

Aerosol-spectrometers for particle number, size and mass detection

F. Schneider¹

Abstract

The aim of this article is to give some basic fundamentals about the functional principles of aerosol spectrometers, procedures for calibration and validation of aerosol spectrometers and some application examples of actual aerosol spectrometer used for particle number, size and mass detection in agricultural or environmental applications.

Keywords: aerosol Spectrometer, particle size distribution, PM10, PM2.5, PM1

Introduction

This paper will focus on the application of light-scattering aerosol spectrometers for particle number, size and mass detection for high particle concentrations, excluding such applications for clean spaces, where the needed specifications of the aerosol spectrometers are very different, e.g. accurate particle size determination is not requested, sample volume flow is comparably high and maximum particle concentrations are very low.

The determination of particle concentration, particle size distribution and particle mass of aerosol particles is required in various application fields, e.g. chemical, pharmaceutical, agricultural, or automotive industry. During the last decade some remarkable steps in aerosol spectrometer development took place, so today aerosol spectrometers are used to obtain all this results, mentioned above.

Aerosol spectrometers

Fundamentals and detailed descriptions can be found in literature about light scattering of single particles (Van der Hulst H. C. 1957, Bohren C. F. and Hufmann D. R. 1998), basic functional principles of aerosol spectrometers (e.g. Seinfeld J. H. and Pandis S. 1998 or in German language Haller P. 1999) and different technical specifications (e.g. Baron P. A. and Willeke K. 2001 or Hinds W. C. 1998).

Light scattering of single particles

Aerosol spectrometers are based on the interaction between an incident light beam and single aerosol particles. The first description and calculation for the elastic scattering and absorption by aerosol particles according to the incident wavelength are the Maxwell equations (Mie G. 1908). Light scattering can be divided in three ranges, defining an optical size parameter x by $x = \pi * D_p / \lambda$, with the aerosol particles circumference $\pi * D_p$ and the incident wavelength λ :

$x \ll 1$: Rayleigh-scattering: particle diameter is much smaller as the incident wavelength. The intensity of the scattered light is proportional to the sixth power of particle diameter ($I \sim D_p^6$). This is typically for e.g. gas molecules interaction with solar radiation in the atmosphere, causing blue sky effect or very small aerosol particles affecting the lower size detection limit of an aerosol spectrometer.

¹ Grimm Aerosoltechnik GmbH & Co KG, Ainring, Germany

$x \cong 1$: Mie-scattering: particle diameter is about the incident wavelength. There is a strong interaction between the aerosol particle and the incident beam, depending although on particle refractive index with no simple relation between scattered intensity and particle diameter (especially for mono chromatic light, e.g. laser). This is most relevant for atmospheric aerosol particles.

$x \gg 1$: Geometric Optics: light rays hitting the particle lead to reflection, refraction and absorption, rays passing the particles edge give rise to diffraction. The scattered intensity is proportional to the particle cross-sectional area ($I \sim D_p^2$). This will cause e.g. cloud optical effects with water droplets or rainbow effect and affects an aerosol spectrometers upper size detection limit for coarse particles.

Beside the incident wavelength and the particle diameter the (complex) refractive index of a aerosol particle defining its scattering and absorption is very important. Depending on their chemical composition and the internal mixture of aerosol particles the refractive index influences the scattering intensity of an aerosol particle. It has to be mentioned, that scattering intensity is not symmetrically around an aerosol particle, but with strong differences in scattering angles (0° = backward direction, 90° scattering or 180° forward direction).

All together, the scattering intensity given by a single particle depends on the wavelength, intensity and polarisation angle of the incident light, the detection angle of the scattered light and the diameter and complex refractive index of the aerosol particle.

So different aerosol particles generate different scattered light impulses, which can be detected to determine particle concentration and determining the intensity of the scattered light impulse with a pulse height analyser it is also possible to determine the size of the aerosol particle.

Functional principles

An aerosol spectrometer operates by leading single aerosol particles through a light beam or through an intensively lighted measuring volume. The light pulse which is scattered by the single particle is measured and also its intensity. Knowing the sample volume flow and the measuring period the particle number concentration can be derived from the number of the counted scattering pulses. The intensity of the scattered light can be interpreted to a particle size. Depending on calibration procedure and information about the micro physical properties of the measured aerosol particles it is also possible to calculate the particle mass for a given particle size fraction.

