only be determined by the PSL sphere check. The pluming time is correct if up- and down-scans show the same result. The scan time has to be long enough because of the slow CPC response and to avoid smearing effects. We recommend an up- or down-scan time of minimum 2 min.

- Mobility particle size spectrometers should also be regularly compared to a reference instrument for a period of few days once per year (if a reference system is available). This intercomparison can be done either within an intercomparison workshop or at the sampling site. If a reference mobility particle size spectrometer is not available, also the total particle number concentration measured by a CPC can be compared to the number integral of the size distribution. The integral of the particle number size distribution should be compared to the directly measured total particle number concentration if no nucleation mode particles are present. Ideally, the difference in particle number concentrations should be smaller than 10% after correction for internal diffusional losses.

- The Zero-check of the system should be also done every month. An absolute particle filter should be connected to the system inlet and scanned for several size distributions. Ideally, the background should be close to zero.

- The DMA and the laminar flow element to determine the aerosol flow rate have to be cleaned once per year. CPCs have to be serviced by an experienced person to clean the saturator and the optics.

- The bipolar diffusion charger should not be opened. The instructions of the manufacturer have to be followed.

- The mobility particle size spectrometers should be operated in an environment of 15-30°C to avoid a malfunction of the particle counter.

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ACTRIS Recommendation for measurements with mobility particle size spectrometers - Part IV Constants and Relevant Equations

Alfred Wiedensohler and Wolfram Birmili

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Constants and equations

This recommendation is based on the article of Wiedensohler et al. (2012).

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

Dynamic gas viscosity at 296.15 K and 1013.25 hPa:

$$\eta_0 = 1.83245 \cdot 10^{-5} \frac{\mathrm{kg}}{\mathrm{m\ s}}$$

$$\eta = \eta_0 \left(\frac{T}{T_0} \right)^{3/2} \left(\frac{T_0 + 110.4\mathrm{K}}{T + 110.4\mathrm{K}} \right)$$

Mean free path at 296.15 K and 1013.25 hPa:

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

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

Cunningham correction:

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

Bipolar charge distribution

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

ranges from 1 to 1000 nm or 20 to 1000 nm particle diameter for n equal to -1, 0, +1 or -2, +2, respectively. The according approximation coefficients are given in Table 1.

Approximation formula:

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

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

Table 1: Approximation coefficients after Wiedensohler 1988

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

Gunn (1956) equation:

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

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ACTRIS In Situ Aerosol:
Guidelines for Manual QC of MPSS Data[MF1]

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Due to the variety of MPSS systems and the complexity of the inversion algorithm for MPSS data, the main manual QC is to be performed on the level 1 data (inverted data). Raw data for level 0 (the raw data output from the instrument) are only augmented with essential information (e.g. from entries in the instrument/station log) and metadata recorded by the instrument and brought to a standardized data format. Level 0 shall be flagged for operation parameter deviations larger than described in Wiedensohler et al. 2018.[MF2]

When performing manual QC for a given time period of data, e.g. a year for an annual submission of data, the parameters contained in the level 0 data listed below are to be plotted as time series, and visually inspected. Data sequences exhibiting issues are to be flagged with an appropriate flag contained in this list below.

For manual QC of level 1 and level 2 data time series plots of individual parameters are not sufficient to identify all possible issues in a data set. Contour plots of the PNSD are helpful to identify major issues in the data (like arcing, possible local pollution or lack of CPC working fluid), which may not be visible in time series plots (see appendix A for examples). Frequency distributions may help to identify small negative values and uneven distributions of the measured values along with the time series plots that are not visible in countourplots (appendix B). Data with issues identified by these tools shall be flagged according to the list below.

Any non-invalidating flag (559, 640, 110) is to be propagated to the average if it occurs during the averaging period.

Group 0: Valid data

Flag	Validity	Description
000	V	Valid measurement

Group 1: Exception flags for accepted, irregular data

Flag	Validity	Description
110	V	Episode data checked and accepted by data originator. Valid measurement

Group 3: Flags for aggregated datasets (used for level 1.5 & 2 only)

Flag	Validity	Description
390	V	Data completeness less than 50%
392	V	Data completeness less than 75%
394	V	Data completeness less than 90%

Group 5: Chemical problem

Flag	Validity	Description
559	V	Unspecified contamination or local influence, but considered valid

Group 6: Mechanical or instrumental problem

Flag	Validity	Description
640	V	Instrument internal relative humidity above 40%
683	I	Invalid due to calibration. Used for Level 0.
686	I	Invalid due to zero check. Used for Level 0

Group 9: Missing flags

Flag	Validity	Description
999	M	Missing measurement, unspecified reason

Regardless in which data level the issue is found, the flags are added to the initial level 0 data version, thereby producing level 0a (manually QCed level 0) as output of the QC process. Some flags are used for level 0 only. The corresponding data lines are marked as missing in level 1, and are excluded from calculating hourly averages in levels 1.5 and 2.

