Intercomparison of mobility particle sizers (MPS)

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1 Introduction

Particles smaller than 100 nm in diameter are generally called ultrafine particles. Ultrafine aerosol particles are ubiquitous: They are found outdoors and indoors, in the environment as well as at workplaces. Most of them are derived from combustion and industrial processes, heating systems, and motor vehicles. The diesel engine is probably the dominant source of ultrafines in urban environment today. Adverse health aspects caused by the inhalation of ultrafine particles have been investigated intensively by e. g. *Oberdörster* [1].

However, neither standards nor commonly accepted guidelines exist defining requirements for suitable measuring devices and for a procedure and methodology to measure ultrafine aerosols. To provide support for ultrafine aerosol measurements at least for the workplace, ten European institutions active in the field of occupational safety and health published a preliminary convention on the principles which should be taken into account [2].

Several methods for measuring number concentrations and size distributions of ultrafine aerosols are known (see for example the electrical low-pressure impactor, ELPI). The measuring principle of combining a differential mobility analyzer (DMA) with a condensation particle counter (CPC) has turned out to be the preferable and most widely used method for environmental and workplace measurements. The physical principle is that the aerosol particles are first brought into charge equilibrium and then classified in a DMA according to their electrical mobility. Electrical mobility is a measure of the particle's ability to move in an electrical field and is inversely proportional to the particle diameter. The electrical mobility diameter is particularly relevant for ultrafine particles as it is related to their diffusion and deposition (e. g. in the human lung). Once the particles are classified, their number concentration is measured by a CPC. A more detailed description is given in the literature, e. g. *Hinds* [3] and *Willeke* and *Baron* [4]. Technologically advanced instruments are the Differential Mobility Particle Sizer (DMPS) and the Scanning Mobility Particle Sizer (SMPS) system. The latter one is commercially available and manufactured by TSI, St. Paul, MN (USA). There are several advantages and disadvantages using a DMPS or a SMPS. The DMPS measures particle size stepwise, uses a simple inversion routine, and is able to measure low and high particle number concentrations (10^2 to 10^7 particles/cm^3). However, sampling times of 5 to 15 min are required, which might be too long for rapidly changing number concentrations. The SMPS utilizes a continuous scanning mode instead of steps, providing complete particle size distributions for submicrometer aerosols in about 1 to 5 minutes. The SMPS has a more complex inversion program. Both systems need careful calibrations of the flows. Attention needs to be paid to use the correct delay time

Abstract *Mobility particle sizers are widely used for the measurement of ultrafine aerosols in the environment as well as at workplaces. The question arises about the comparability of the different systems' measurements. For assessing this, eleven institutes from Austria, Switzerland and Germany with their devices participated in an intercomparison exercise, which took place in the testing ground of the Institut für Gefahrstoff-Forschung – IGF in Dortmund. The test aerosols were generated by a diesel engine and by a soot generator. According to the outcome of this intercomparison it is essential to guarantee uniform sampling conditions and instrument parameters for getting comparable results. Attention should be paid to the correct adjustment of the flow rates and to the applied software. User skill is very important. On these conditions the MPS-technique is a valid tool to measure size distributions and number concentrations of ultrafine particles and the comparability of the results is regarded to be acceptable. The intercomparison will be repeated on refined conditions for finding out the variability and the limits of the different device configurations.*

