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MEASUREMENT AND ANALYSIS OF PARTICLE NUMBER SIZE DISTRIBUTIONS IN AMBIENT AIR USING THE DAS

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MEASUREMENT AND ANALYSIS OF PARTICLE NUMBER SIZE DISTRIBUTIONS IN AMBIENT AIR USING THE DAS

1. PURPOSE AND APPLICABILITY

This OP contains the protocol for performing measurements of particle number size distribution (10 nm - 10 μm in diameter) in outdoor air for the EU-multicenter study ULTRA-2. The spectrometer consists basically of a modified Scanning Mobility Particle Sizer (SMPS, TSI), 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

CPC condensation particle counter
DMA differential electrical mobility particle analyzer without CPC
SMPS Scanning Mobility Particle Sizer
LAS Laser aerosol spectrometer
Gillibrator: soap-bubble flow meter

Main particle size ranges:
$NC_{0.01-2.5}$: total particle number concentration (cm^{-3}) in the size range 0.01 - 2.5 μm
$NC_{0.01-0.1}$: particle number concentration (cm^{-3}) in the size range 0.01 - 0.1 μm
$NC_{0.1-1.0}$: particle number concentration (cm^{-3}) in the size range 0.1 - 1.0 μm
$NC_{1.0-2.5}$: particle number concentration (cm^{-3}) in the size range 1.0 - 2.5 μm

Sub particle size ranges:
$NC_{0.01-0.03}$: particle number concentration (cm^{-3}) in the size range 0.01 - 0.03 μm
$NC_{0.03-0.1}$: particle number concentration (cm^{-3}) in the size range 0.03 - 0.1 μm
$NC_{0.1-0.5}$: particle number concentration (cm^{-3}) in the size range 0.1 - 0.5 μm
$NC_{0.5-1.0}$: particle number concentration (cm^{-3}) in the size range 0.5 - 1.0 μm

OP: operating procedure

3. REFERENCES

Khlystov, A. et al. (1998) .... to be submitted.
Wang, S.C. and Flagan, R.C. (1990) Scanning electrical mobility spectrometer. Aerosol Sci. Technol. 13, 230.
Yoo, S.-H., Chae, S.-K., Liu, B.Y.H. (1996) Influence of particle refractive index on the lower detection limit of light scattering aerosol counters. Aerosol Sci. Technol., 25, 1-10.

4. DISCUSSION

NA

5. RESPONSIBILITIES

NA

6. EQUIPMENT AND MATERIALS

6.1 Equipment

6.1.1 SMPS

a) DMA (Model EC3071, TSI)
b) Kr-85 neutralizer (Model 3077, TSI)
c) CPC (Model 3010, TSI)
d) personal computer (Advantech)
e) data acquisition card (PCL-818H, Advantech)
f) digital flow controllers (Model 5850S, Brooks)
g) PT100 element
h) spare parts SMPS (filters, fitting, tubing, computer cables)
i) spare parts CPC (filters, butanol)

6.1.2 LAS

a) LAS-X (PMS)
b) Spare parts (laser, filters, fitting, tubing, cleaning equipment)

6.1.3 Flow meter

a) Gillibrator

6.2 Materials

a) coarse filters for SMPS (Siemens)
b) HEPA filters for the sheath flow of DMA (Gelman Sci.)
c) HEPA filters for the sheath flow of CPC (Gelman Sci.)
d) HEPA filters for the sheath flow of LAS (Gelman Sci.)
e) 1-butanol extra pure (Merck)

6.3 Paper materials

a) Field forms to record performance parameters of SMPS in the field

b) Field forms to record performance parameters of LAS in the field
c) Laboratory forms to record data acquisition

7. PROCEDURES

Note: All of the maintenance procedures should be recorded in the laboratory book.

7.1 Daily maintenance of SMPS

a) check whether the voltage indicated by the program is changing sinchronically with the voltage indicated on the DMA
b) check butanol level in CPC, fill if necessary (do not leave refill bottle connected to the CPC). Record the time of filling.
c) check indicator LED on CPC front.
d) check (remotely) flows and their std of CPC, excess air and sheath air (see 8.3.1). This check needs to be recorded on a separate laboratory form, see 6.3.c).
e) check comparability with other instruments (see 8.3.2). This check needs to be recorded on a separate laboratory form, see 6.3.c.

