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Intercomparison of number concentration measurements by various aerosol particle counters[☆]

A. Ankilov [1], A. Baklanov [1], M. Colhoun [2], K.-H. Enderle [3],
J. Gras [4], Yu. Julanov [5], D. Kaller [6], A. Lindner [7],
A.A. Lushnikov [5], R. Mavliev [1], F. McGovern [2], A. Mirme [8],
T.C. O'Connor [2], J. Podzimek [8], O. Preining, G.P. Reischl,
R. Rudolf [10], G.J. Sem [9], W.W. Szymanski, E. Tamm [8],
A.E. Vrtala [11], P.E. Wagner [*], W. Winklmayr [6], V. Zagaynov [5]

Institut für Experimentalphysik, Universität Wien, Boltzmanngasse 5, A-1090 Vienna, Austria

Abstract

Total aerosol particle number concentrations, as measured by means of 16 different measurement systems, have been quantitatively compared during an international workshop at the Institute for Experimental Physics of the University of Vienna, Austria, which was coordinated within the Committee on Nucleation and Atmospheric Aerosols (ICCP-IUGG). The range of measuring instruments includes Pollak counters (PCO) in use already for several decades, presently available commercial particle counters, as well as laboratory prototypes. The operation of the instruments considered was based on different measurement principles: (1) adiabatic expansion condensation particle counter, (2) flow diffusion condensation particle counter, (3) turbulent mixing condensation

particle counter, (4) laser optical particle counter, and (5) electrostatic particle measurement system. Well-defined test aerosols with various chemical compositions were considered: DEHS, sodium chloride, silver, hydrocarbons, and tungsten oxide. The test aerosols were nearly monodispersed with mean particle diameters between 4 and 520 nm, the particle number concentrations were varied over a range from about 4×10^1 to 7×10^6 cm^{-3}. A few measurements were performed with two-component aerosol mixtures. For simultaneous concentration measurements, the various instruments considered were operated under steady state conditions in a linear flow system. A series of at least 10 single concentration measurements was performed by each individual instrument at each set of test aerosol parameters. The average of the concentration data measured by the various instruments was defined as a common reference. The number concentrations obtained from the various instruments typically agreed within a factor of about two over the entire concentration range considered. The agreement of the measured concentrations is notable considering the various different measurement principles applied in this study, and particularly in view of the broad range of measurement instruments used. Significant deviations and nonlinear response were observed only in a few cases and are possibly related to calibration errors. For certain conditions, a dependence of aerosol counter response on particle composition has been found. The scatter of the number concentrations obtained from each individual instrument during measurements with constant test aerosol typically did not exceed 20% to 25%. At concentrations below 10^3 cm^{-3}, however, several of the instruments, including electrostatic particle measurement systems, tend to show increased experimental scatter. © 2002 Elsevier Science B.V. All rights reserved.

1. Introduction

Formation and dynamics of atmospheric aerosols have received considerable attention in recent years. Particularly the possible influence of atmospheric aerosols on the evolution of the global climate has been discussed by several authors (see, e.g. Blanchet, 1989; Koepke, 1992; Charlson and Wigley, 1994). At this point, unfortunately, the global database on atmospheric aerosols appears to be still quite incomplete and the present measuring network needs to be improved (Deepak and Vali, 1991).

One of the most frequently determined parameters used for the characterization of atmospheric aerosols is number concentration. According to recent international recommendations (Global Atmosphere Watch, 1999), aerosol number concentration is among the aerosol parameters to be measured at global stations. Determination of aerosol number concentration can be performed by means of various methods based on a number of different measuring principles.

In recent years, calibrations and intercomparisons of several concentration measuring instruments were reported (Bartz et al., 1985; Dreiling et al., 1986; Wiedensohler et al., 1997) mainly considering flow diffusion condensation particle counters. In order to contribute to a comprehensive standardization of aerosol number concentration measurement systems, we compared various types of condensation particle counters as well as several other instruments based on different measurement principles during an international workshop, at the Institute for Experimental Physics of the University of Vienna, Austria, which was coordinated within the Committee on Nucleation and Atmospheric

Aerosols, ICCP-IUGG. A broad range of different instruments was considered, including Pollak counters in use already for several decades, presently available commercial particle counters, as well as laboratory prototypes. Particularly we used various types of adiabatic expansion condensation particle counters, flow diffusion condensation particle counters, turbulent mixing condensation particle counters, laser optical particle counters, electrostatic aerosol spectrometers and a Faraday Cup Electrometer.

In order to allow quantitative intercomparisons of aerosol number concentration measurements under well-defined reproducible conditions, test aerosols with specified

Table 1
Aerosol measurement instruments used in the present study

Symbol	Acronym	Principle	Description	Institution
#	PCP	EXP	Pollak Counter	University of Missouri, Rolla, MO, USA
+	PCO	EXP	Pollak Counter	University of Galway, Galway, Ireland
×	PCG	EXP	Pollak Counter	Institute of Natural Resources and Environment, Aspendale, Victoria, Australia
■	GIV CNC 440	EXP	Pollak Type Counter Netzsch CNC 440	Netzsch Gerätebau, Germany GIV, Breuberg, Germany
●	SANC 1	EXP	Size Analyzing Nuclei Counter	Institut für Experimentalphysik, Universität Wien
▲	MULSS	EXP	Multiple Light Scattering System	Institut für Experimentalphysik, Universität Wien
△	TSI 3010	DIFF	TSI Condensation Particle Counter 3010	TSI, St. Paul, MN, USA
○	TSI 3022A	DIFF	TSI Condensation Particle Counter 3022A	TSI, St. Paul, MN, USA
▽	TSI 3025A	DIFF	TSI Ultrafine Cond. Particle Counter 3025A	TSI, St. Paul, MN, USA
◇	ADB	MIX	Automated Diffusion Battery	Institute of Chemical Kinetics and Combustion, Novosibirsk, Russia
□	DAS	MIX	Diffusion Aerosol Spectrometer	Karpov Institute of Physical Chemistry, Moscow, Russia
Y	TM CNC	MIX	Turbulent Mixing Type Cond. Nuclei Counter	Institute of Chemical Kinetics and Combustion, Novosibirsk, Russia
*	PMS LAS-X	OPT	Laser Optical Particle Counter LAS-X	Particle Measuring Systems, Boulder, CO, USA
◆	EAS 4	STAT	Electric Aerosol Spectrometer	Tartu University, Tartu, Estonia
$	HAUKE EMS-07	STAT	Electrical Mobility Spectrometer EMS-07	Hauke, Gmunden, Austria
%	HAUKE EMS-08	STAT	Electrical Mobility Spectrometer EMS-08	Hauke, Gmunden, Austria
	FCE	STAT	Faraday Cup Electrometer FCE-07	Institut für Experimentalphysik, Universität Wien

Measuring principles of the instruments: EXP: adiabatic expansion condensation nuclei counter (working fluid/water); DIFF: flow diffusion condensation nuclei counter (working fluid/*n*-butanol); MIX: turbulent mixing condensation nuclei counter (working fluid/dibutylphthalate); OPT: optical particle counter; STAT: electrostatic particle measurement system.

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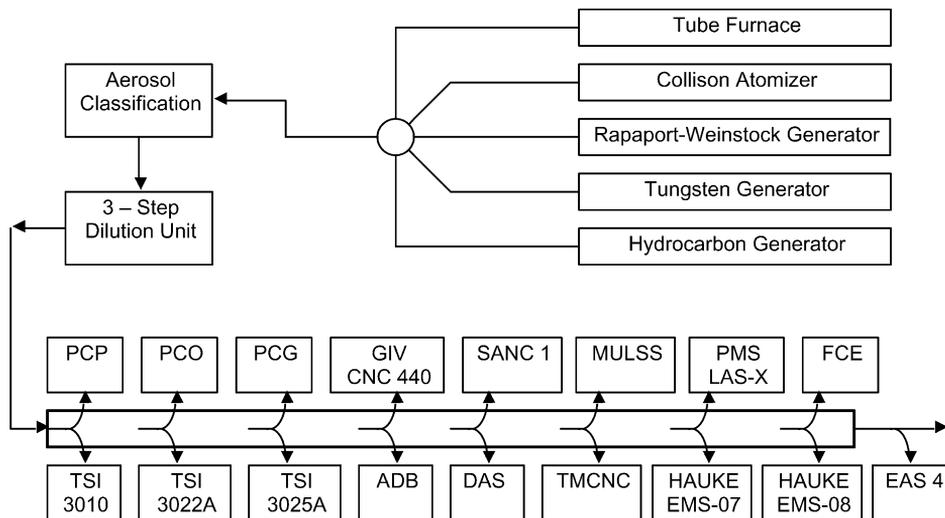
Fig. 1. Schematic diagram of the experimental arrangement showing the systems for aerosol generation, classification and dilution as well as the linear aerosol flow system and the various aerosol measurement instruments considered. The instruments used are listed in Table 1.

properties and known particle compositions have been used. We considered nearly monodispersed aerosols with mean particle diameters covering a comparatively wide range including ultrafine aerosols. Furthermore, mixtures of two monodispersed aerosol fractions

Table 2
Test aerosols used in the present study

Particle compound	Geometric mean particle diameter (nm)	Geometric standard deviation	Aerosol generation principle	Concentration (1)/ concentration (2)
One-component systems				
DEHS[a]	50–58	1.2	Collison atomizer + classifier	
DEHS[a]	130	1.3	Collison atomizer + LaMer generator	
DEHS[a]	520	1.2	Collison atomizer + LaMer generator	
NaCl	4	1.3	tube furnace + classifier	
NaCl	20	1.2–1.5	tube furnace	
Ag	7	1.3	tube furnace	
Hydrocarbons	30	1.5	photochemical reaction generator	
Tungsten oxide	11	1.4	heated wire generator	
Two-component systems				
DEHS[a]	54	1.20	Collison atomizer + classifier	1
NaCl	18	1.05	tube furnace	
Ag	7	1.04	tube furnace + classifier	0.3
NaCl	18	1.04	tube furnace + classifier	

[a] Di (2-ethyl hexyl) sebacate.

were studied. Test aerosols with various compositions including hygroscopic as well as nonhygroscopic particles were used in order to investigate a possible dependence of the response of aerosol particle counters on the composition and physico-chemical properties of the aerosols.

