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MEASUREMENT OF PARTICLE NUMBER SIZE DISTRIBUTIONS IN AMBIENT AIR USING THE MOBILE AEROSOL SPECTROMETER

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MEASUREMENT OF PARTICLE NUMBER SIZE DISTRIBUTIONS IN AMBIENT AIR USING THE MOBILE AEROSOL SPECTROMETER

1. Purpose and applicability

This OP contains the protocol for performing measurements of particle number size distribution in outdoor air for the EU-multicenter study ULTRA-2. The spectrometer consists basically of an differential electrical mobility particle analyzer (DMPS, TSI) and an optical particle spectrometer (LAS-X, PMS). This is an evaluation version of an anticipated standard operating procedure (SOP) which will result from experiences with this OP. Due to this fact this OP is subject to changes. Every addition to this OP will be added as an Appendix during this study.

2. Definitions

d mobility equivalent particle diameter in µm
CPC condensation particle counter
DMA differential electrical mobility particle analyzer without CPC
DMPS Differential electrical mobility particle analyzer
LAS Laser aerosol spectrometer

Main particle size ranges:

$NC_{0.01-2.5}$: total particle number concentration (cm^{-3}) diameter range between 0.01 - 2.5 µm
$NC_{0.01-0.1}$: particle number concentration (cm^{-3}) diameter range between 0.01 - 0.1 µm
$NC_{0.1-1.0}$: particle number concentration (cm^{-3}) diameter range between 0.1 - 1.0 µm
$NC_{1.0-2.5}$: particle number concentration (cm^{-3}) diameter range between 1.0 - 2.5 µm

Sub particle size ranges:

$NC_{0.01-0.03}$: particle number concentration (cm^{-3}) diameter range between 0.01 - 0.03 µm
$NC_{0.03-0.1}$: particle number concentration (cm^{-3}) diameter range between 0.03 - 0.1 µm
$NC_{0.1-0.5}$: particle number concentration (cm^{-3}) diameter range between 0.1 - 0.5 µm
$NC_{0.5-1.0}$: particle number concentration (cm^{-3}) diameter range between 0.5 - 1.0 µm

OP: Operating procedure

3. References

Brand, P., Gebhart, J., Below, M., Georgi, B., Heyder, J. (1991), Characterisation of Environmental Aerosol on Helgoland Island, *Atmospheric Environment*, **25A**, 581-585.

Brand, P., Ruoß, K. (1992) Technical note: Performance of a Mobile Aerosol Spectrometer for in situ characterisation of an environmental aerosol in Frankfurt city, *Atmospheric Environment*, **26A**, 2451-2457.

Tuch, Th., Brand, P., Wichmann, H.E., Heyder, J. (1997) , Variation of Particle Number and Mass Concentration in Various Size Ranges of Ambient Aerosols in Eastern Germany., *Atmospheric Environment*, **31**,4193-4197.

4. Discussion

NA

5. Responsibilities

NA

6. Equipment and materials

6.1 Equipment

6.1.1 DMPS

a) DMA complete (TSI, model 3071A)
b) CPC complete (TSI, model 3010)
c) Kr85 neutralizer (Model 3077. TSI)
d) Spare parts DMPS (filters, fittings, tubing)
e) Spare parts CPC (butanol, filters)

6.1.2 LAS

a) LAS complete (sample flow: 1 cm^3/s)
b) Spare parts (filters, cleaning equipment, fittings, tubing)

6.1.3 Data Acquisition

a) MAS-Software Version 15

6.1.4 Flow Control

a) Gilibrator

6.2 Materials

a) filters for DMPS: TSI P/N 1602015, Gelman 12144
b) filter for CPC: TSI P/N 1602028, TSI P/N 1602059, Balston P/N DFU-9922-05-AQ
c) filters for LAS: Pall P/N FLF6001N
d) 1-butanol extra pure (Merck)

6.3 Paper materials

a) Field forms to record performance parameters of DMPS in the field
b) Field forms to record performance parameters of LAS in the field
c) Laboratory book MAS

7. Procedures

ALL DAILY AND WEEKLY MAINTENANCE PROCEDURES MUST BE RECORDED IN THE RESPECTIVE FIELD FORM (Figure 1+2)