The flags for aggregated datasets in group 3 apply only to levels 1.5 and 2. They indicate which fraction of the averaging period is covered by active sample time of the instrument.

The following parameters are to be inspected for the issues:

Level 0:

1. **Periods of zero and span checks**
   If not done automatically by the data acquisition software, periods of zero checks and calibrationscks are to be flagged with flags 686 and 683, respectively.
2. **Sample pressure, sample inlet temperature**
   Sample pressure varies with ambient pressure. Other types of variations should not occur, e.g. variations with fluctuating sample flow. Sample temperature at inlet and outlet normally varies only with lab temperature, and during zero and sizing checks. Other variations and spikes need to be inspected, the reason determined, and flagged according to the issue if needed.
3. **Sample relative humidity at inlet and outlet**
   Sample relative humidity varies with ambient relative humidity and the temperature difference between ambient and lab. The sample should be dried so that the sample has rH < 40% already at the instrument inlet. If rH is higher, apply flag 640. Spikes of rH can occur during zero and sizing checks. These periods need to be flagged 999. Other variations and spikes need to be inspected, the reason determined, and flagged according to issue if needed.
4. **Sample flow and sheath air flow**
   Sample flow and sheath air flow should be constant, with small variations caused by wind gusts. Sample flow under normal operation should typically be at least 1 l/min, with variations smaller than 5%. Sheath air flow should be stable as well. Variations and

spikes exceeding 5% need to be inspected, the reason determined, and flagged according to issue if needed.

5. **CPC operation parameters**
   Recorded CPC operation parameters need to be within manufacturer specifications. Time periods with deviations from these specifications must be inspected for data validity as such deviations might only be due to a faulty sensors in the CPC. A good indicator for the validity of data during such periods is the comparison of the integral particle number concentration determined by the MPSS with the number concentration reported by an extra total CPC.

Level 1:

1. Visually inspect contour plots of the PNSD on a daily or maximal biweekly basis and identify possible local pollution, arcing DMA or periods like zero checks or lack of CPC working fluid. See appendix A for examples. Flag data accordingly.
2. It is helpful to create histograms of the frequency distributions of all measured parameters. Typically these frequency plots are log normally distributed. Histograms can therefore easily help to identify unusual measurements, which cannot be identified in contour plots. Flag data where necessary.
3. Compare MPSS derived integral particle number concentration with total CPC (see level 0)

Level 2:

1. **Particle number size distribution**
   Create monthly contourplots of the PNSD to verify data integrity of the hourly average data check time series for flags.

Reference: A. Wiedensohler, A. Wiesner, K. Weinhold, W. Birmili, M. Hermann, M. Merkel,T. Müller, S. Pfeifer, A. Schmidt, T. Tuch, F. Velarde, P. Quincey, S. Seeger & A. Nowak (2018) Mobility particle size spectrometers: Calibration procedures and measurement uncertainties, Aerosol Science and Technology, 52:2, 146-164, DOI: 10.1080/02786826.2017.1387229

Appendix A: Problems identified by contour plots:

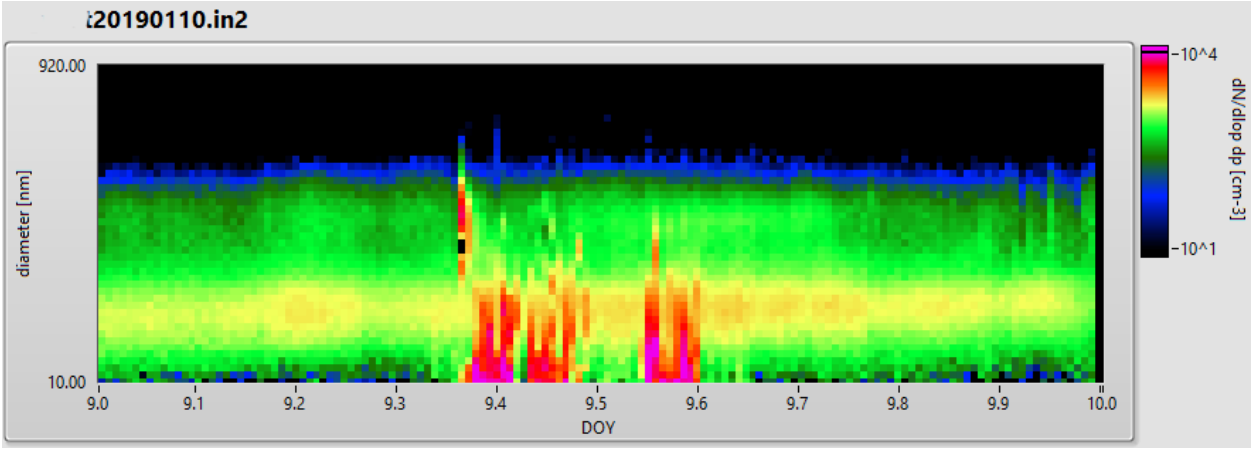

Fig 1: Possible local pollution

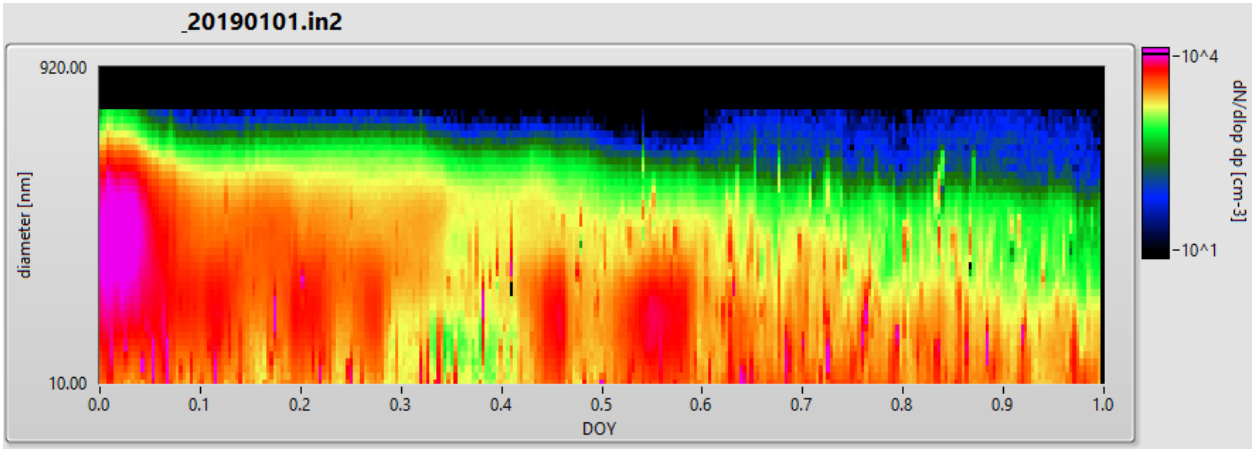

Fig 2: Local pollution (new years eve fireworks)