There are many types of aerosol spectrometers commercially available, differing in their technical setups and main application fields. In the following the main components of an aerosol spectrometer will be explained, with regards to

different technical solutions.

As light source either lasers (e.g. diode-lasers, He-Ne-lasers) or intensive white light (e.g. xenon high-pressure lamps) is used. The light has to be focuses by a beam optic into a well defined, homogeneously illuminated optical measuring volume. The beam optic has to full fill several aims. Aerosol spectrometer using a laser have to compensate inhomogeneity of the laser beam intensity, caused by the Gauss-shaped distribution over the laser beam diameter. Also the border zone error, caused by aerosol particles which not fully pass the optical measuring volume or pass the laser beam in an area where its intensity is lower and the coincidence error, caused by two or more aerosol particles in the optical measuring volume at the same time have to be avoided (e.g. by an optical measuring volume limitation) or minimized. Beside the optical limitation the aerosol flow can be focused into the light beam, to achieve a so called aerodynamic measuring volume limitation.

After the optical measuring volume the beam might be collected by a light trap.

The aerosol flow passing the light beam in the measuring volume has to be strictly volume controlled, to assure the known time while an aerosol particle stays in the measuring volume and to be able to calculate the particle concentration in the sample.

The scattered light is collected by the detector optics under a certain solid angle with known aperture and led on a detector, typically a photodiode or a photo-multiplier. Aerosol spectrometers are available with detectors placed under different angles, namely in backward (appr. 0° - 45°), forward ($\sim 180^\circ$) or 90° direction. The detector position and aperture can be chosen in a way to minimize the influence of refractive index on scattering intensity and to compensate Mie-interferences in scattering intensity when laser light is used. On the other hand signal to noise ratio of the scattering signal also is strongly influenced by the detector position and aperture angle. Finally the signal has to be amplified and analyzed by a pulse height analyzer.

The setup of an aerosol spectrometer will influence the specification such as precision, particle size range, number of size classes, maximum concentration range but also price, costs for calibration and service, size and weight or roughness. Beside the technical specification of the optical detection mentioned above, aerosol spectrometers are available with different setups for special applications, e.g.:

- automatic coincidence indication, to increase counting efficiency
- isokinetic sampling inlets for measuring in high velocities
- implementation of Nafion dryers to reduce humidity in sampling air without heating and without affecting particle concentration or size distribution

- integrated filters to validate particle mass calculation or enable further investigation of analysed aerosol particles (gravimetrically, microscopically or chemically)
- ex-proof or temperature resist sensors to enable measurements in critical atmospheres and conditions
- battery powered for portable, mobile use

If a device measures the scattering intensity not from single particles but from multiple particles this photometers or nephelometers are not able to determine particle size distribution. Also particle concentration only can be measured with these systems, when certain requirements are met (particle size distribution and refractive index must be constant during a series of runs). This is for many applications most unlikely, so this type of photometers must not be mixed with aerosol spectrometers.

Calibration

Actually there is no national or international standard for calibration of optical aerosol spectrometers, yet. But both international (ISO) and national (VDI) working groups have been initialised to close this gap. The aim of these guidelines is to describe a calibration procedure and a validation method of aerosol spectrometers, to minimise the deviation in the results measured by a single aerosol spectrometer and to minimize difference in the results measured by different instruments.

Particle size can be calibrated by mono disperse standard particles, poly disperse aerosols in combination with cyclones (Binning J. et al. 2006) and theoretically by calculating calibration curve (scattering intensity vs. particle size for given refractive index or materials). For the theoretical calculation it is fundamental to know all parameters of the optical setup, e.g. published by the manufacturer, otherwise the results are completely wrong as published e.g. by Vetter T. 2004.