7.1 Daily maintenance of DMPS

a) clean impactor plate
b) check butanol level in CPC, fill if necessary (do not leave refill bottle connected to the CPC)
c) check indicator LED on CPC front
d) check and record:
e) Monodisperse Flow indicated on DMA voltmeter
f) Excess Air Flow indicated on DMA voltmeter
g) Sheath Air Flow indicated on DMA voltmeter
h) Pressure Drop of impactor
i) If pressure drop differs from normal value more than 2 cm H_20: Check the sample flow. If it deviates from nominal value adjust the pressure drop. The pressure drop may change as a result of narrowing of the impactor nozzle, while the flow remains (almost) not changed.
j) Adjust flow rates if different from nominal value and indicated value is larger than 0.01 meter range 100V, nominal values should be indicated on the instrument and in the diary.
k) repeat e) through i) until no changes in flow rates are noticeable after the made adjustments
l) if any adjustment was necessary record old and new values
m) check if deviation between 3 consecutive hourly CPC number concentration and DMPS indicated number concentration is within 30 % each other.

If any adjustment yields no sufficient result (i.e. normal operating conditions can not be achieved) or number concentration differences between CPC and DMPS remain larger than 30 % personal doing the routine maintenance is requested to report to an experienced operator as soon as possible.

7.2 Daily maintenance of LAS

check and record:
a) sample and sheath air flow indicated on rotameters of the instrument
b) Laser reference meter
c) inform an experienced operator to clean mirror and/or Brewster window if reference voltage is lower than 5.5 V. (Try and find out before the campaign whether a laser-reference will survive the next 24 hours or not, extensive cleaning activities should be avoided). Avoid laser-reference values larger than 9.5 Volts.
d) adjust air flow rates if rotameter indicated flowrates deviate more than 30 % from nominal values. (this large range is necessary due to the relatively bad flow meters of the LAS). Use the Gilibrator for this adjustment.
e) Check number concentration in the overlap area between LAS and DMPS. Visually check 3-5 particle size distributions by an experienced operator every working day. This can be done using remote data access.

If any adjustment or cleaning yields no sufficient result (i.e. normal operating conditions can not be achieved) or number concentration differences in the overlap range between LAS and DMPS/ are larger than typical personal doing the routine maintenance is requested to report to an experienced operator as soon as possible.

7.2 Weekly maintenance of DMPS

a) Drain CPC and refill with fresh butanol (to be done by routine personal). Record the time of refill operation.
b) Visually check coarse filter of the DMA sheath air inlet. Replace if necessary.
c) Check zero count rate of the DMPS:
    - Switch high voltage manually to zero
    - CPC countrate must be smaller than 0.04 cm^{-3}
    - Switch high voltage manually to 5 kV
    - Apply high concentrated aerosol to sample line and inlet of the DMA
    - CPC readout must not be affected by this aerosol
    - Switch high voltage back to external adjustment

7.4 Weekly maintenance of LAS

a) check sizing properties with at least one Latex aerosol (0.3µm)
b) if the peak of the monodisperse aerosol located in the wrong channel (difference larger than 1 channel compared to the usual value at the given laser reference) adjust nozzle (Only if necessary, try to avoid this ! This operation will usually take very long and cause a missing value in daily averages, adjustment may only be performed by primary operator)

7.5 Weekly calibration procedures

a) mobility calibration

7.6 Biweekly maintenance of DMPS (to be done by an experienced operator)
RECORD ALL ACTIONS IN THE LABORATORY BOOK OF THE MAS

a) clean impactor nozzle using ultrasonic bath.
b) visually check sheath air filter, replace if necessary.
c) check all flow rates including CPC and pressure drop with the Gilibrator.
d) check for leakage after reconnection of all fittings.
e) change Labels with nominal flow rates on the DMA and in the field form for daily maintenance if necessary and inform routine personal about changes.

7.7 Biweekly maintenance of LAS (to be done by an experienced operator)
TRY TO BE AT THE STATION WHEN ROUTINE MAINTENANCE IS DONE. CHECK IF THE ROUTINE MAINTENANCE IS STILL DONE ACCORDING TO THIS OP.

RECORD ALL ACTIONS IN THE LABORATORY BOOK OF THE MAS

a) check sample flow with Gilibrator, adjust if necessary
b) change operation parameters in daily maintenance field form and inform routine personal if necessary
c) Check agreement of total particle number concentration using hourly or daily averages
d) Check agreement between $PM_{2.5}$ and $MC_{0.01\text{-}2.5}$. The typical difference should be smaller than 20 %. Occasional values up to 30 % difference may occur.

ANY EXCESS MAINTENANCE ON THE MAS IS TO BE AVOIDED UNLESS ABSOLUTELY NECESSARY. IF THE INSTRUMENT IS RUNNING SMOOTHLY ONLY MAKE CHANGES ACCORDING TO THIS OP.