Vergleichsmessungen von Mobilitätsspektrometern

Zusammenfassung *Mobilitätsspektrometer werden weit verbreitet zur Messung ultrafeiner Aerosole eingesetzt, sowohl in der Umwelt als auch an industriellen Arbeitsplätzen. Dabei stellt sich die Frage nach der Vergleichbarkeit der mit den verschiedenen Systemen erzielten Messergebnisse. Zur Beantwortung dieser Frage beteiligten sich elf Institute aus Österreich, der Schweiz und Deutschland mit ihren Geräten an einer Vergleichsuntersuchung, die im Technikum des Instituts für Gefahrstoff-Forschung – IGF in Dortmund stattfand. Als Aerosolquellen dienten ein Dieselmotoraggregat und ein Rußgenerator. Die Ergebnisse dieser Vergleichsuntersuchungen zeigten, dass zur Erzielung vergleichbarer Ergebnisse einheitliche Probenahmebedingungen und Geräteparameter einzuhalten sind. Insbesondere ist auf die Kalibrierung der Luftdurchsatzraten und auf die Auswerte-Software zu achten. Die Anwendung der Gerätetechnik erfordert qualifiziertes Personal. Unter diesen Bedingungen erwies sich die eingesetzte Messtechnik als geeignet zur Messung der Anzahlkonzentration und der Partikelgrößenverteilung ultrafeiner Aerosole. Die Vergleichbarkeit der Messergebnisse ist akzeptabel. Die Vergleichsuntersuchungen sollen unter verfeinerten Bedingungen wiederholt werden, um die Variabilität und die Grenzen der verschiedenen Gerätekonfigurationen herauszufinden.*

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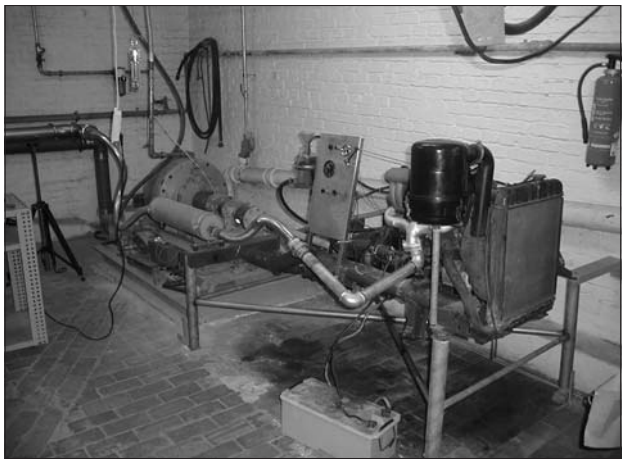

Figure 1 | **Diesel engine used for aerosol generation.**

within the SMPS-program to maintain a correct sizing. The number concentration measurement must exceed a concentration in the order of a few hundred to thousand particles/cm³ to provide good counting statistics or alternatively the scan time has to be increased to the order of the DMPS. The upper concentration limit can be as high as up to 10^7 particles/cm³ (depending on the SMPS-configuration).

It is essential that published data on the number concentrations and the particle size distributions of ultrafine aerosols must be comparable. The first question that arises is the comparability of the various DMPS- and SMPS-devices including the software used for operation and data.

On suggestion of the Institut für Gefahrstoff-Forschung – IGF, Bochum, eleven institutes from Austria, Germany, and Switzerland participated in an experimental intercomparison workshop at IGF's testing ground in Dortmund [5]. The purpose of this workshop was to compare different DMA-based particle sizers and to derive recommendations for the correct use of SMPS in workplace measurements.

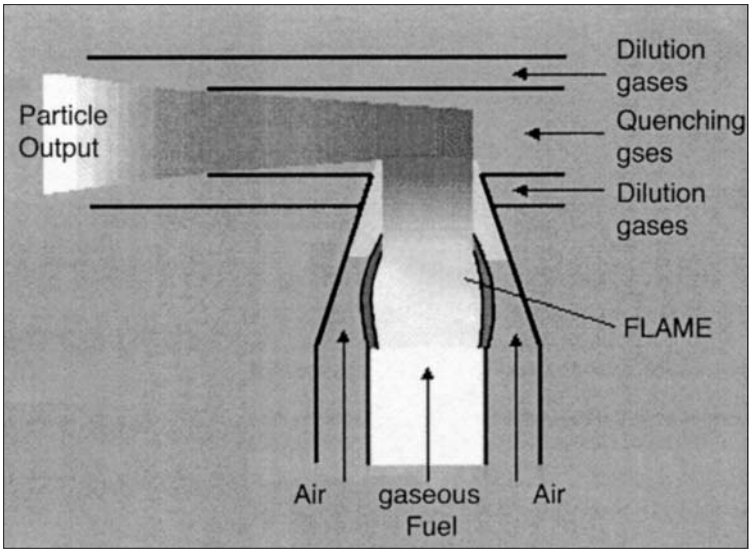

Figure 2 | **Schematic diagram of the soot generator.**

2 Participants and methodology

The following institutions participated in the intercomparison workshop (given in alphabetical order):