Two different types of size standards so called primary and secondary standards are available: Mono disperse, spherical reference particles, certified by the manufacture as primary calibration standards mostly in aqueous suspension, like polystyrene-latex (Boundy R. H. and Boyer R. F. 1952). The size of these particles usually was examined microscopically (light or electron microscope). A second possibility to use standard particles is to generate mono disperse reference particles directly into the gas phase, a so-called secondary standard. The users are responsible on its own to full fill repeatability and accuracy of this method. The dispersion of an aerosol using a generator is influenced by the electrically charge of the particles, so the standard aerosol must be neutralised by suitable methods. Water coatings on the standard particles, stabilisation from the solution or agglomerates of standard particles might

cause problems during calibration procedure (Haller P. 1999).

For the calibration of particle concentration no primary standard exists. The counting efficiency only can be determined by comparison of an aerosol spectrometer with a well-defined reference unit (aerosol spectrometer or other e.g. Differential Mobility Analyzer, DMA and Condensation Particle Counter CPC).

Application examples for particle counting, sizing and mass determination

The selected examples show results from particle measurements both counting, sizing and mass determination. All measurements have been carried out with laser aerosol spectrometers of various models manufactured by Grimm Aerosoltechnik, Ainring.

All Grimm aerosol spectrometers operate with a very similar optical detection principle:

- diode-laser with 780 nm or 683 nm
- 15 channel or 31 size channels
- 90° scattering light detection with a given aperture of about 60°
- maximum particle concentration without coincidence 2,000 particles/cm³
- 1.2 l/min sampling flow

The spectrometers are available as portable model measuring particle concentration or particle mass for all size channels and equipped with an integrated 47 mm-PTFE filter for gravimetrically control of dust mass. Other Version is 19" rack version for continuously measurements of PM₁₀, PM_{2.5} and PM₁ (simultaneously) or particle concentration in 31 size channels. These models are equipped with a special sampling pipe to avoid condensation and to control relative humidity and optionally with external sensors, special weather housing for continuously measurements.

Figure 1 and 2 show a measurement during 24 hours in a laying hen stable in summer (figure 1) and winter (figure 2) performed with a 15 size channel aerosol spectrometer Model 1.108, Grimm and a time resolution of 1 minute mean values. The size channels were summed up to one fraction smaller 1 µm and one fraction bigger 1 µm.