7.8 Beginning and end of the study

Check Activity of the Kr85 neutralizer**.** Use an appropriate counter. Record count rate, counter model and serial number of the counter in the laboratory book of the MAS. Activity may be checked at a marked point of the housing of the DMA.

8. Analytical procedures

8.1 Averaging of hourly and daily mean size distributions

Strange particle size distributions must only be rejected from the data set if the unusual size distributions are caused by an instrument failure.
Hourly average concentrations will be calculated if at least 66% of the data for one hour are available. Daily average number concentrations are valid, if at least 16 hourly averages are available for one day.
Daily means will be provided for time intervals 12:00 to 12:00 hours next day.

8.2 Particle number concentrations and Spectral data

8.2.1 Calculation of particle number concentrations

Number concentrations of the total spectrum and the following size ranges will be given as hourly and daily mean values:

Main particle size ranges:

$NC_{0.01-2.5}$: particle number concentration (cm^{-3}) diameter range between 0.01 - 2.5 µm
$NC_{0.01-0.1}$: particle number concentration (cm^{-3}) diameter range between 0.01 - 0.1 µm
$NC_{0.1-1.0}$: particle number concentration (cm^{-3}) diameter range between 0.1 - 1.0 µm
$NC_{1.0-2.5}$: particle number concentration (cm^{-3}) diameter range between 1.0 - 2.5 µm

Sub particle size ranges:

$NC_{0.01-0.03}$: particle number concentration (cm^{-3}) diameter range between 0.01 - 0.03 µm
$NC_{0.03-0.1}$: particle number concentration (cm^{-3}) diameter range between 0.03 - 0.1 µm
$NC_{0.1-0.5}$: particle number concentration (cm^{-3}) diameter range between 0.1 - 0.5 µm
$NC_{0.5-1.0}$: particle number concentration (cm^{-3}) diameter range between 0.5 - 1.0 µm

8.2.2 Calculation of daily average particle number distribution

Daily average particle number distributions will be calculated and supplied as separate files to the coordinating center. These files will contain diameter and dN/dlogd. The method to calculate concentrations in size ranges different from the main and sub-particle size ranges as given in 8.2 and 8.5 will be added as a Text file to the complete data set.

8.3 Quality control

Quality control of the data is achieved on daily basis by:

a) checking the critical operation parameters of the instruments (flow rates etc., see 8.3.1)
b) comparing number concentrations in the overlapping size ranges of different instruments (see 8.3.2)
c) visual checks of hourly averaged size distributions (see 8.3.3)
d) Check of the daily average particle number concentration of MAS and CPC

8.3.1 Check for critical operation parameters

The following operation parameters are checked according to this OP:

a) sheath and excess flow rates of DMPS should be within 1% of the set points
b) sample flow rate of DMPS should be within 5% from 0.61 l/min
c) sample flow rate of LAS should be within 10% from 0.06 l/min
d) the reference voltage of LAS should not be lower than 5.5 V.
e) sizing accuracy of DMPS as determined with PSL is within 10%
f) sizing accuracy of LAS as determined with PSL should be within one LAS channel

If one or more operating parameters are deviating from the nominal, the instrument in question should be checked and adjusted to the nominal parameters as soon as possible. Data collected with that instrument during the period of malfunction should be corrected, if possible, for the observed deviations

8.3.2 Comparison of number concentrations in the overlapping size ranges

Hourly averaged number concentrations in the following overlap regions are checked:

a) MAS vs. CPC 10 - 2500 nm

If two instruments deviate for 3 consecutive hours by more than 30%, both instruments should be checked as soon as possible by an experienced operator. If instrument is found to be malfunctioning, it should be adjusted to normal operation. Data collected with that instrument during the period of malfunction should be corrected, if possible, for the observed deviations from nominal operating parameters (see 8.4). If no malfunctioning is found, data from both instruments is accepted as is and is not rejected. However, it should be marked appropriately to indicate the discrepancy between the two instruments.

8.3.3 Visual checks of hourly averaged size distributions

Hourly-averaged size distributions should be checked visually by an experienced operator for persistent (for more than 3 hours in a row) “gaps”, i.e. whether the spectra appear to be “missing” particles at some sizes.

If such persistent “gaps” are found the instruments should be checked as soon as possible by an experienced operator. If instrument is found to be malfunctioning, it should be adjusted to normal operation. Data collected with that instrument during the period of malfunction should be corrected, if possible, for the observed deviations from nominal operating parameters (see 8.4). If no malfunctioning is found, data from the instruments is accepted as is and is not rejected.