In the next chapter, the measuring principles of the various aerosol concentration measuring instruments considered in the present study are briefly reviewed. The following chapter contains a description of the general experimental arrangement used in the inter-comparison experiments including the aerosol generation systems used. Subsequently, the measurement results are presented in detail and few conclusions are drawn.

2. Instrumentation

In the present intercomparison study, we considered 16 different instruments for the measurement of aerosol number concentration, which are based on five different measurement principles. In these instruments, measurement of aerosol number concentration is
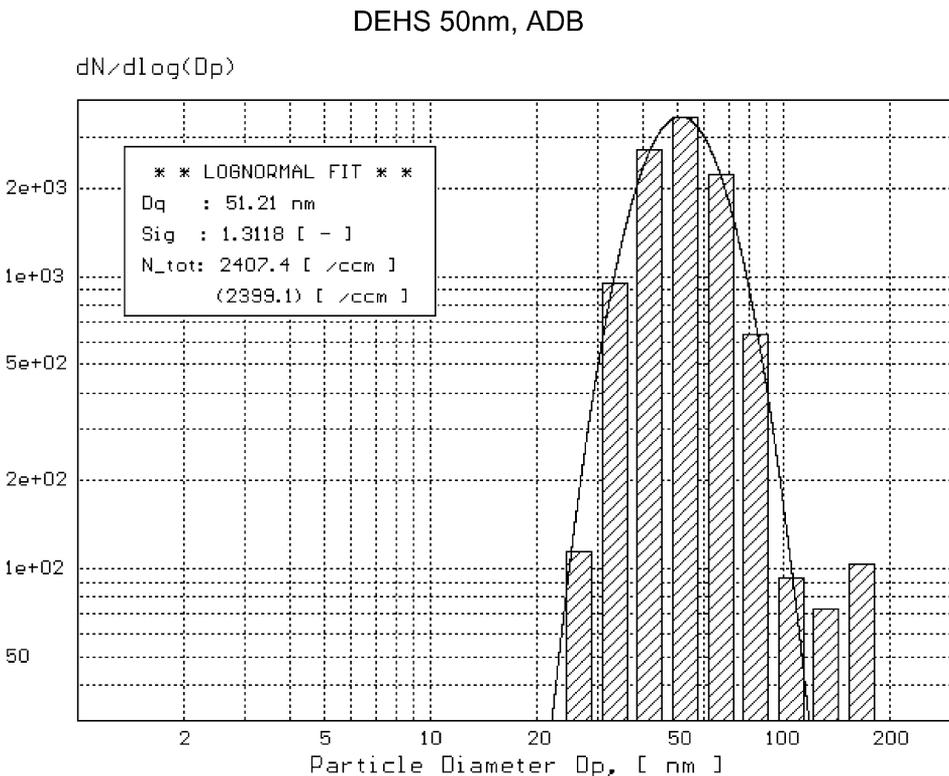

Fig. 2. Size distribution of DEHS aerosol, geometric mean particle diameter around 50 nm, as measured by means of the Automated Diffusion Battery (ADB).
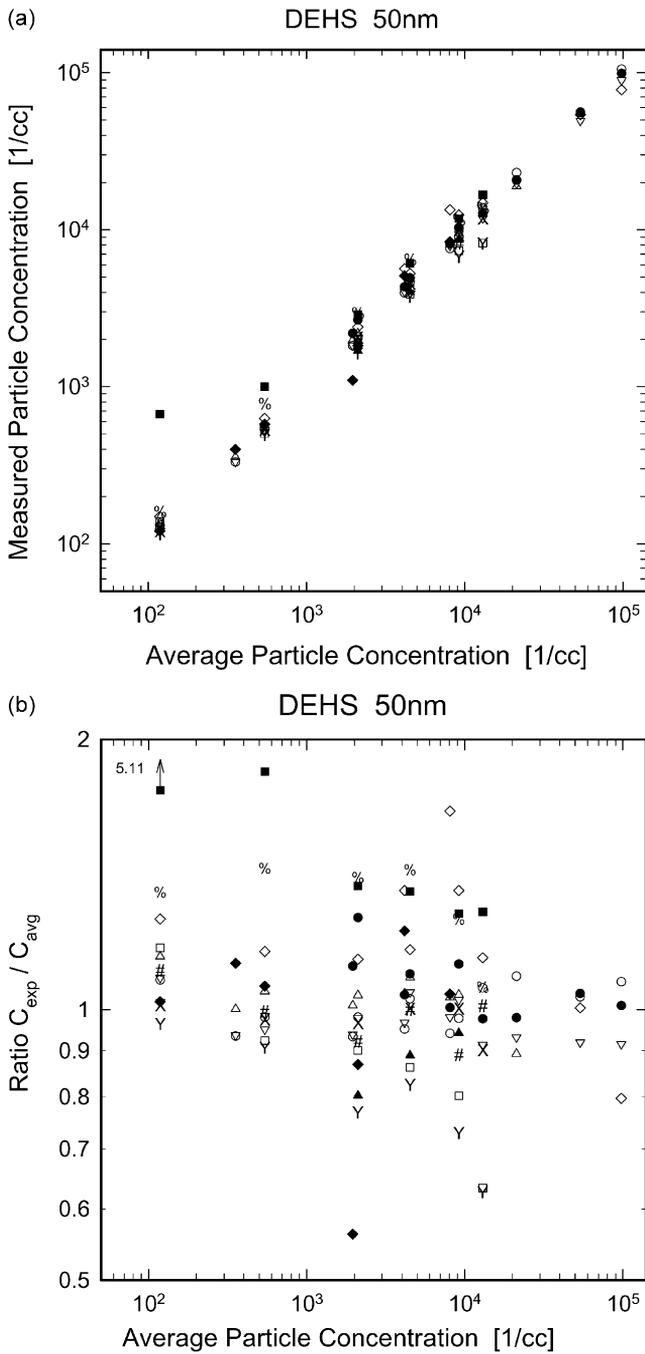
(a)

DEHS 50nm

(b)

DEHS 50nm

Fig. 3. (a–c) Results of the concentration intercomparisons for the DEHS aerosols, geometric mean particle diameter 50–58 nm.
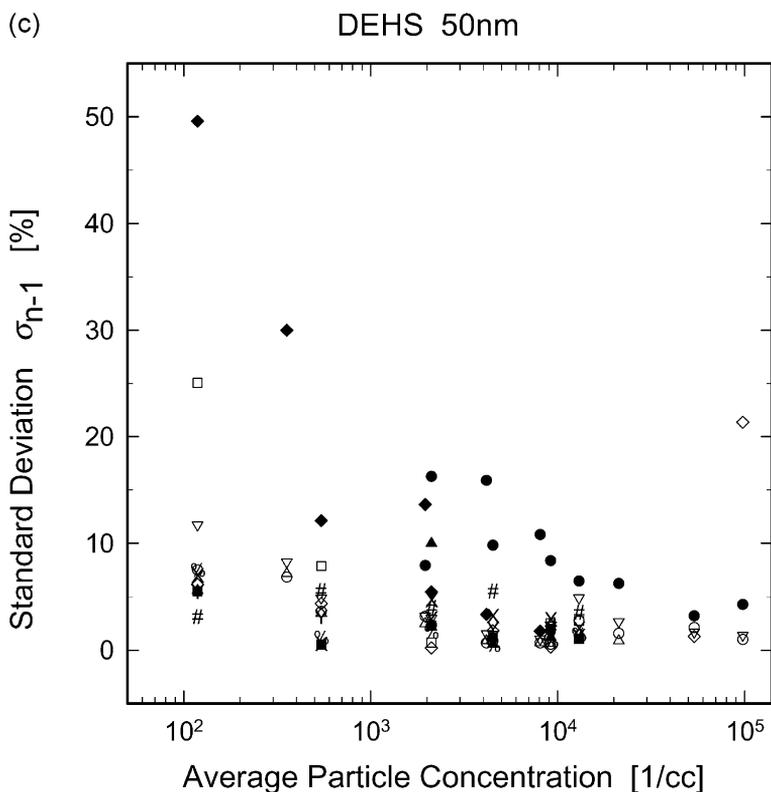
(c)

DEHS 50nm

Fig. 3 (*continued*).

partly performed by single particle counting, partly the concentration is determined from the measurement of an integral parameter. Various of these devices can be considered as absolute instruments, which are independent of empirical calibration relative to external reference standards, few instruments, however, require empirical calibrations. While all of the measuring systems considered provided data on particle number concentration, some of the instruments furthermore allowed the determination of aerosol size distribution. The latter instruments were used for the characterization of the various test aerosols used.