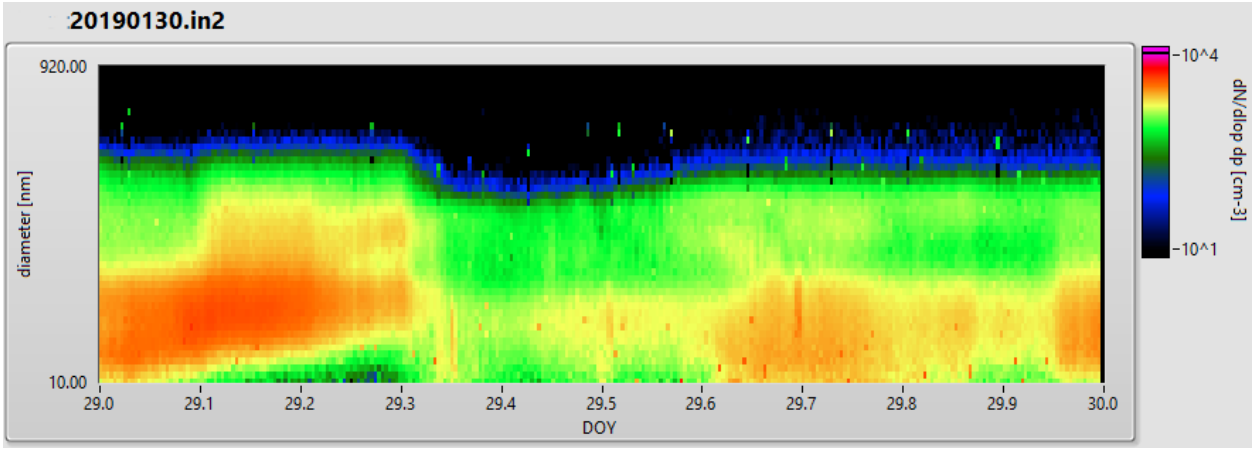

Fig.3: DMA is arcing

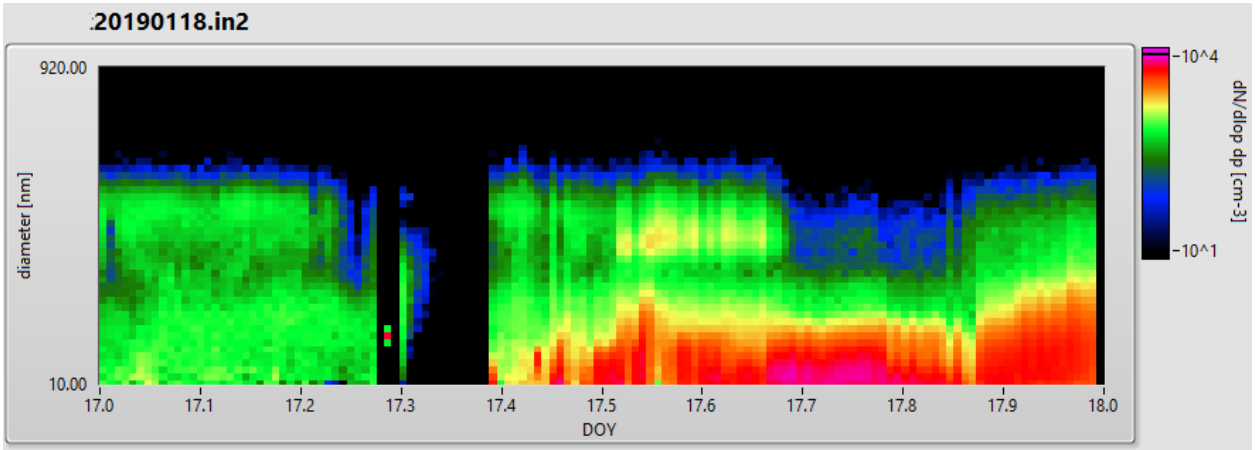

Fig 4: Zero check

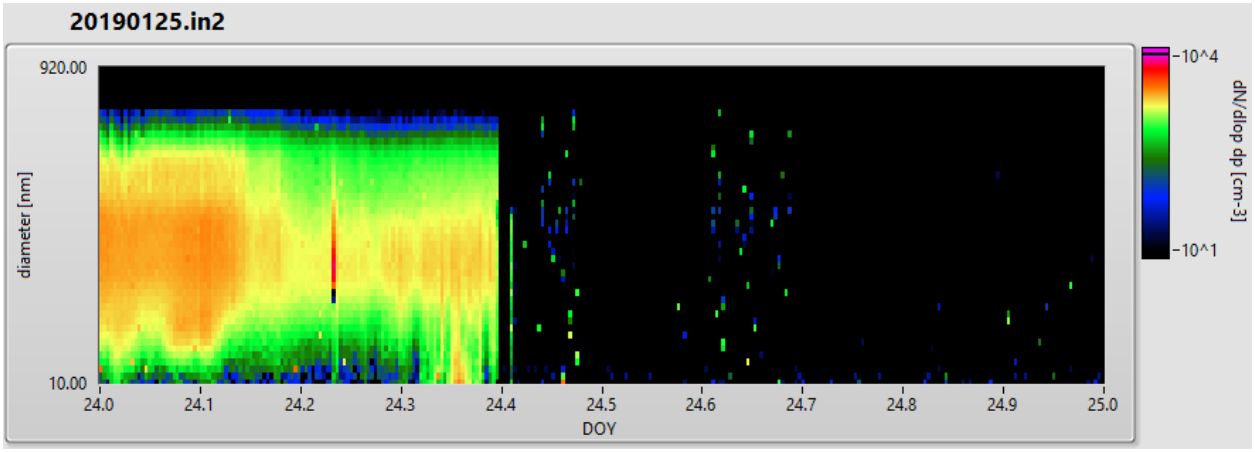