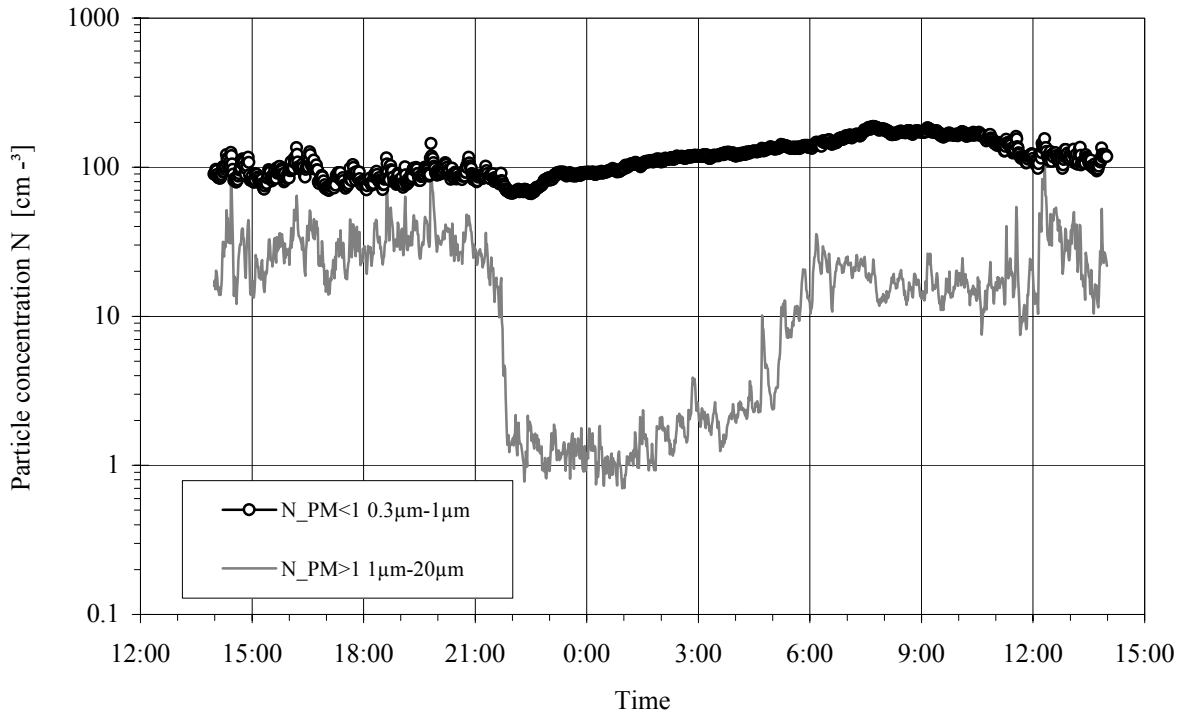


Figure 1:

Particle concentration in summer time in a laying hen stable for two size ranges (0.3 μm - 1 μm and 1 μm - 20 μm) during 24 hours, with a time resolution of 1 minute (Schneider C. 2005)

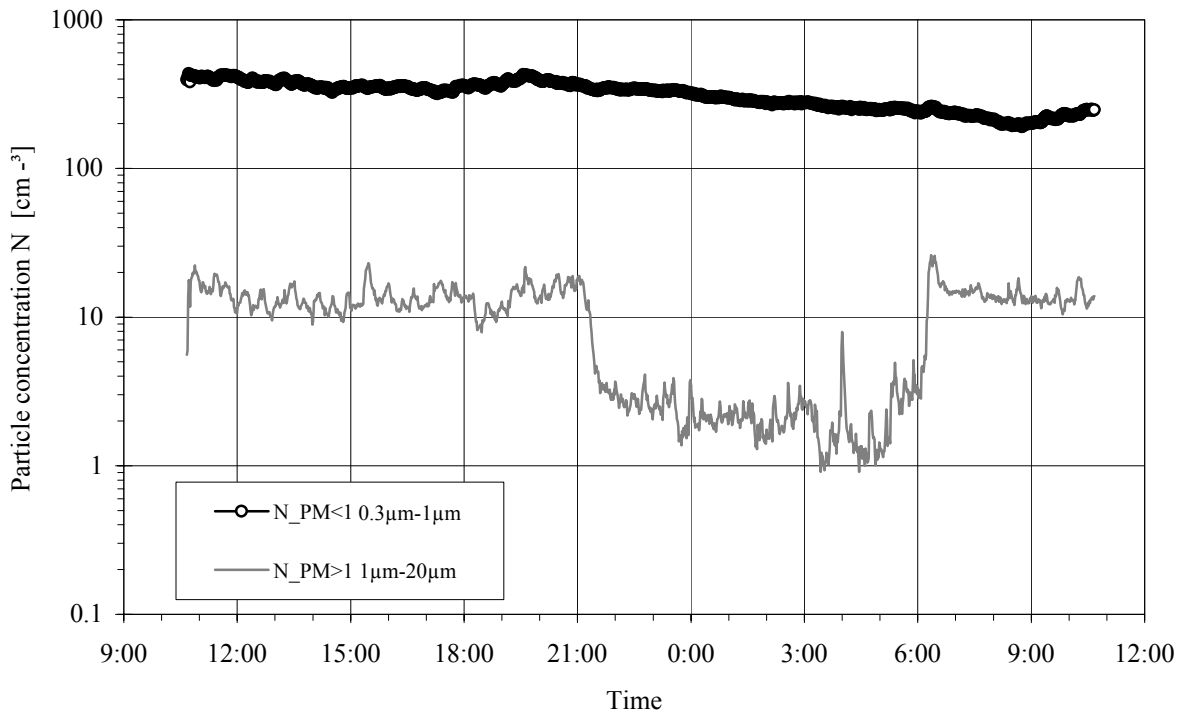


Figure 2:

Particle concentration in winter time in a laying hen stable for two size ranges (0,3 μm - 1 μm and 1 μm - 20 μm) during 24 hours, with a time resolution of 1 minute (Schneider C. 2005)

The figures above clearly show the differences in particle number concentration between the seasons. High ventilation rates during summer (figure 1) cause a dilution of particles smaller $1\ \mu\text{m}$. During night a seasonable influence on coarse particles can be neglected. Size selective concentration measurements with high temporal resolution containing a lot of information about air quality, ventilation performance or animal hygiene.