The devices used and their measuring principles are listed in Table 1. The Pollak Counters (PCP, PCO, PCG) and the Pollak Type Counter (GIV CNC 440) are based on adiabatic expansion of initially water-saturated aerosols causing vapor supersaturation and drop growth. The particle number concentration is determined from the measurement of light extinction using an empirical calibration. A more detailed description of these instruments is given by Gras et al. (2002). The Size Analyzing Nuclei Counter (SANC 1) and the Multiple Light Scattering System (MULSS) are based on adiabatic expansion as well, however, the number concentration of the growing droplets is determined from simultaneous light scattering and extinction measurements and quantitative comparison to
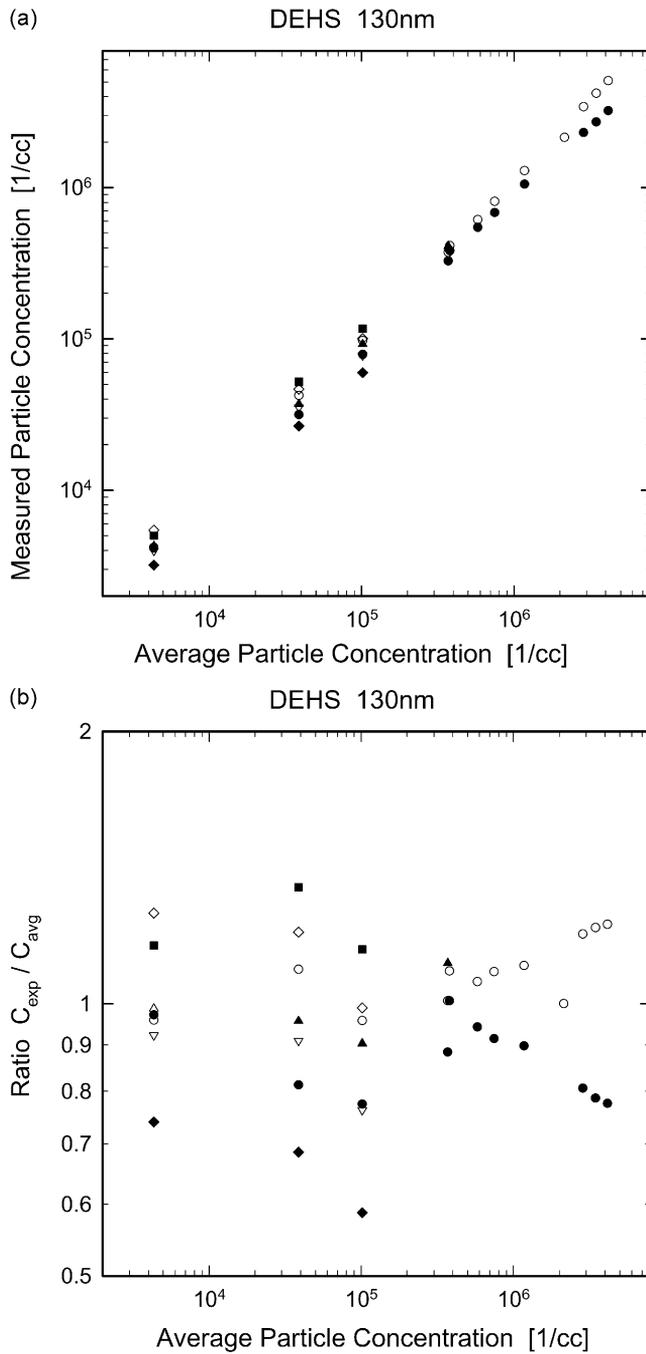
(a)

DEHS 130nm

(b)

DEHS 130nm

Fig. 4. (a–c) Results of the concentration intercomparisons for the DEHS aerosols, geometric mean particle diameter 130 nm.
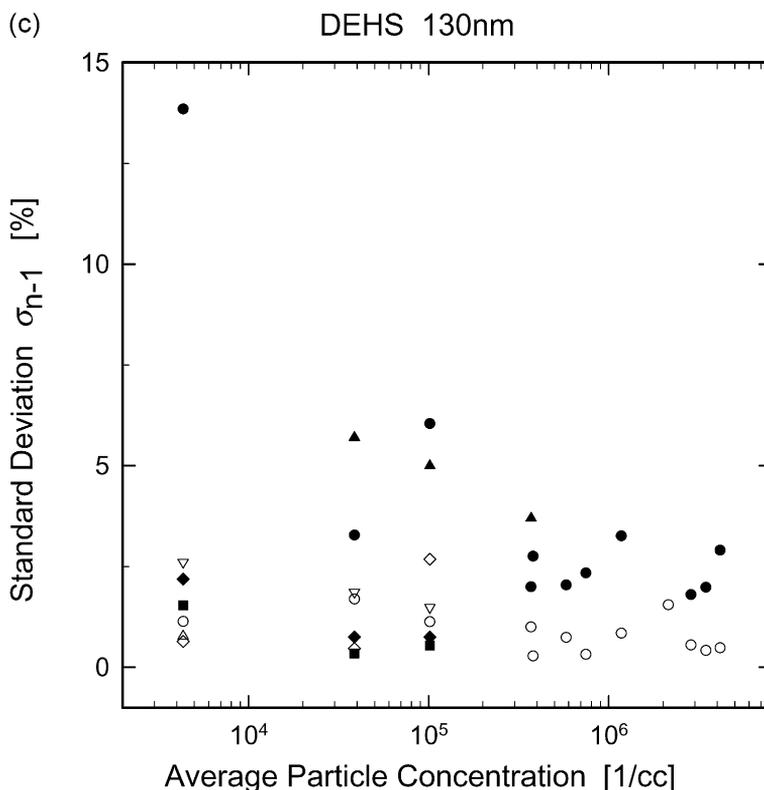
(c)

DEHS 130nm

Fig. 4 (*continued*).

Mie scattering theory. This method allows the absolute number concentration measurement without calibration relative to an external standard (see Szymanski and Wagner, 1990; Szymanski, 2002).

The group of instruments TSI 3010, TSI 3022A and TSI 3025A is based on nonisothermal flow diffusion. Aerosols initially saturated with *n*-Butanol vapor are passed in laminar flow through a cooled cylindrical tube and heat conduction lead to vapor supersaturation and drop growth. Drop concentration is determined either by optical single particle counting or, at high concentration, by an integral optical method requiring empirical calibration. A detailed description is given by Sem (2002).

Both the Automated Diffusion Battery (ADB) and the Diffusion Aerosol Spectrometer (DAS) are devices providing total particle number concentrations as well as aerosol size distributions. For detection, the aerosol particles are magnified by the condensation of supersaturated low volatility vapors (dibutylphthalate, dioctylsebacate) obtained by mixing of two gas flows at different temperatures. The aerosol number concentration is measured by optical single particle counting. More details are given by Julanov et al. (2002).

In the Turbulent Mixing Type Condensation Nuclei Counter (TM CNC) (Mavliev and Wang, 2000; Mavliev, 2002) supersaturated dibutylphthalate vapor is obtained by
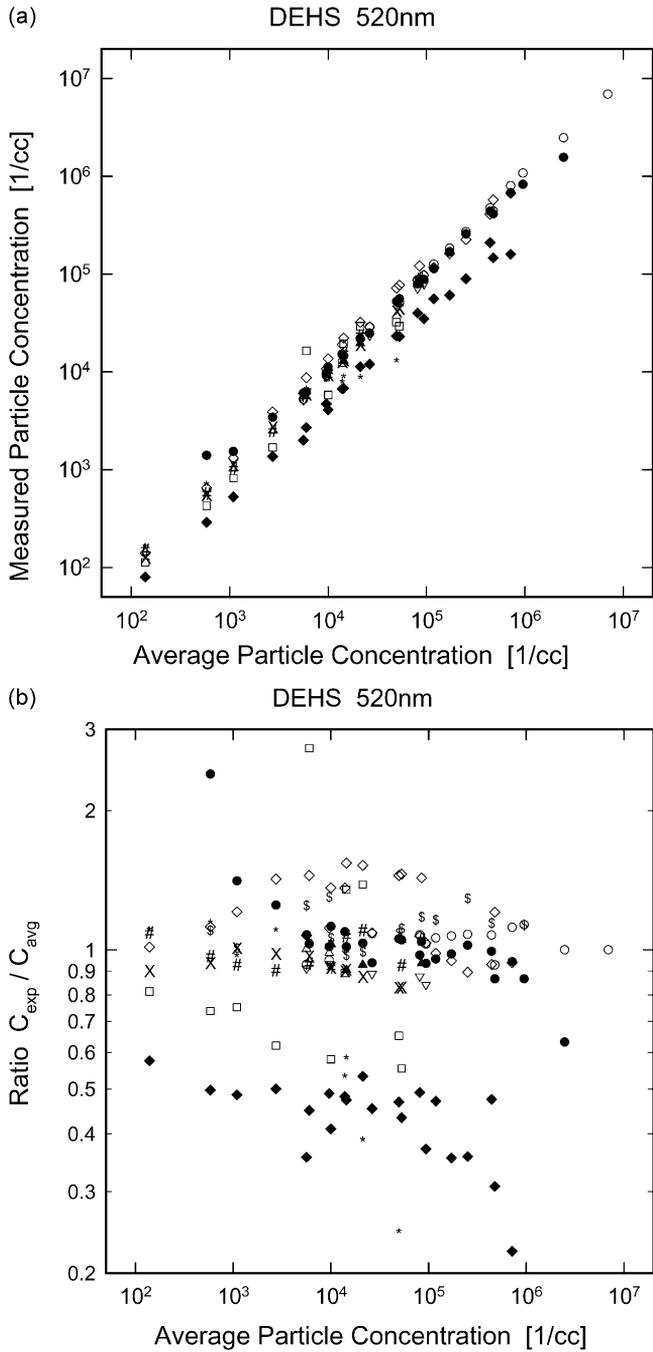
(a)

(b)

Fig. 5. (a–c) Results of the concentration intercomparisons for the DEHS aerosols, geometric mean particle diameter 520 nm.
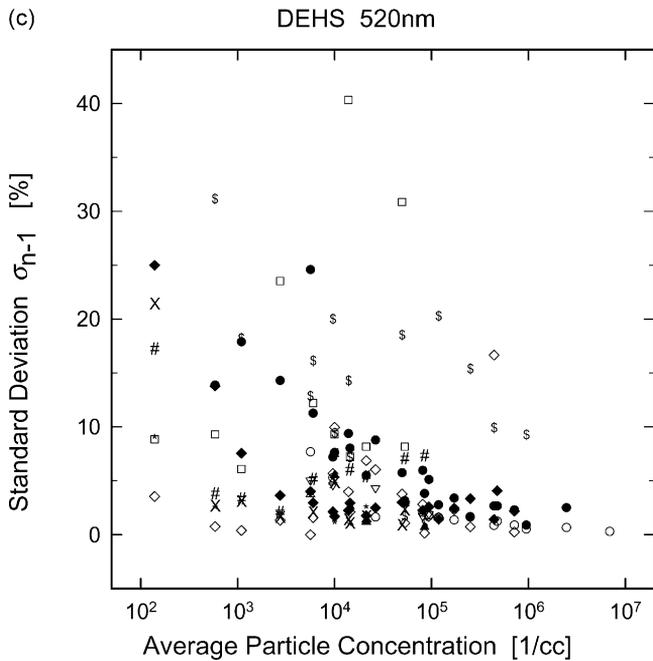
Fig. 5 (*continued*).

turbulent mixing of a hot vapor-saturated particle-free flow with a cold aerosol flow. The aerosol particles grow to visible sizes and the number concentration is measured by optical single particle counting.