Fig 5: Lack of CPC working fluid

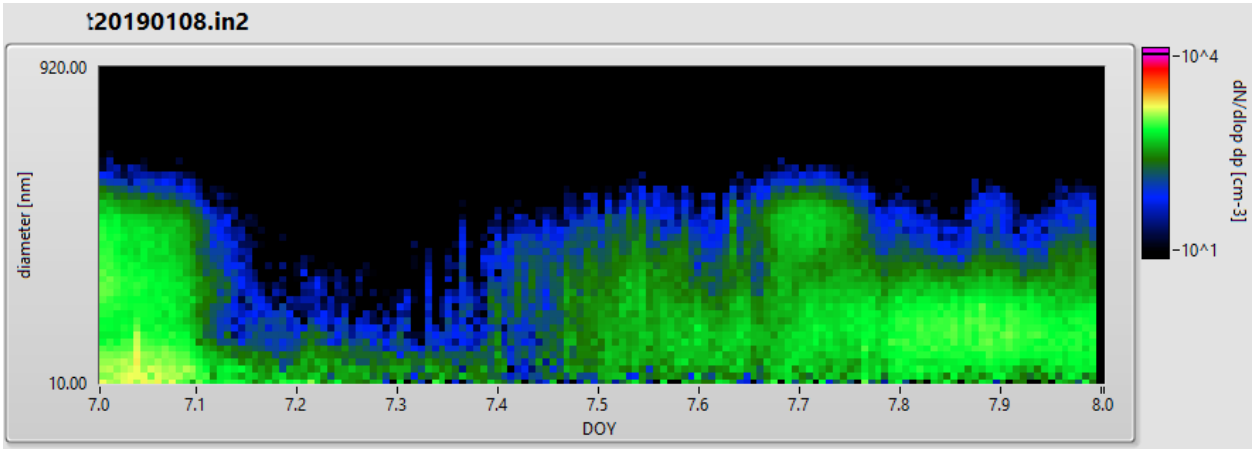

Fig 6: This is not a faulty measurement, just a period of clean air!