- Berufsgenossenschaftliches Institut für Arbeitssicherheit – BIA, Sankt Augustin, Germany,
- Fachhochschule Aargau, Windisch, Switzerland,
- Bundesforschungsanstalt für Landwirtschaft – FAL, Institut für Technologie und Biosystemtechnik, Braunschweig, Germany,
- Institut für Troposphärenforschung – IfT, Leipzig, Germany,
- Institut für Gefahrstoff-Forschung – IGF, Bochum, Germany,
- Institut für Maschinen-Messtechnik und Kolbenmaschinen – IMKO der Otto-von-Guericke Universität, Magdeburg, Germany,
- Labor für Festkörperphysik, ETH, Zürich, Switzerland,
- METAS, Swiss Federal Office of Metrology and Accreditation (formerly EAM, Eidgenössisches Amt für Messwesen), Bern, Switzerland,
- Ökozentrum Langenbruck, Langenbruck, Switzerland,
- Österreichische Staub- (Silikose-)Bekämpfungsstelle – ÖSBS, Leoben, Austria,
- Volkswagen AG, KEFUT, Wolfsburg, Germany.

All participants, except the IfT, used SMPS-systems of different ages, series and configurations. Here, three different SMPS-configuration types were employed. None of the SMPS-systems had been specifically checked or calibrated prior to the intercomparison to reflect instrument conditions as they were. The IfT used a Twin-DMPS [6] (TDMPS, two Hauke type DMAs with two different TSI CPCs) as an independent comparison spectrometer. This TDMPS was used together with one SMPS-system under conditions of fixed parameters during all the experiments whereas the variation of SMPS scan times and flows was an important aspect of this intercomparison workshop. Especially, different SMPS-configurations should be investigated for their comparability.

The experiments were supported by TSI in Aachen (Germany), the European Headquarters of the SMPS' manufacturing company, who provided assistance by technical staff throughout the complete workshop. This proved especially helpful since SMPS-systems need careful flow setting and skilled personnel for operation.

The measurements took place at the diesel test facility of IGF in Dortmund which is described in detail in [5], where a 44 kW aspiration type diesel engine was used as an aerosol generator (see **Figure 1**). In addition, a soot generator based on a quenched propane flame [7] was used alternatively (see **Figure 2**).

The IGF diesel test facility contains a room of about 20 m³ volume, which is constantly flushed at a very low flow rate of about 0.02 m³ exhaust gas per minute. It has been shown that the aerosol is homogeneously mixed in the room [8]. The aerosols were extracted from this homogenization chamber through a polished stainless steel tube (**Figure 3**) and distributed to each of the participating instruments (**Figure 4**) via a special manifold. This manifold guaranteed identical conditions for all participants (identical sampling line lengths, flow rates etc.). Every participant used the same length of flexible tubing made from electrically conducting material for connecting the manifold with the instrument. It was decided to keep the conditions of aerosol generation as simple as

Figure 3 | **Distribution of test aerosols via a manifold.**

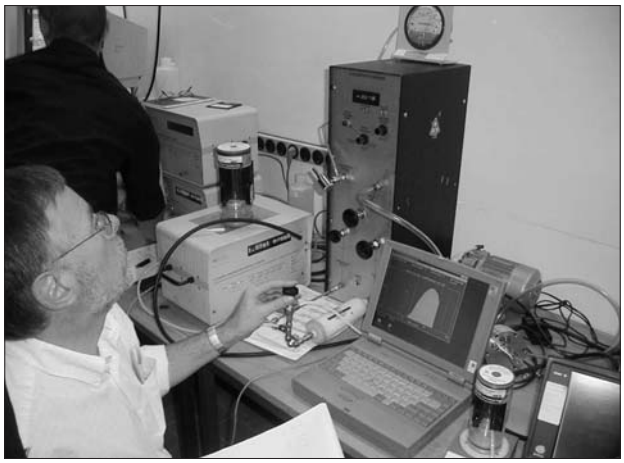

Figure 4 | **Typical measuring device of a participant.**

possible and to vary the concentrations in only three steps (plus a special set of experiments using the CAST™ device).