8.4 Correction of data for recoverable instrumental malfunction

If an instrumental malfunction was found, the data can be corrected for the observed deviation, in the following cases:

a) sample flow of DMPS deviates by less than 30% from 0.61 l/min
b) sheath /excess flows of DMPS deviate by less than 30% from 6.1 l/min
c) sample flow of LAS deviates by less than 50% from 0.06 l/min

Corrected data should be marked, the reason and the procedure of correction should be described.

8.5 Data validation

The data are considered invalid, if it is discovered that the instrument was operated outside the nominal operating conditions, for which no correction can be applied, or if one or more critical parts of the instrument (i.e. laser of CPC or LAS, pumps, empty butanol reservoir of CPC, etc.) were not functioning or were out of order.

8.6 Calculation of particle mass concentrations

Particle number distributions are converted into particle volume concentrations by the MAS program. These particle volume distributions are then converted into particle mass distributions using an apparent density of 1530 kg m^{-3}. This apparent density has been calculated from approximately 1000 parallel measurements of PM2.5 obtained with a Harvard Impactor collocated at the measurement site of the MAS in Erfurt and the respective particle volume concentration in the size range 0.01 µm -2.5 µm obtained from MAS measurements. Mass concentrations of the total spectrum and the following size ranges will be given as hourly and daily mean values:

Main particle size ranges:

$MC_{0.01\text{-}2.5}$: total particle mass concentration (cm^{-3}) diameter range between 0.01 - 2.5 µm
$MC_{0.01\text{-}0.1}$: particle mass concentration (cm^{-3}) diameter range between 0.01 - 0.1 µm
$MC_{0.1\text{-}1.0}$: particle mass concentration (cm^{-3}) diameter range between 0.1 - 1.0 µm
$MC_{1.0\text{-}2.5}$: particle mass concentration (cm^{-3}) diameter range between 1.0 - 2.5 µm

Sub particle size ranges:

$MC_{0.01-0.03}$: particle mass concentration (cm^{-3}) diameter range between 0.01 - 0.03 μm
$MC_{0.03-0.1}$: particle mass concentration (cm^{-3}) diameter range between 0.03 - 0.1 μm
$MC_{0.1-0.5}$: particle mass concentration (cm^{-3}) diameter range between 0.1 - 0.5 μm
$MC_{0.5-1.0}$: particle mass concentration (cm^{-3}) diameter range between 0.5 - 1.0 μm

8.7 Comparison of $MC_{0.01-2.5}$ with $PM_{2.5}$

Daily average total mass concentration $MC_{0.01-2.5}$ measured by the MAS and a parallel running $PM_{2.5}$ Harvard Impactor should not differ more than 30 %. If there is no instrumental reason for a larger difference data should be marked in the data set but should not be rejected.

8.8 Data storage

All original spectral data and the cleaned sets of data will be stored in duplicate on appropriate media for further re-evaluations.

9.0 Attachments.

Figure 1. ULTRA II – Field Form Daily/Weekly DMPS Checks
Figure 2. ULTRA II – Field Form Daily/Weekly LAS-X Checks

Figure 1. ULTRA II – Field Form Daily/Weekly DMPS Checks

		Check:							Remarks:	Signature
						clean	visible	green	<---nominal values, only to be changed by primary operator	
Day	**Date**	**Pressure drop**	**Mono**	**Poly**	**Sheath**	**Impact Plate**	**Butanol**	**LEDs**	**e.g. flow adjustement, CPC refill, operator call**	
Monday										
Tuesday										
Wednesday										
Thursday										
Friday										
Saturday										
Sunday										

Butanol Change: date/time:	Sheath air filter:	Zero measurement:	

gray fields may only be filled by primary operator

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Figure 2. ULTRA II – Field Form Daily/Weekly LAS-X Checks

ULTRA II - Field Form Daily/weekly DMPS Checks

			Check:				Remarks:	Signature:
				20 +/- 5 cm³/s	5.5-9.5 V	gap < 30%	<-------nominal values, only to be changed by primary operator	
Day	**Date**		**Sample Flow**	**Sheath Air**	**Laser Voltage**	**Overlap LAS/DMPS**	**e.g. flow adjustement, Clean Mirror, operator call**	
Monday		old						
		new						
Tuesday		old						
		new						
Wednesday		old						
		new						
Thursday		old						
		new						
Friday		old						
		new						
Saturday		old						
		new						
Sunday		old						
		new						

weekly maintainance date:

Operator:	start time: stop time:	sample flow [cm³/min] old: new:	
Mobility calibration:	Latex Check: Range/Channel	Zero counts:	
Filename:	Filename:	Filename:	

gray fields may only be filled by primary operator

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