Appendix B: Problems identified by frequency plots/time series plot

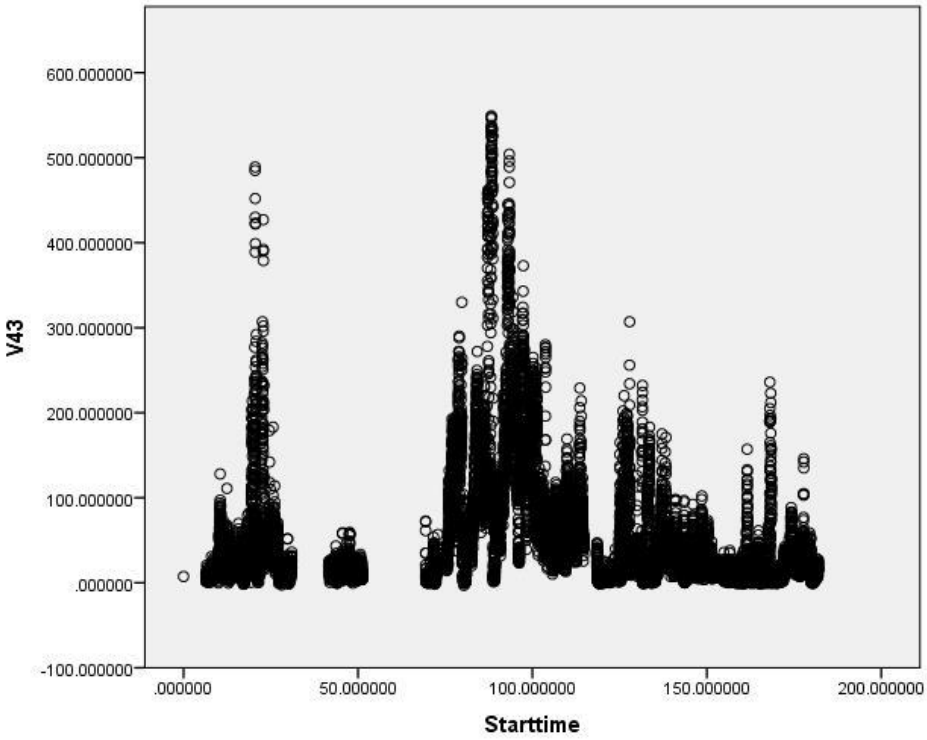

Fig 7: Time series plot bin 35 looks ok

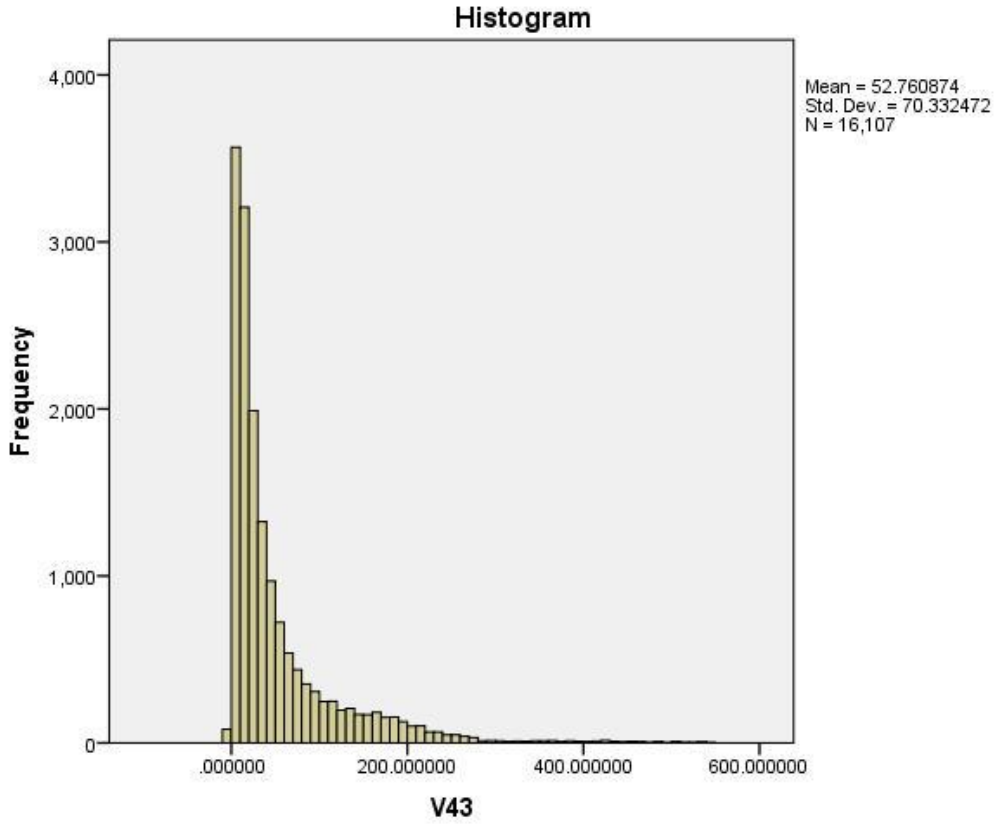

Fig. 8: But the histogram reveals almost 100 values less than zero due to inversion artefacts