In total, 14 different runs of 30 or 60 minutes length (depending on the scan time of the instruments agreed upon in the respective run) were performed. Every participant recorded as many spectra as possible during these steady state periods of identical sampling conditions. The protocol shown in **Table 1** for the different runs was agreed on.

Remarks:

– Starting from run D, each participant used the SMPS-option "down scan first".

– As each run was supposed to consist of ten individual complete scans, those runs with a scan duration of 6 minutes ("Scan: 5/1") lasted a total of 60 minutes, all others (esp.: "Scan: 2/1") lasted a total of 30 minutes.

– During the evaluation, it was found that different versions of the TSI supplied SMPS-software yielded slightly differing results. As a consequence all results were then calculated using the software version 3.2. The TDMPS-data were inverted by a routine of *Stratmann* and *Wiedensohler* [9].

– The concentration levels were only general targets, and it was tried to reach them using the expertise of the test facility's operators (for a more detailed description of the levels reached see **Table 2**).

Table 1 | **Protocol of the different runs performed.**

Run identification	Aerosol-properties *	Scan	Flow	Integration-limits in nm
A	M	np	np	16 – 220
B	L	np	np	16 – 220
C	H	np	np	16 – 220
D	H	5/1	0.3/3	16 – 600
E	H	2/1	0.3/3	16 – 600
F	H	5/1	1/10	10 – 220
G	H	2/1	1/10	10 – 220
H	L	2/1	1/10	10 – 220
I	L	5/1	1/10	10 – 220
J	M	np	np	16 – 220
K	200 nm (CAST)	2/1	0.3/3	16 – 600
L	200 nm (CAST)	5/1	0.3/3	16 – 600
M	100 nm (CAST)	2/1	0.3/3	16 – 220
N	40 nm (CAST)	2/1	1/10	16 – 220

*

L = low concentration of diesel particulate matter (≈ 10^4 particles/cm³),

M = medium concentration of diesel particulate matter (≈10^5 particles/cm³),

H = high concentration of diesel particulate matter (≈10^6 particles/cm³).

np = no pre-set conditions, every participant used their default settings.

xxx nm (CAST) = CAST™-generated aerosol with maximum of number concentration at xxx nm.

The participants were asked to report their data in a standardized format using the spreadsheet given in **Table 3**. In addition, the participants reported their complete sets of primary data for a more detailed analysis including mode fittings. Because there was too much variation within the runs themselves, it was not to be expected that differences due to the construction and composition of the varying measurement trains (DMA-CPC combinations) would manifest themselves in differing curve shapes of the instruments. The analysis shows some general findings like standard deviations of the set of different SMPS with specific settings.

In parallel to the SMPS-experiments, a set of additional measurements was performed. The aerosol concentration was also monitored by stationary dust sampling over the complete period of one respective run. The results of these samplers [10] are reported here for comparison reasons, as they give an estimate of the general aerosol concentrations of DPM achieved in the experiments. The respective sampler (MPG II, DEHA Hahn und Wittmer, Friolzheim) was positioned directly in front of the inlet of the steel manifold, which distributed the aerosols to the participants. The results are given in Table 2. They show that the experiments were performed under conditions representative for real workplace atmospheres [11].

Table 2 | General concentration levels obtained.