The laser optical particle counter (PMS LAS-X) allows the determination of particle size distributions as well as the total number concentrations. The range of particle sizes is restricted to visible sizes, beyond a certain number concentration counting errors due to particle coincidences occur. For more details see Szymanski (2002).

The Electric Aerosol Spectrometer (EAS 4), as described in some detail by Tammet et al. (2002) and the Electrical Mobility Spectrometers (HAUKE EMS-07 and HAUKE EMS-08) (see Reischl et al., 1997) allow the determination of particle number size distributions. To this end, the aerosol particles, which may either be in bipolar electrical charge equilibrium or carry unipolar charges are electrostatically classified. The concentrations of the thereby obtained series of aerosol mobility fractions are determined from the measurements of electrical currents occurring in Faraday Cup Electrometers. The number size distributions are calculated accounting for the actual charging probabilities and the total particle number concentration is obtained by integration over the entire size range.

The Faraday Cup Electrometer (FCE) is applicable only for the measurement of the concentration of particles carrying unipolar electrical charges. In the present study, the FCE has only been used, when the test aerosols considered were obtained by electrostatical classification of polydispersed primary aerosols. For sufficiently small particle diameters, the influence of multiple charges on the particles is negligible.

3. Experimental procedure

For intercomparison of aerosol measurement instruments, it is important that well-defined and stable test aerosols are used and that each of the instruments considered is sampling aerosol with equal properties. In order to meet these requirements, either an aerosol plenum volume or an aerosol flow system can be used. An aerosol plenum volume is applicable in the case of sufficiently stable aerosols and for a limited number of instruments to be connected simultaneously. In the present study, aerosols with number concentrations up to 10^7 cm^{-3} and with particle diameters down to 4 nm were considered. These aerosols are generally undergoing quite rapid dynamic changes causing significant instabilities in an aerosol plenum volume. Furthermore, due to the comparatively large number of aerosol measurement instruments considered in this study, a simultaneous connection of all instruments to one aerosol plenum volume is difficult.

In order to minimize the above-mentioned difficulties, a linear aerosol flow system has been used in the present study. This arrangement has the advantage that steady state conditions can be achieved even for the case of comparatively unstable test aerosols. Furthermore, for an appropriate choice of the length of the aerosol flow channel, a sufficient number of instruments can be connected simultaneously. Of course the stability of the test aerosols obtained in a flow system critically depends on the stability of the aerosol generation, classification and dilution systems. Accordingly, the aerosol generators as well as the classification and dilution systems were operated under carefully controlled conditions, and the properties of the test aerosols were continuously monitored during each measurement series.

A schematic diagram of the experimental arrangement is shown in Fig. 1. Five different generators were available for the generation of various test aerosols with particle diameters ranging from about 4 to 500 nm. Measurement series were performed with one-component test aerosols generated by one of the generators as well as with two-component systems obtained from two simultaneously operated generators. In the following, the operating principles of the generators used in the present study are briefly outlined.

Generation of di(2-ethyl hexyl) sebacate (DEHS) particles was performed by atomizing dilute solutions of DEHS in iso-propanol by means of a Collison atomizer (TSI 3076) and subsequent drying in a diffusion dryer. Monodispersed aerosol fractions with particle diameters around 50 nm were obtained using an electrostatic aerosol classifier (TSI 3071). Nearly monodispersed DEHS aerosols with larger particle diameters up to about 500 nm were generated by evaporation and subsequent condensation of the polydispersed primary aerosols in a LaMer-type (Rapaport–Weinstock) generator.

NaCl and Ag aerosols were generated by evaporation in a tube furnace and subsequent condensation (Scheibel and Porstendörfer, 1983). In some cases, monodispersed NaCl aerosol fractions were obtained by electrostatic aerosol classification.

Fairly monodispersed and stable aerosols consisting of hydrocarbons were generated by means of photochemical reactions in benzene halides and subsequent nucleation. The resulting particles consist of polyaromatic oxygen- and halogen-containing molecules.

Finally, for the generation of tungsten oxide particles, a tungsten wire was heated to a temperature around 1000 °C, where tungsten is stable, while tungsten oxide will eva-

porate. Oxidation at the heated tungsten surface leads to the evaporation of tungsten oxide and subsequent nucleation results in tungsten oxide particles.

The aerosol particles generated were nearly in charge equilibrium. However, as mentioned above, for some measurement series electrostatic classification of the generated aerosols was performed. In these cases, a nearly monodispersed fraction of unipolarly charged particles was obtained.

The aerosol was then diluted in a three-stage dilution unit in order to achieve various constant aerosol number concentrations in a range from about 10^2 to 10^7 cm^{-3}. Subsequently, the aerosol was passed through a cylindrical aerosol flow channel with an inner diameter of 50 mm and a length of about 2.5 m. In order to minimize aerosol losses in the channel, comparatively high aerosol flowrates ranging from 60 to 90 l/min were chosen. Aerosol concentration measurements by means of the two of the instruments used in this study (TSI 3025A and SANC 1, see Table 1) were routinely performed at inlet and exit of the aerosol flow channel indicating that particle losses in the aerosol flow channel were negligible.

All aerosol measurement instruments considered in a particular measuring series were *simultaneously* connected to the aerosol channel. Test aerosol not sampled by the
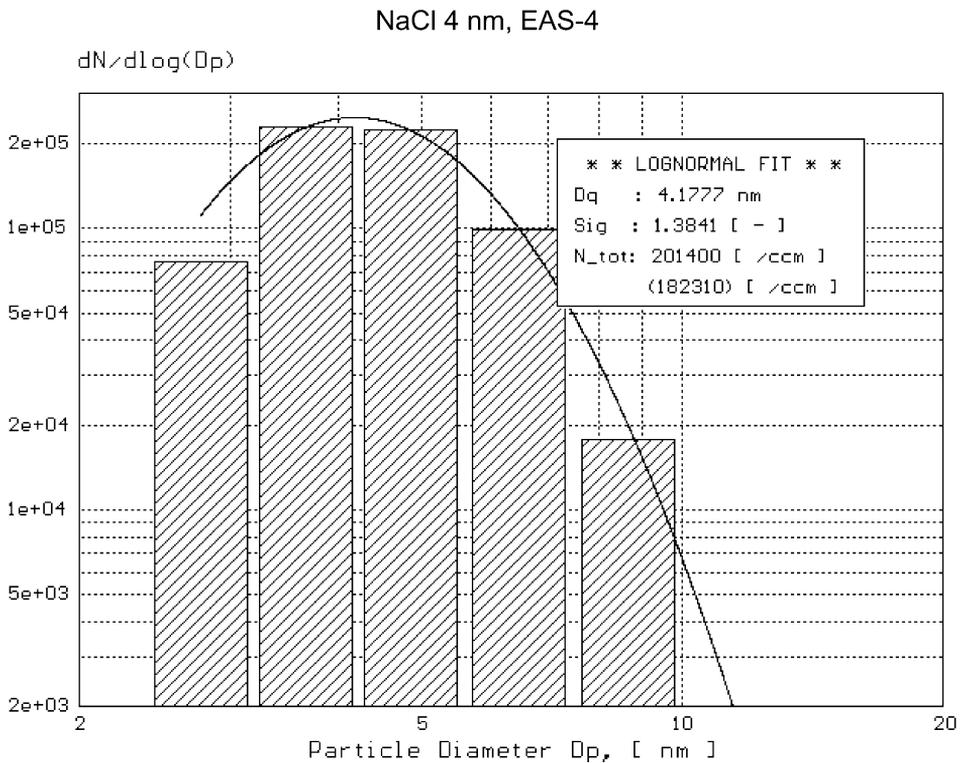

Fig. 6. Size distribution of NaCl aerosol, geometric mean particle diameter 4 nm, as measured by means of the Electric Aerosol Spectrometer (EAS 4).
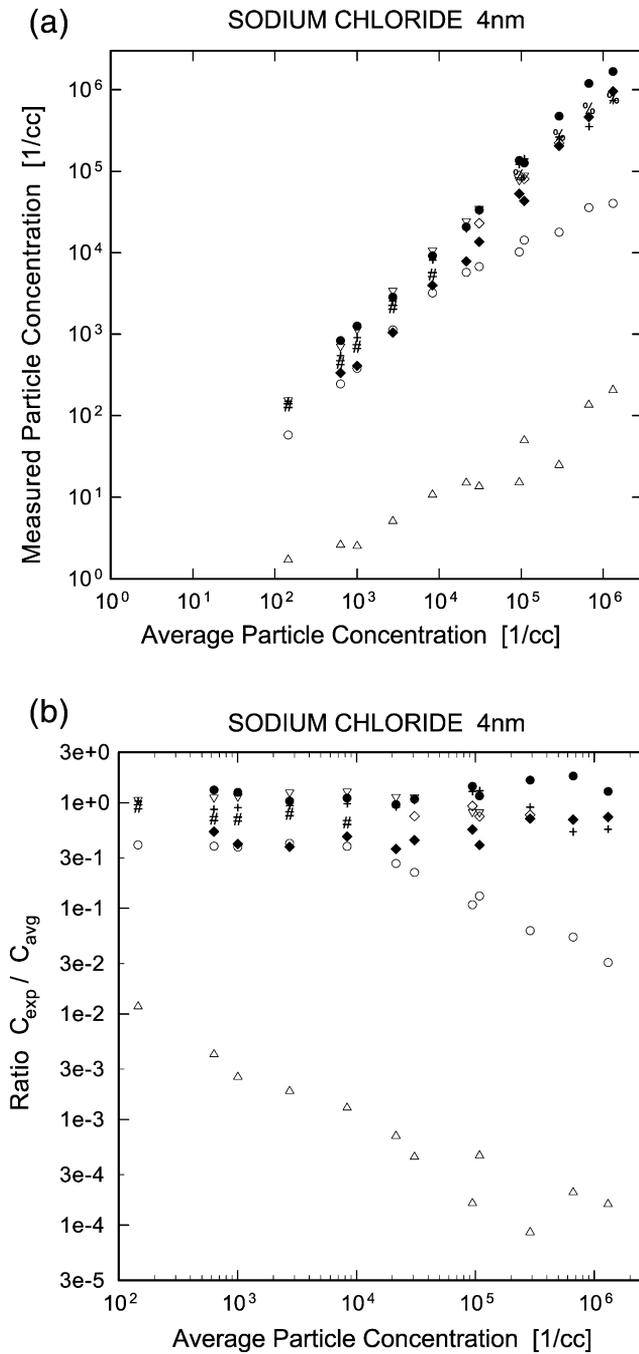
Fig. 7. (a–c) Results of the concentration intercomparisons for the NaCl aerosols, geometric mean particle diameter 4 nm.
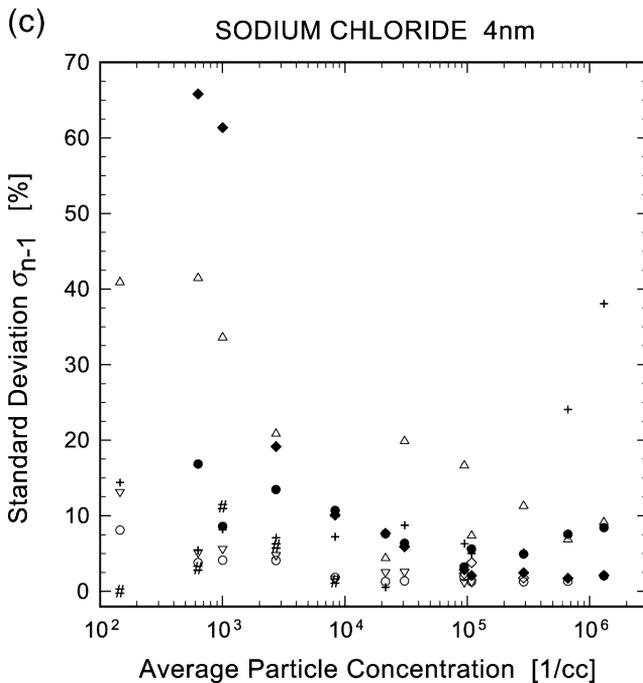
Fig. 7 (*continued*).