7.2 Daily maintenance of LAS

a) check and record:
    1) sample and sheath air flow indicated on rotameters of the instrument
    2) laser reference meter

If laser reference voltage is lower than 4 V, inform an experienced operator to clean mirror and/or Brewster window. Avoid laser-reference values larger than 9 Volts.
Note 1: The given value of 4 V is the nominal value provided by the manufacturer, but is subject to changes for a specific instrument).
Note 2: Cleaning of the mirror/Brewster window can only be performed by experienced operators!

a) adjust air flow rates if rotameter indicated flow rates deviate more than 30 % from nominal values. Record the old values and time of adjustment. (this large range is necessary due to the relatively bad flow meters of the LAS)
b) check comparability with the SMPS in the overlap range (see 8.3.2)

7.3 Weekly maintenance of SMPS

a) drain CPC and refill with fresh butanol (to be done by routine personal).
b) visually check coarse filters of the DMA, replace if necessary
c) check and record sample flow rate with the Gilibrator, inform an experienced operator if the flow deviates by more than 10% from 1 l/min.
d) check null counts of the SMPS, while scanning, by placing an absolute filter in front

of the sample inlet. Record the time and file name.

e) check sizing properties with monodisperse PSL aerosol of 0.2 μm. Record the time and file name.

7.4 Weekly maintenance of LAS

a) check sample flow with Gilibrator
b) change label with operation parameters on front of the instrument and inform routine personal if necessary
c) check sizing properties with monodisperse PSL aerosol of 0.2 μm (to be done simultaneously with the calibration of SMPS, see 7.3). Record the time and file name.

7.5 Handling of persistent instrumental deviations

If any adjustment of the operating parameters (see 7.1 and 7.2) yields no sufficient result (i.e. normal operating conditions can not be achieved) or number concentration differences between the instruments (see 8.3) remain unacceptable, personal doing the routine maintenance is requested to report the problem and actions undertaken to solve it to an experienced operator as soon as possible.

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

7.6 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. 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

Hourly average concentrations are calculated if at least 66% of the valid data (see 8.5) 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.
Strange particle size distributions must only be rejected from the data set if the unusual size distributions are caused by an instrument failure (see 8.5).

8.2 Number concentrations and spectral data

8.2.1 Calculation of particle number concentrations per size range

The particle number concentrations, total as well as in the specified below size ranges, is done by summing up the number concentrations of hourly averaged (see 8.1) instrumental channels which fall into the specified size range. If the range boundary falls within one of the instrumental channels, only the proportional fraction of the concentration in that channel is added to the concentration in the size range, i.e. if 1/3 of the channel is in the specified size range, only one third of the concentration in that channel is attributed to that size range.

Number concentrations in the following size ranges are calculated:

Main particle size ranges:

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

Sub particle size ranges:

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

8.3.1. Check for critical operation parameters

The following operation parameters are checked on daily basis:

a) sheath and excess flow rates of SMPS should be within 1% of the set points of the flow controllers
b) the standard deviation of both sheath and excess flows of SMPS during one scan is within 2%
c) sample flow rate of SMPS should be within 10% from 1 l/min
d) sample flow rate of LAS should be within 10% from 0.3 l/min
e) the reference voltage of LAS should not be lower than 4 V (see note in 7.2a)
f) sizing accuracy of SMPS as determined with PSL is within 10%
g) 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 (see 8.4).

8.3.2. Comparison of number concentrations in the overlapping size ranges

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

a) SMPS vs CPC: 10 - 300 nm
b) LAS vs SMPS: 175-275 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) Agaps≅, i.e. whether the spectra appear to be Amissing≅ particles at some sizes.

If such persistent Agaps≅ 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 SMPS deviates by less than 50% from 1 l/min
b) sheath /excess flows of SMPS deviate by less than 50% from 10 l/min
c) std of sheath and excess flows in SMPS during one scan is less than 10%
d) sample flow of LAS deviates by less than 50% from 0.3 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 (see 8.4), 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 volume and mass concentrations

8.6.1. Convolution to particle volume distributions

The number concentration distribution data will be convoluted into particle volume distributions assuming spherical particles of the nominal particle diameter of the given size intervals.