Run identification	Respirable dust in mg/m³	EC in mg/m³	OC in mg/m³
A	0.28	0.047	0.064
B	nd (LOD=0.08)	nq (LOQ=0.02)	nq (LOQ=0.036)
C	0.59	0.190	0.164
D	0.43	0.187	0.158
E	0.54	0.202	0.189
F	0.58	0.232	0.223
G	0.47	0.157	0.168
H	0.25	0.035	0.134
I	0.15	0.027	0.071
J	nq (LOQ=0.21)	0.035	0.046
K	0.32	0.120	0.064
L	0.27	0.114	0.063
M	0.22	0.031	0.070
N	-		0.078

EC = elemental carbon

OC = organic carbon

nd (LOD=xxx) = "not detectable" (Limit of Detection=xxx mg/m³)

nq (LOQ=yyy) = "not quantifiable (Limit of Quantification=yyymg/m³)

3 Results and Discussion

The main goal of the intercomparison was to clarify basic quality management data of the MPS-systems. Therefore, the results obtained by the instruments were compared directly. They were reported by the participants using the spreadsheet given in Table 3.

From these spreadsheets, the following data were extracted for each instrument and run (within the integration limits given in Table 1):

- the median values for the particle diameters,
- the number concentrations.

Figures 5 to **8** show bar charts of both sets of data for a selection of runs (run E, G, H, I).

The horizontal line indicates the mean value for all participants. The x-axis shows the participants (random order), where participant A5 and B3 did not change their parameters during all the different runs. As can be seen in the example and during almost all other similar plots, these particular instruments were very close to the average of all the other participants.

Table 3 | Standardized report spreadsheet.

Institute/Company xxx

SMPS-System CPC-Type Ser.-No.

DMA-Type Ser.-No.

Flow-Control Ser.-No.

Software

Remarks

Evaluation with Charge Correction

	File		File		File		File		File		File	
	Sheath [l/min]		Sheath		Sheath		Sheath		Sheath		Sheath	
	Aerosol [l/min]		Aerosol		Aerosol		Aerosol		Aerosol		Aerosol	
	Up Scan [s]		Up Sc.		Up Sc.		Up Sc.		Up Sc.		Up Sc.	
	Down Scan [s]		Down Sc.		Down Sc.		Down Sc.		Down Sc.		Down Sc.	
	Delay [s]		Delay		Delay		Delay		Delay		Delay	
	Dp min [nm]		Dp min (View-Range)		Dp min		Dp min		Dp min		Dp min	
	Dp max [nm]		Dp max (View-Range)		Dp max		Dp max		Dp max		Dp max	
Scan	Dp-Mode[nm]	part/cm³	Dp-Mode	part/cm³	Dp-Mode	part/cm³	Dp-Mode	part/cm³	Dp-Mode	part/cm³	Dp-Mode	part/cm³
1												
2												
3												
4												
5												
6												
7												
8												
9												
10												

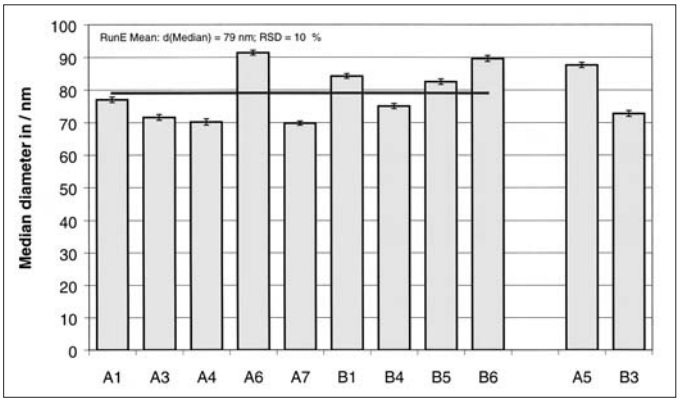

Figure 5 | UFP median diameters as measured in run E.

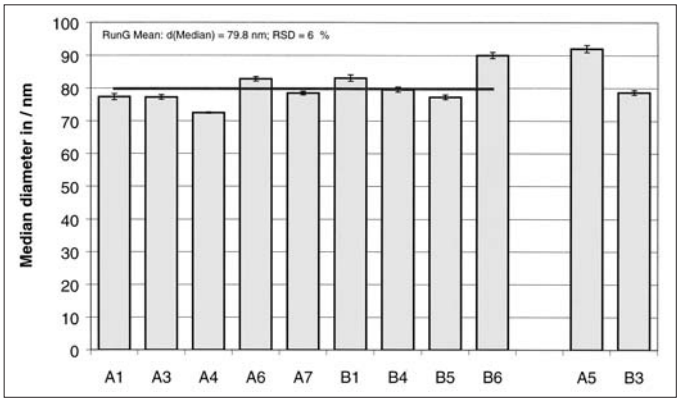

Figure 6 | UFP median diameters as measured in run G.