instruments connected to the channel was vented to an exhaust. Except for the Electric Aerosol Spectrometer (EAS 4) (see Table 1), all instruments were connected via aerosol neutralizers. Accordingly, the test aerosol sampled by the various instruments was in charge equilibrium. In this connection, it should be recalled that for measuring series with electrostatically classified test aerosols the aerosol particles passing through the flow channel were unipolarly charged. Particle losses in the connecting tubes to the individual instruments were accounted for using the expression by Gormley and Kennedy (1949) and considering the corresponding sample flowrates.

During each measuring series at least 10 single measurements were simultaneously performed by each of the instruments considered, while the parameters of the test aerosol were kept constant. The particle number concentration of the test aerosol was found to vary typically within a few percent during one measuring series. In the rare cases, when a significant drift of the aerosol number concentration was observed during one measurement series, the corresponding measuring results were discarded.

Measurement series were performed for a number of different test aerosols. During *concentration intercomparisons*, the particle number concentration was varied by selecting different dilution ratios at the three-stage dilution unit while keeping the average particle diameter constant. On the other hand, *detection efficiency intercomparisons* were performed, where the average particle diameter was varied while keeping the particle number concentration, as monitored by the Faraday Cup Electrometer (FCE) (see Table 1) at a

Fig. 8. (a–c) Results of the concentration intercomparisons for the NaCl aerosols, geometric mean particle diameter 20 nm.
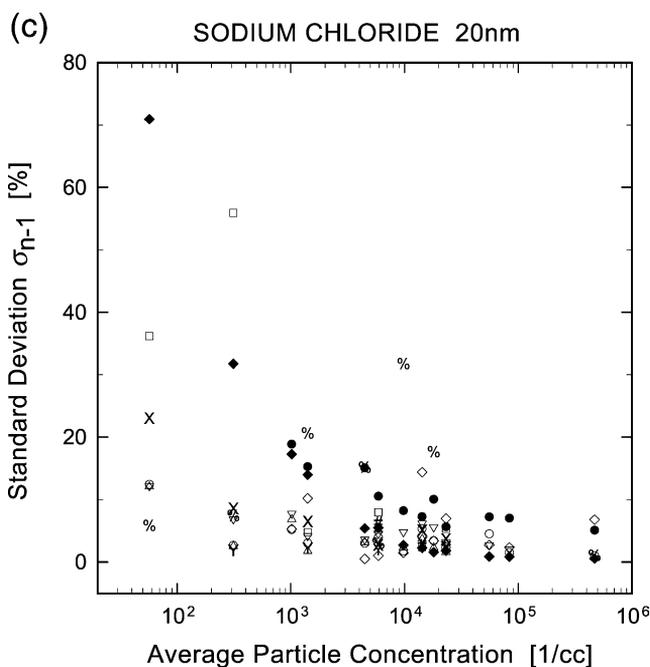
Fig. 8 (*continued*).

constant value. In the following, the results of the concentration intercomparisons will be presented in detail. The results of the detection efficiency intercomparisons are included in Ankilov et al. (2002).

4. Results and discussion

Simultaneous measurements of aerosol particle number concentration were performed by all instruments listed in Table 1 except for the Faraday Cup Electrometer (FCE), which is applicable only to unipolarly charged aerosol particles. As mentioned earlier, the test aerosols sampled by the various instruments were nearly in charge equilibrium except for the Electric Aerosol Spectrometer (EAS 4), which was sampling unipolarly charged aerosol during measuring series with electrostatically classified test aerosols. The test aerosols were continuously monitored during each measurement series and particle number size distributions were routinely measured by means of the Electrical Mobility Spectrometers (HAUKE EMS-07 and HAUKE EMS-08), the Automated Diffusion Battery (ADB), and the Electric Aerosol Spectrometer (EAS 4).

The instruments considered are based on various different measurement principles. Some of the instruments had to be calibrated against external reference standards previous to the intercomparison measurements performed in this study, other instruments are

independent of empirical calibration. Accordingly, it does not appear to be useful to identify one particular instrument as a common reference. Comparison of the various concentration measurements was therefore performed with respect to the *average* of the concentration data obtained during one measuring series. Usually, all concentration measurements were included into the concentration average. In cases, however, were an instrument was obviously operated outside its measuring range and clearly yielded erroneous results, the corresponding concentration data were not included into the concentration average. In this way, it was always possible to determine unique average concentrations for each measurement series. These average concentrations were found to provide a useful reference, particularly for measuring series, where a comparatively large number of instruments were operated simultaneously.

In the following, the measured particle number concentration C_{exp}, the ratio of measured and average particle number concentration C_{exp}/C_{avg}, and the standard deviation σ_{n-1} of the data obtained from each individual instrument, are presented as functions of the average particle number concentration C_{avg}. For the various test aerosols considered, we show particle number size distributions, as measured by means of the Elec-

Fig. 9. Size distribution of Ag aerosol, geometric mean particle diameter 7 nm, as measured by means of the Automated Diffusion Battery (ADB).
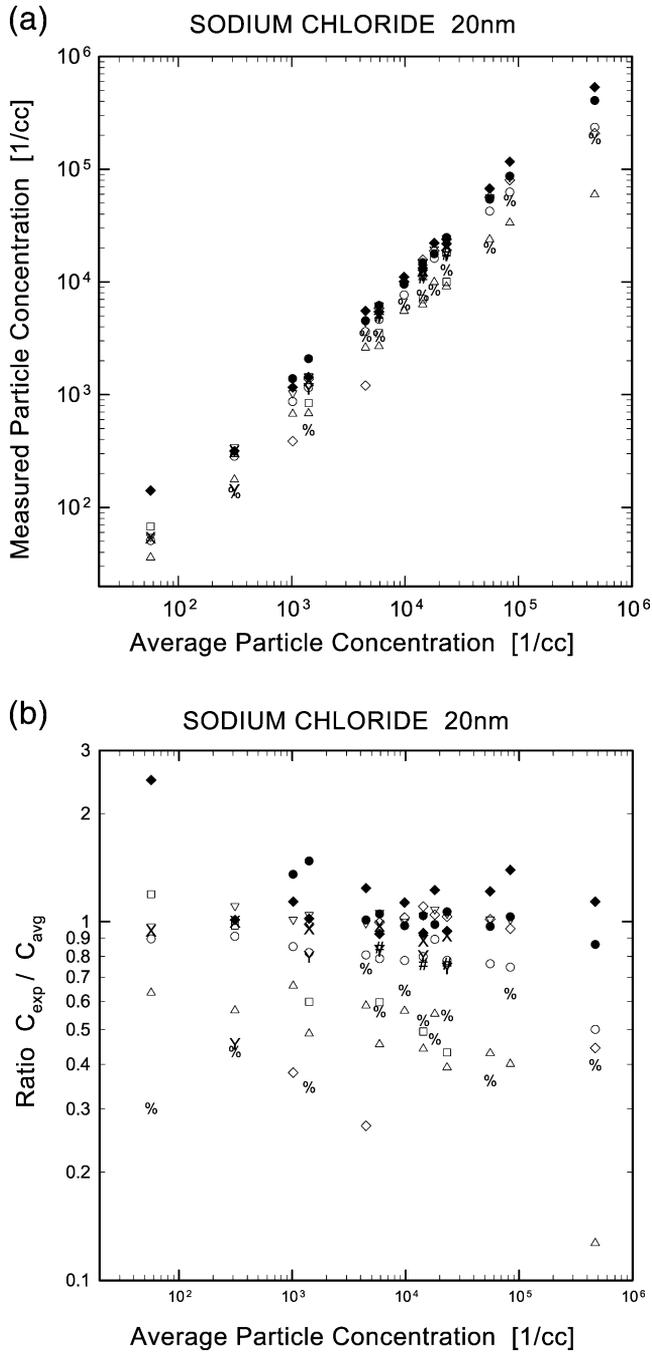
Fig. 10. (a–c) Results of the concentration intercomparisons for the Ag aerosols, geometric mean particle diameter 7 nm.
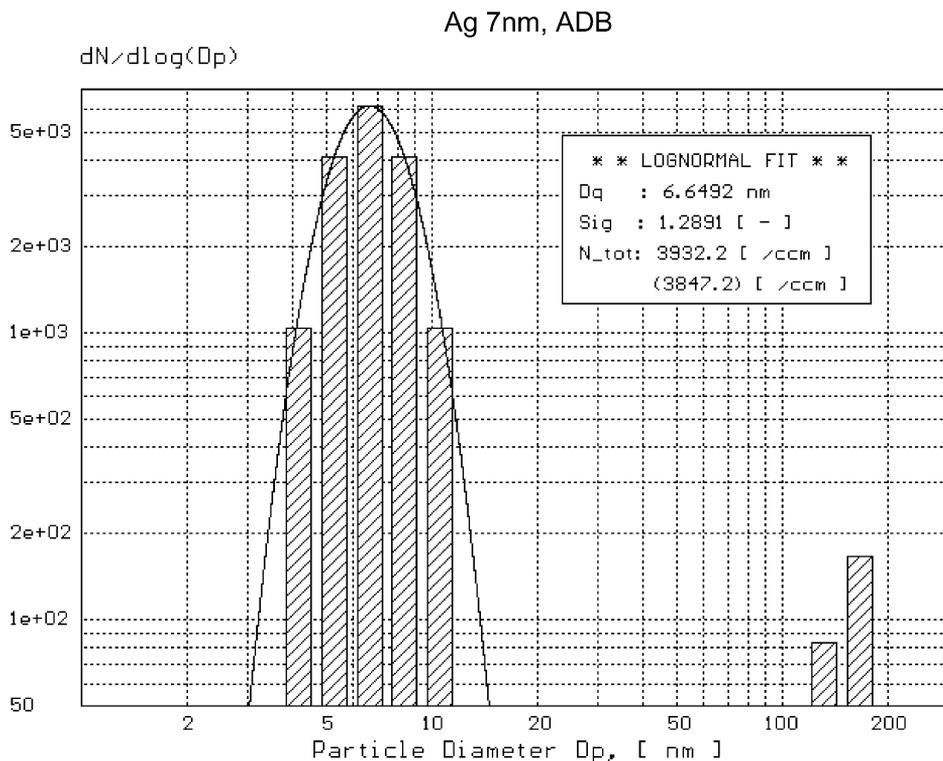
Fig. 10 (*continued*).