8.6.2. Calculation of apparent particle density

From the daily means of the particle volume concentrations and the daily PM2.5 measurements an apparent density will be determined for each day which will be averaged over the entire measuring period.

8.6.3. Calculation of particle mass concentrations

Based on the particle volume distriubtion data and the mean apparent particle density particle mass concentration will be calculated for the following size ranges:

Main size ranges:
$MC_{0.01\text{-}2.5}$: total mass concentration ($\mu g\ m^{-3}$) in the size range between 0.01 - 2.5 μm
$MC_{0.01\text{-}0.1}$: mass concentration ($\mu g\ m^{-3}$) in the size range between 0.01 - 0.1 μm
$MC_{0.1\text{-}1.0}$: mass concentration ($\mu g\ m^{-3}$) in the size range between 0.1 - 1.0 μm
$MC_{1.0\text{-}2.5}$: mass concentration ($\mu g\ m^{-3}$) in the size range between 1.0 - 2.5 μm

Sub particle size ranges:
$MC_{0.01\text{-}0.03}$: mass concentration ($\mu g\ m^{-3}$) in the size range between 0.01 - 0.03 μm
$MC_{0.03\text{-}0.1}$: mass concentration ($\mu g\ m^{-3}$) in the size range between 0.03 - 0.1 μm

$MC_{0.1-0.5}$: mass concentration ($\mu g\ m^{-3}$) in the size range between 0.1 - 0.5 μm
$MC_{0.5-1.0}$: mass concentration ($\mu g\ m^{-3}$) in the size range between 0.5 - 1.0 μm

8.7. 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. DAS daily and weekly maintenance field form.
Figure 2. DAS daily maintenance lab form.

Figure 1. DAS daily and weekly maintenance field form.

DAS DAILY AND WEEKLY MAINTENANCE FIELD FORM

Daily procedures Year: 199...... Week number:......................

			SMPS				LAS						
			butanol fill		LED indicator			Flows					
	Date	Operator	yes / no	time of fill	LASER	TEMP		sample	sheath	laser, V	time	Notes	signiture
MO							old						
							new						
TU							old						
							new						
WE							old						
							new						
TH							old						
							new						
FR							old						
							new						
SA							old						
							new						
SU							old						
							new						

Weekly procedures date:.............................. start time:.......................... stop time:.......................... Operator:................................

SMPS	
Drain / refill CPC (date and time):	
Coarse filters replase (yes / no):	
Sample flow (l/min):	
Null-counts (counts, file name):	
PSL calibration (size, file name):	

LAS	
Sample flow (l/min):	
Null-counts (counts, file name):	
PSL calibration (size, file name):	

Notes: ....
....
....

Figure 2. DAS daily maintenance lab form.

DAS DAILY MAINTENANCE LAB FORM

Daily procedures

Year: 199......   Week number:.....................

	Date	Operator	SMPS				Overlap comparison		Notes	Signiture
			average flows, l/min	maximum deviatio from set point, %	maximum std during 1 scan, %	OK, yes /no	CPC vs SMPS	LAS vs SMPS		
MO			sheath .... excess .... CPC	sheath .... excess .... CPC	sheath .... excess .... CPC					
TU			sheath .... excess .... CPC	sheath .... excess .... CPC	sheath .... excess .... CPC					
WE			sheath .... excess .... CPC	sheath .... excess .... CPC	sheath .... excess .... CPC					
TH			sheath .... excess .... CPC	sheath .... excess .... CPC	sheath .... excess .... CPC					
FR			sheath .... excess .... CPC	sheath .... excess .... CPC	sheath .... excess .... CPC					
SA			sheath .... excess .... CPC	sheath .... excess .... CPC	sheath .... excess .... CPC					
SU			sheath .... excess .... CPC	sheath .... excess .... CPC	sheath .... excess .... CPC					

Explanation:

SMPS parameters are OK if *:

1) sheath and excess flows are within 1% of set points of the flowcontrollers (10 l/min)
2) sample flow is within 10% of 1 l/min
3) std deviation during one scan is lower than 2%

* the criteria may be changed