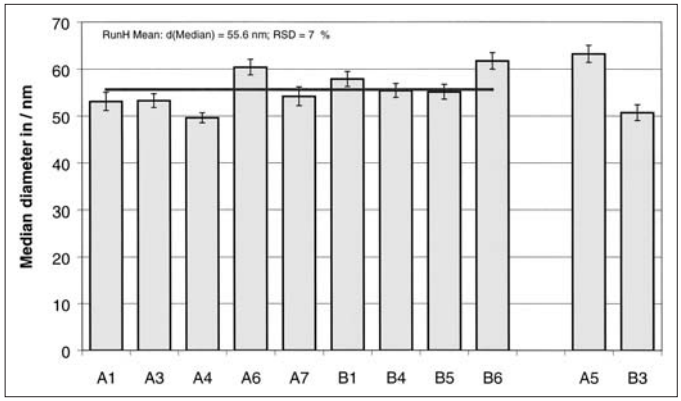

Figure 7 | **UFP median diameters as measured in run H.**

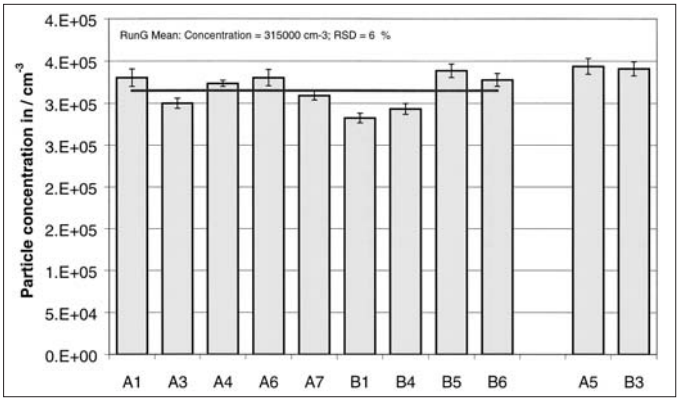

Figure 10 | **UFP particle number concentrations as measured in run G.**

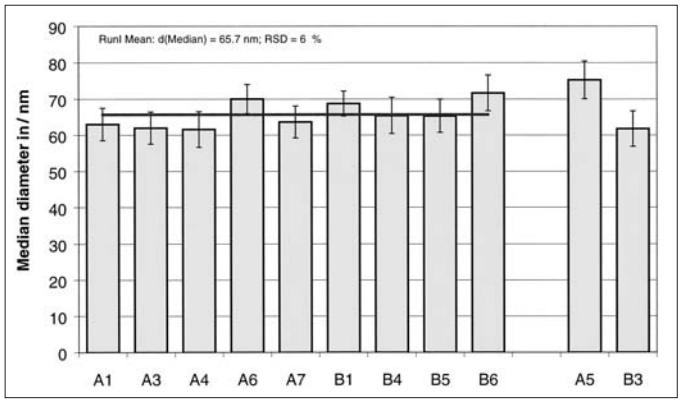

Figure 8 | **UFP median diameters as measured in run I.**

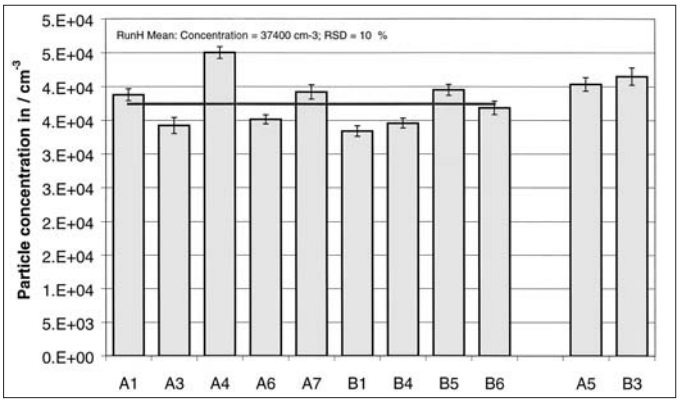

Figure 11 | **UFP particle number concentrations as measured in run H.**

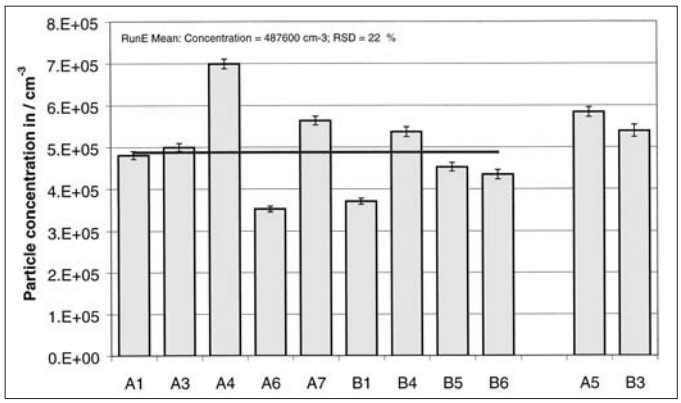

Figure 9 | **UFP particle number concentrations as measured in run E.**

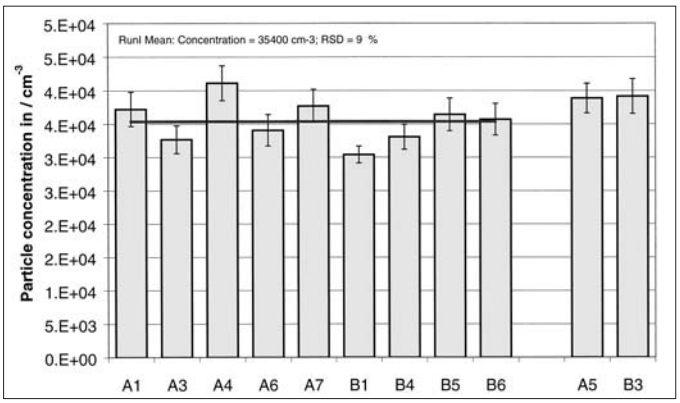

Figure 12 | **UFP particle number concentrations as measured in run I.**

Similar bar charts were obtained for the particle number concentrations. **Figures 9** to **12** show the respective data.

During the experiments the settings of the SMPS-systems were optimized and flows were corrected using an external flow meter (Gilibrator, Gillian Instrument Corp., Wayne, NJ, USA), which yielded significant improvements of performance in some cases. As an example the relative deviations of the median diameter measured by one participant from the average during all the runs (A to N in **Figure 13**) are shown. In this case, the mentioned intervention was performed after run D after which the deviations from the average decreased significantly.

These figures were selected to demonstrate some conclusions possibly to be drawn from the experiments. By comparison of the residual standard deviations (rsd) between the different instruments under specific conditions we derived the following statements:

Influence of sheath air flow: Under otherwise identical conditions (except integration limits), an increase of the sheath air flow rate decreases the rsd for particle size determination from 10 % at a flow rate of 3 l/min to 6 % at a flow rate of 10 l/min (see run E compared to run G, Figure 5 compared to Figure 6). A comparison of the rsd for the number concentrations does not make sense because of the differing integration parameters.