trical Mobility Spectrometer (HAUKE EMS-08), the Automated Diffusion Battery (ADB), and the Electric Aerosol Spectrometer (EAS 4). Lognormal fits to the experimental size distributions were performed and the test aerosols are characterized by their geometric mean particle diameter $\bar{d}_p$ and geometric standard deviation σ_g. Measuring series were performed for various test aerosols. One-component as well as two-component aerosols were considered. The test aerosols used and their main properties are listed in Table 2.

Concentration intercomparisons were performed for DEHS aerosols at three different mean particle sizes. A particle number size distribution for geometric mean particle diameter around 50 nm is shown in Fig. 2. The results of the concentration intercomparisons for the DEHS aerosols are shown in Figs. 3, 4 and 5. During these measurements, comparatively large particle diameters between 50 and 520 nm were considered and a wide concentration range from about 10^2 to 10^7 cm^{-3} was covered. Each of the various instruments considered exhibited a quite similar behaviour for the three different particle diameters selected. As can be seen from Fig. 3a, b, the concentrations obtained from the Pollak Type Counter Netzsch CNC 440 (GIV CNC 440) are significantly too high for particle concentrations below 10^3 cm^{-3} indicating a calibration error of this instrument. Fig. 3c shows that the data obtained from the Electric Aerosol Spectrometer (EAS 4) exhibit quite large experimental scatter for concentrations below about 10^3 cm^{-3}, which might be connected to electrometer noise. In Fig. 4b, it can be seen that for

particle concentrations above 5×10^5 cm^{-3} the Condensation Particle Counter (TSI 3022A) and the Size Analyzing Nuclei Counter (SANC I) yield deviating results. The TSI 3022A concentration ratios are observed to increase while at the same time the SANC I concentration ratios decrease with increasing average concentration. In this connection, it should be noted that for concentrations above 5×10^5 cm^{-3} only data from these two instruments were available and thus these data were plotted with respect to the averages *from the two instruments only.* In an independent study (Filipovicova, 1996), it has been shown that the SANC I yields fairly accurate number concentration values up to several 10^7 cm^{-3}. Accordingly, the results shown in Fig. 4b indicate that the calibration of the TSI 3022A might be inaccurate for concentrations above 5×10^5 cm^{-3}. A similar behaviour is observed in Fig. 5b. As seen from Figs. 4c and 5b, the SANC I data are fairly scattered for concentrations below 10^4 cm^{-3}, at a concentration below 10^3 cm^{-3} the SANC I yielded a considerably high concentration value. Furthermore, Figs. 4b and 5b indicate that for increasing concentrations the Electric Aerosol Spectrometer (EAS 4) yields increasingly too low concentration values, which might be
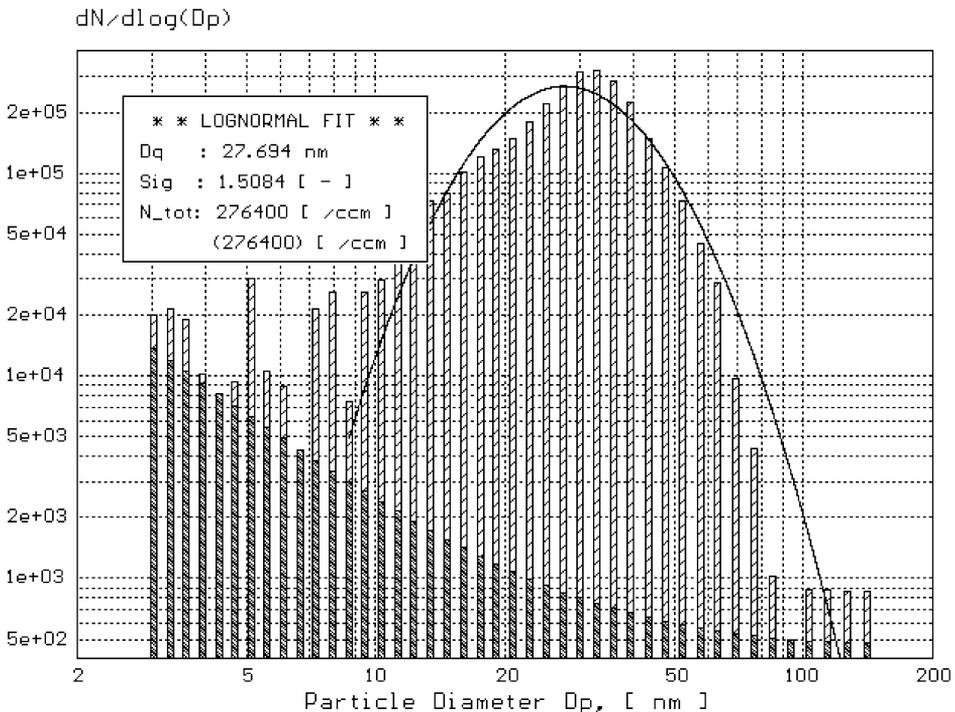
Fig. 11. Size distribution of the hydrocarbon aerosol, geometric mean particle diameter 30 nm, as measured by means of the Electrical Mobility Spectrometer (HAUKE EMS-08).
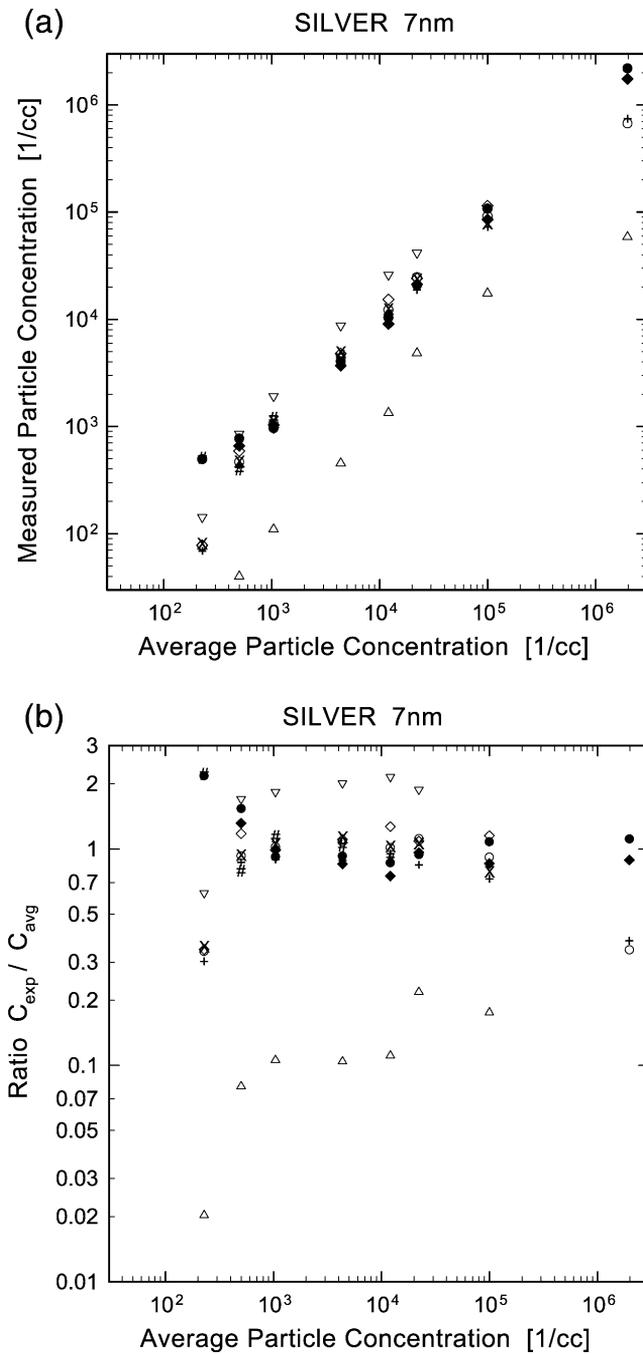
(a)