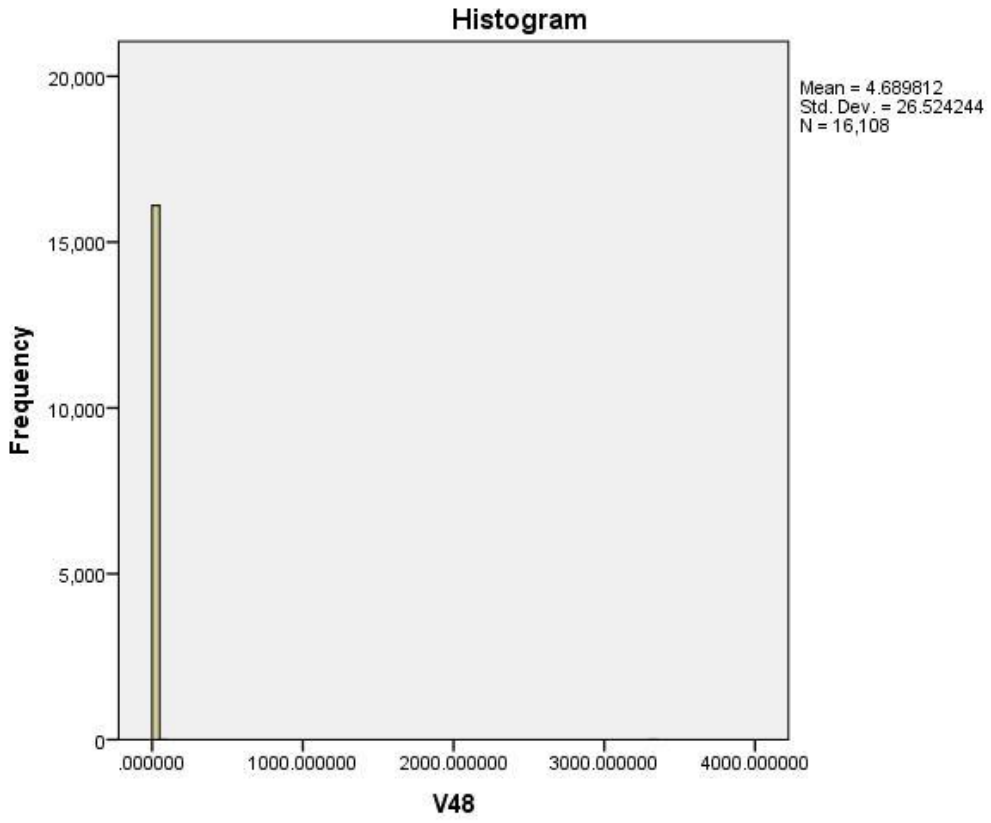

Fig 9: The histogram of bin 48 has maximum of 4000 at a mean value of 4.7

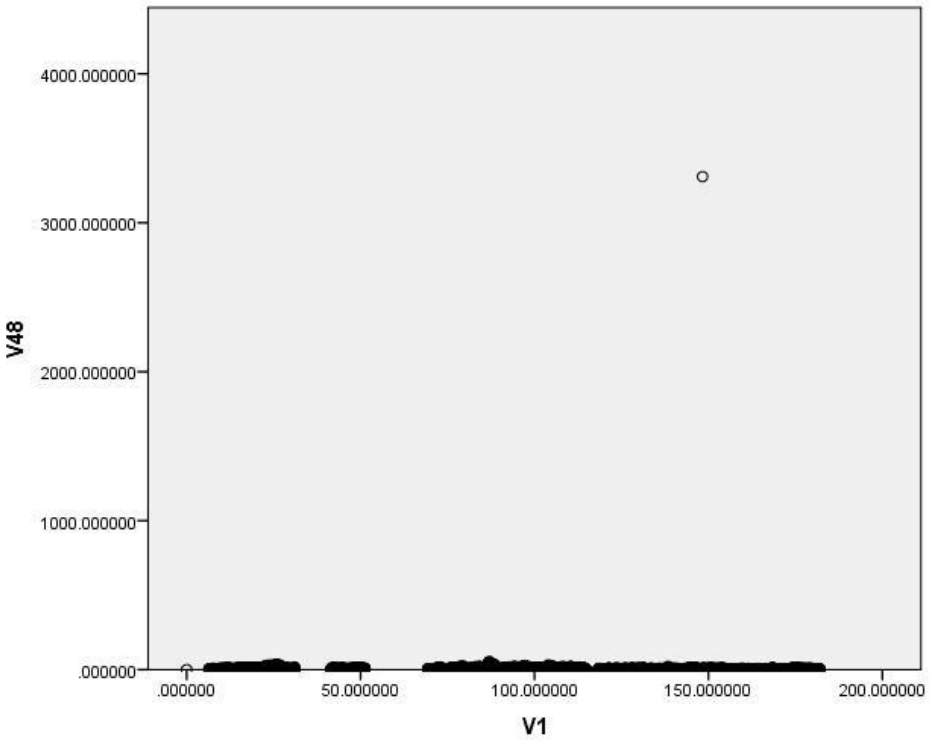

Fig 10: Due to one single outlier