Influence of scan time: By increasing the scan time from 2 to 5 minutes there is no significant improvement being observed in the rsd for the particle diameter as well as for the

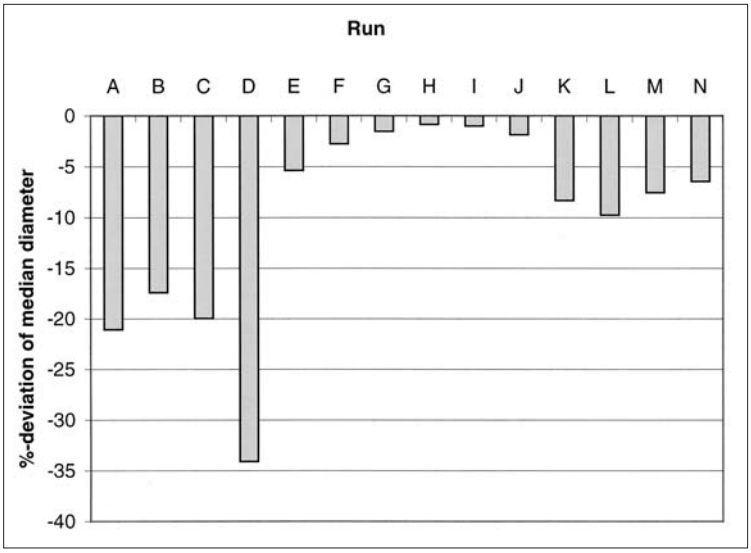

Figure 13 | **Deviations of UFP median diameters as measured by a specific participant laboratory.**

number concentration (see run H vs run I, Figures 7 vs 8, 11 vs 12).

Influence of particle concentration of the test aerosol: "High" particle concentrations in the test aerosol vs "low" ones (runs G vs I) cause lower rsd for the particle concentration measurement (Figure 10 vs 12) but does not improve the determination of the median diameter (Figure 6 vs 8).

4 Conclusions

Some general conclusions can be drawn from the intercomparison:

1) It is very important to guarantee uniform instrument parameters (this does include the software version used for evaluation) and sampling conditions for using the MPS-technique. There is urgent need for standardization.

2) Under conditions of "good practice" (especially by using the fixed parameters mentioned above), the SMPS-technique is a valid tool to measure size distributions and number concentrations of ultrafine particles. The comparability of results seems to be acceptable.

3) However, user skill is very important. SMPS-devices are no black boxes. The user must understand the critical parameters and the principles of operation.

4) All participants of the intercomparison workshop appreciated the improvement in their skills operating SMPS-systems, which turned out to be essential.

5) The experimental set-ups (test stand as well as CAST generator) were well-suited to the task.

The intercomparison will be repeated using the experience gained by the current experiments to find out the variability and limits of different SMPS-configurations.

A set of recommendations for the use of MPS in workplace measurements will be formulated and published elsewhere.

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Aus der Regelungsarbeit der EU

Neue Richtlinien der Europäischen Union zu Gefahrstoffen

In den Amtsblättern der Europäischen Gemeinschaft ist folgende Regelung zu Gefahrstoffen veröffentlicht worden:

Entscheidung der Kommission vom 14. Februar 2001 betreffend die Entscheidung über die mögliche Aufnahme bestimmter Wirkstoffe in Anhang I der Richtlinie 91/414/EWG des Rates (2001/134/EG)

Die Bewertung einer Reihe von Wirkstoffen in Pflanzenschutzmitteln hat ergeben, dass die übermittelten Informationen zum Nachweis, ob Pflanzenschutzmittel mit dem betreffenden Wirkstoff die Anforderungen gemäß Artikel 5 Abs. 1 Buchstaben a) und b) der Richtlinie 91/414/EWG unter den vorgeschlagenen Anwendungsbedingungen generell erfüllen oder nicht, nicht ausreichen. Eine Entscheidung, ob diese Wirkstoffe in Anhang I der Richtlinie 91/414/EWG aufgenommen werden können, ist somit zurzeit nicht möglich.

Diese Entscheidung der Kommission legt daher Randbedingungen (Verfahren und Fristen) fest, unter denen die Antragsteller für die 17 im Anhang zu dieser Entscheidung genannten Wirkstoffe die zur endgültigen Beurteilung zur Aufnahme in den Anhang erforderlichen Unterlagen vorzulegen haben. ABl. EG Nr. L 49 vom 20. Februar 2001, S. 13.

Dr. *Ulrich Welzbacher,*
Berufgenossenschaftliche Zentrale für Sicherheit und Gesundheit – BGZ, Sankt Augustin.