(b)
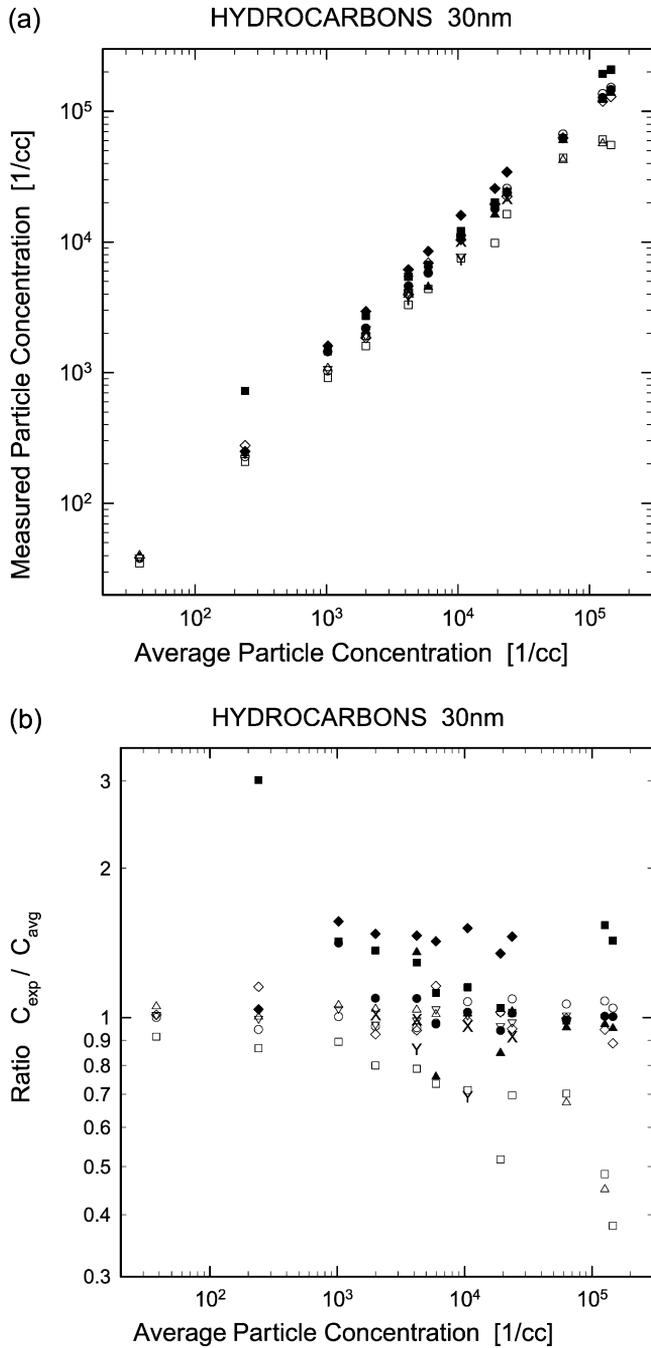
Fig. 12. (a–c) Results of the concentration intercomparisons for the hydrocarbon aerosols, geometric mean particle diameter 30 nm.
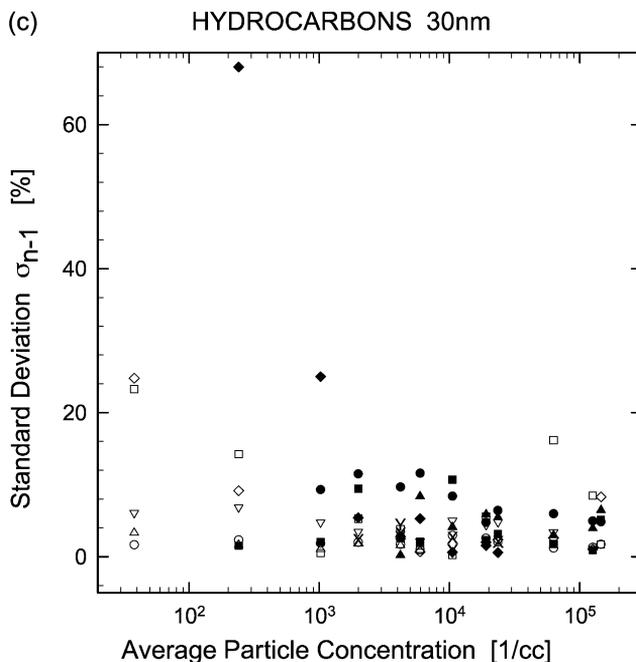
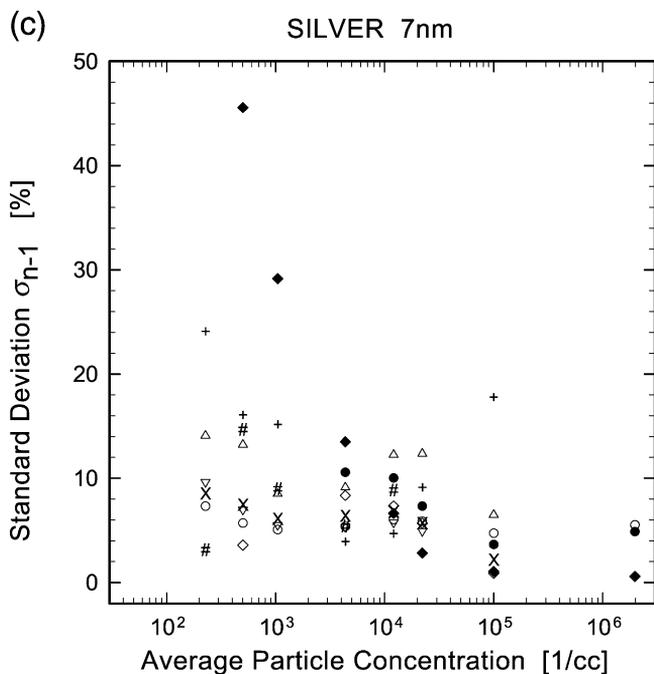Fig. 12 (*continued*).

connected with a calibration problem. This behaviour seems to be mainly pronounced at larger particle sizes, as can be expected for dielectric particles (Tammet et al., 2002). From Fig. 5b, it is also seen that for concentrations close to 10^5 cm^{-3} the laser optical particle counter LAS-X (PMS LAS-X) is apparently operated outside its concentration measuring range. According to Fig. 5c, the scatter of the concentration data as obtained from the Electrical Mobility Spectrometer EMS-07 (HAUKE EMS-07) is considerably increasing with decreasing number concentrations. This behaviour is explained by electrometer noise.

Series of concentration intercomparisons was performed for the water-soluble NaCl particles. The particle size distribution for a mean particle diameter of 4 nm is shown in Fig. 6. The results of these concentration intercomparisons are presented in Figs. 7 and 8. Particle sizes in the ultrafine range and concentrations between about 10^2 and 10^6 cm^{-3} were considered. A somewhat different behaviour as compared to the DEHS measurement series was observed. As can be seen from Fig. 7a, b, for a particle diameter of 4 nm, the concentration readings of the TSI Condensation Particle Counter 3010 (TSI 3010) are substantially too low. Considerable scatter of the corresponding concentration data is observed in Fig. 7c for concentrations below about 10^3 cm^{-3}. In this connection, it is important to note, however, that the particle diameter of 4 nm is outside the specified particle size range of this instrument. As can be seen from Fig. 8b, at a particle diameter of 20 nm, the concentration readings of the TSI 3010 are typically low only by a factor of two or less, for a concentration above 10^5 cm^{-3}, however, a low value is observed. As can be

(a)

TUNGSTEN OXIDE 11nm

(b)

TUNGSTEN OXIDE 11nm

Fig. 13. (a–c) Results of the concentration intercomparisons for the tungsten oxide aerosols, geometric mean particle diameter 11 nm.

Fig. 13 (*continued*).

seen from Fig. 7a, b, at a particle diameter of 4 nm and for concentrations above 10^4 cm^{-3}, the concentration readings of the TSI Condensation Particle Counter 3022A (TSI 3022A) are increasingly low. A similar behaviour can be observed for a particle diameter of 20 nm, as seen from Fig. 8a, b. In this connection, it is important to note that the TSI 3022A exhibits the opposite trend for the DEHS particles, as seen from Fig. 4b. This behaviour may be related to the fact that the TSI Condensation Particle Counters are operated with *n*-Butanol as the working liquid. As DEHS is soluble, whereas NaCl is insoluble in *n*-Butanol, the efficiency of activation and detection might be higher for DEHS particles than for NaCl particles. The dependence of the detection efficiency on particle composition is discussed in Ankilov et al. (2002). Similar as for the DEHS measuring series, the Electric Aerosol Spectrometer (EAS 4) exhibits quite large experimental scatter for concentrations below about 10^3 cm^{-3}, as seen from Fig. 7c. Furthermore, from Fig. 7c it can be observed that the concentration data obtained from the Pollak Counter (PCO) show considerable scatter at concentrations of about 10^6 cm^{-3}.

Concentration intercomparisons were also performed for Ag aerosols with geometric mean particle diameter 7 nm, the corresponding particle size distribution is shown in Fig. 9. The results of these intercomparisons are presented in Fig. 10a, b, c. Similar as for the NaCl particles with a diameter of 4 nm (see Fig. 7 a, b), the concentration readings of the TSI Condensation Particle Counter 3010 (TSI 3010) are substantially too low, because the particle diameter of the Ag particles is outside the specified particle size range of this instrument. Furthermore, at a concentration of about 10^6 cm^{-3}, the concentration readings

(a) DEHS 54nm + NaCl 18nm

(b) DEHS 54nm + NaCl 18nm

Fig. 14. (a–c) Results of the concentration intercomparisons for mixtures of DEHS aerosols with particle diameter 54 nm and NaCl aerosols with particle diameter 18 nm.

(c)

Fig. 14 (*continued*).

of the TSI Condensation Particle Counter 3022A (TSI 3022A) and the Pollak Counter (PCO) are somewhat low.

The size distribution of the Hydrocarbon particles with 30 nm diameter is shown in Fig. 11. For the Hydrocarbon particles, it can be seen from Fig. 12b, that the Pollak Type Counter Netzsch CNC 440 (GIV CNC 440) yields a high value for a concentration around 2×10^2 cm^{-3}. The concentration values obtained from the Electric Aerosol Spectrometer (EAS 4) tend to be somewhat high. Above concentrations of 10^4 cm^{-3}, the concentration readings of the Diffusion Aerosol Spectrometer (DAS) are low. Also for the Tungsten oxide particles with 11 nm diameter, Fig. 13b shows that the Diffusion Aerosol Spectrometer (DAS) yields a low value at a concentration of around 5×10^5 cm^{-3}. Similarly, the TSI Condensation Particle Counter 3010 (TSI 3010) yields low values at concentrations above 10^5 cm^{-3}.