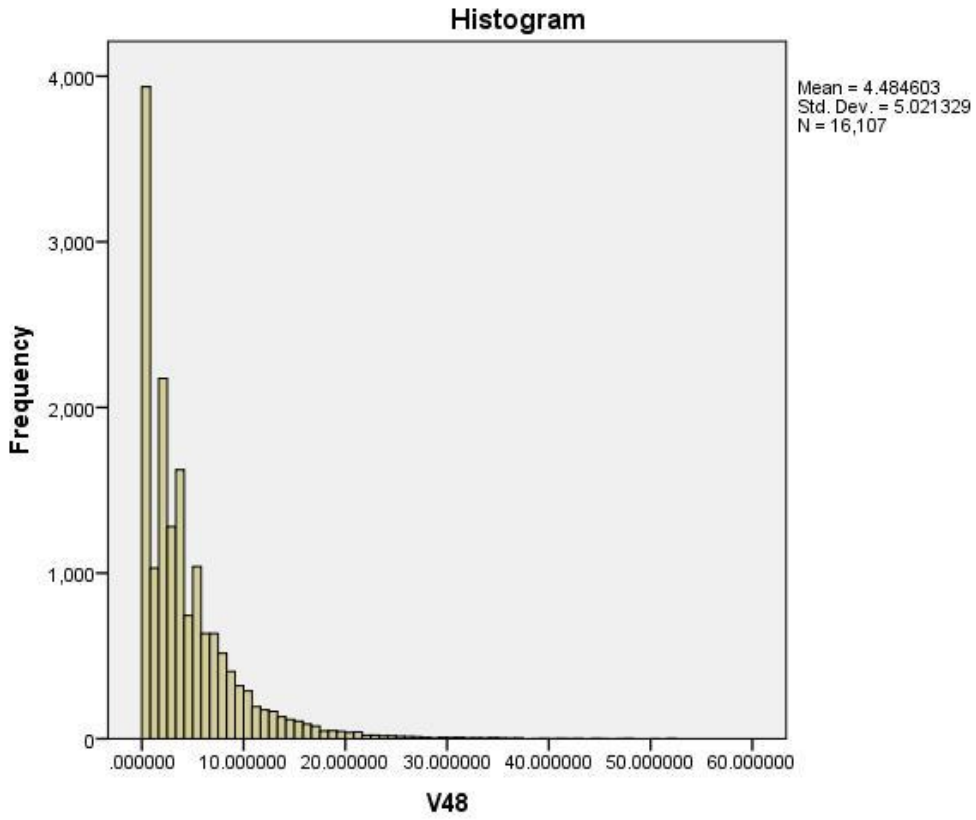

Fig 11: With this outlier removed the histogram looks good

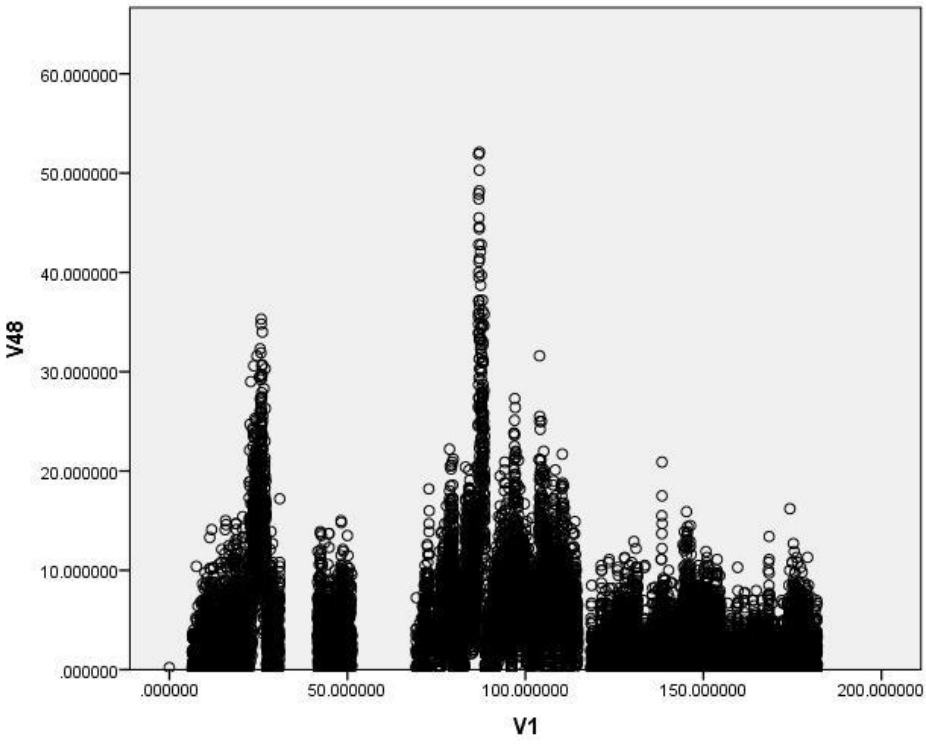

Fig 12: And so does the time series