The results of the concentration intercomparison measurements for the two-component aerosols DEHS+NaCl and Ag+NaCl are presented in Figs. 14 and 15, respectively. The total particle number concentrations of these two-component systems were comparatively low. Accordingly, as can be seen from Figs. 14c and 15c, several instruments show somewhat increased experimental scatter. Figs. 14b and 15b show that for both systems the Pollak Type Counter Netzsch CNC 440 (GIV CNC 440) yields high values at concentrations below 10^3 cm^{-3}. Furthermore, at concentrations above 10^4 cm^{-3} somewhat low concentration values are obtained from the Diffusion Aerosol Spectrometer (DAS). A similar behaviour of Pollak Type Counter Netzsch CNC 440 (GIV CNC 440) and

(a)

Ag 7nm + NaCl 18nm

(b)

Ag 7nm + NaCl 18nm

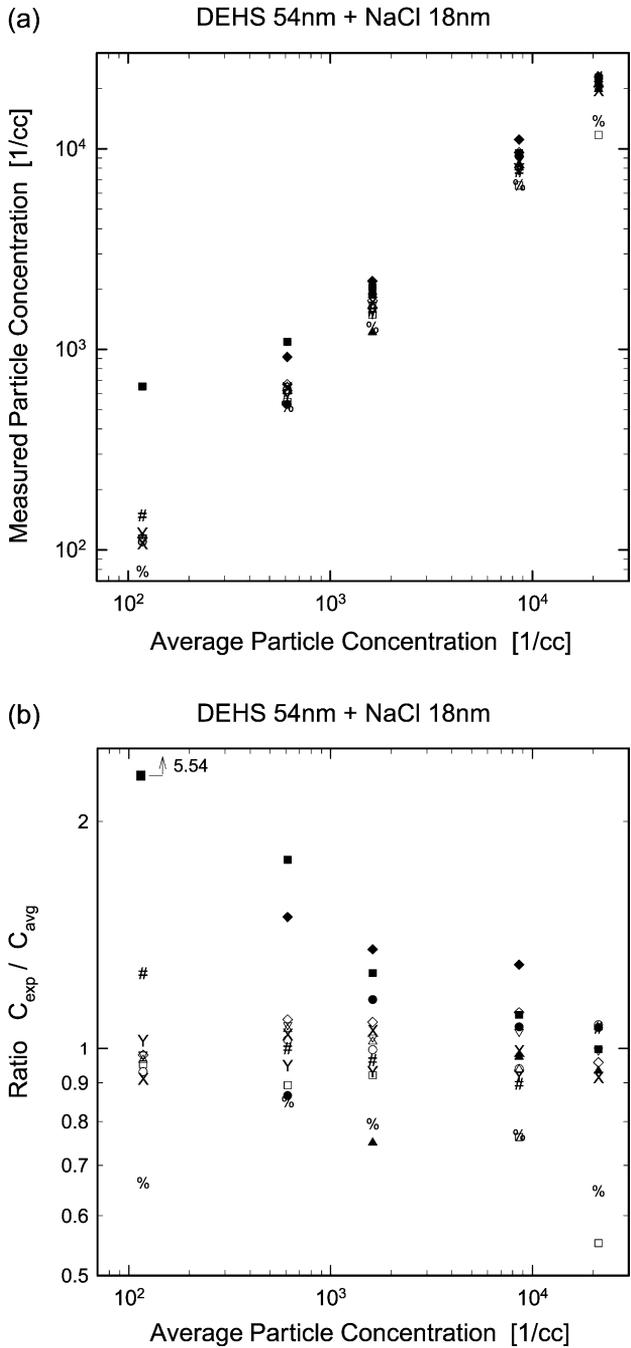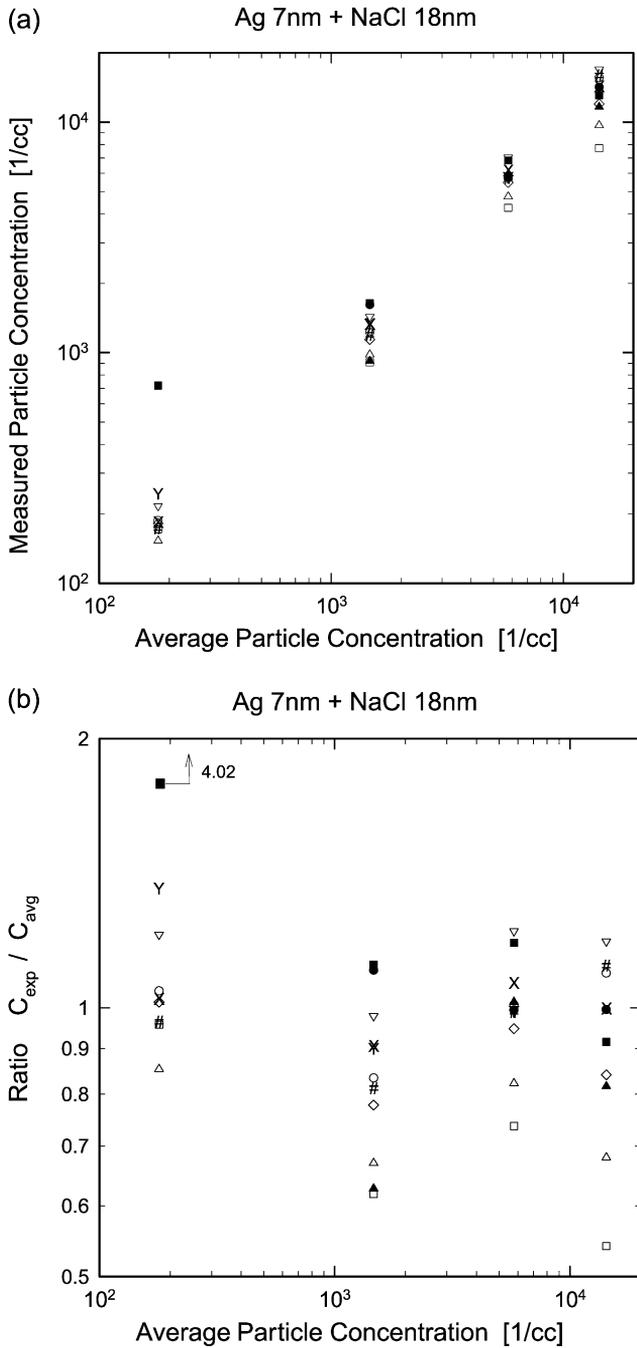Fig. 15. (a–c) Results of the concentration intercomparisons for mixtures of Ag aerosols with particle diameter 7 nm and NaCl aerosols with particle diameter 18 nm.
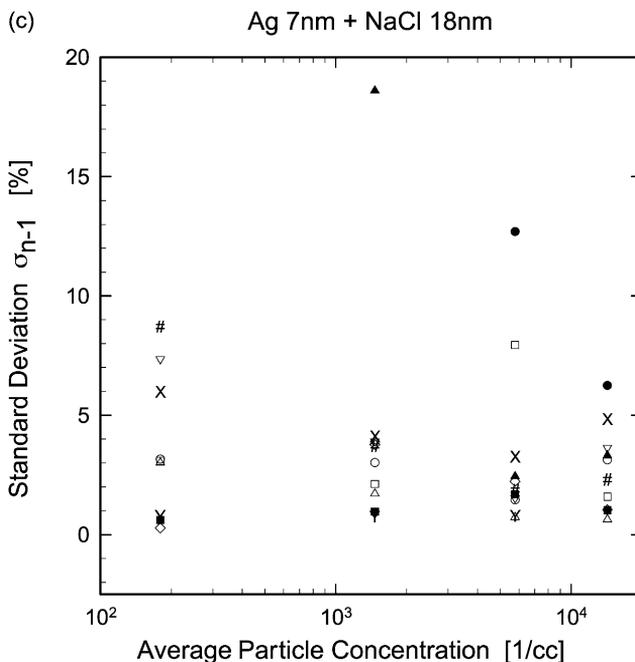
(c)

Ag 7nm + NaCl 18nm

Fig. 15 (*continued*).

Diffusion Aerosol Spectrometer (DAS) has been observed for the hydrocarbon particles, as seen from Fig. 12b.

5. Conclusions

In joint experiments, we have compared total aerosol particle number concentrations, as measured by various aerosol measurement systems. Sixteen different instruments were considered, of which six were adiabatic expansion condensation particle counters, three were flow diffusion condensation particle counters, three were turbulent mixing condensation particle counters, one was a laser optical particle counter and three were electrostatic particle measurement systems. Well-defined monodispersed or nearly monodispersed test aerosols were considered. A few measurements were performed with two-component aerosol mixtures with bimodal size distributions. In several cases, monodispersed aerosol fractions were obtained by means of electrostatic classification. Comparatively wide ranges of particle size and number concentration were covered, five different particle compounds were studied.

Except for a few cases, the particle number concentrations obtained from the various instruments typically agreed within about a factor of two. Only a few of the instruments considered exhibited significant calibration errors in ranges of either low or high particle number concentrations. Strong deviations of up to four orders of magnitude were observed

only in situations, where an instrument was operated outside its specified range. During measurement series with electrostatically classified test aerosols, the Electric Aerosol Spectrometer (EAS 4) was sampling unipolarly charged aerosol, and accordingly in these cases, the accuracy of the total number concentrations obtained from this instrument tends to be increased.

The scatter of the number concentrations measured by each individual instrument at constant conditions typically did not exceed 20% to 25%. Several of the instruments, including electrostatic particle measurement systems, tend to show increased experimental scatter at concentrations below about 10^3 cm^{-3}.

At somewhat elevated concentrations, flow diffusion condensation particle counters partly tend to underestimate the number concentrations, particularly for NaCl particles, whereas a different trend was observed with DEHS particles. This behaviour may be related to the fact that these particle counters are operated with *n*-Butanol as the working liquid, and DEHS is soluble, while NaCl is not soluble in *n*-Butanol. The response of aerosol counters for different particle compositions is further discussed in Ankilov et al. (2002).

Acknowledgements

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