Figure 3 and 4 show a measured normalized particle size distribution with a Grimm Wide Range Aerosol Spectrometer consisting of an aerosol spectrometer (Grimm Model 1.108) in combination with a SMPS system for nano particle counting (CPC, Grimm Model 5.400) and sizing (DMA, Grimm Model 5.500). The measurement took place in a cattle stable (figure 3) and in a pig stable (figure 4).

The combination of optical particle counters with a SMPS system enables to measure a complete size range from $5\ \text{nm}$ up to $20\ \mu\text{m}$ with one set up.

The graphs in figure 5 and 6 show data sets from a simultaneous measurement of two environmental dust monitors, Grimm, mode 1 107, during a winter day. These aerosol spectrometers are able to obtain PM_{10} , $\text{PM}_{2.5}$ and PM_1 mass concentration simultaneously. The spectrometers were mounted in a special weather housing for temperature and humidity control. One spectrometer (figure 6) was equipped with a heated sampling inlet, the other spectrometer (figure 5) was standard version with no temperature modification.

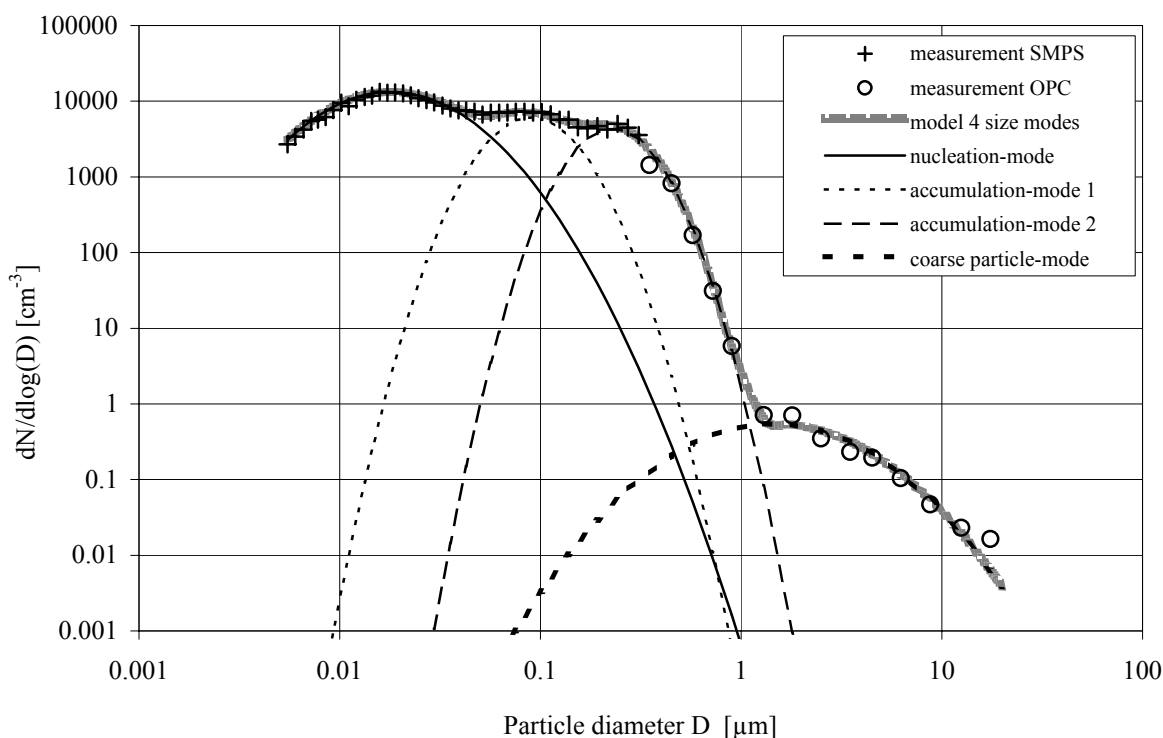


Figure 3:

Normalized particle size distribution in a cattle stable measured with an optical particle counter (OPC $0.3\ \mu\text{m} < D < 20\ \mu\text{m}$) and a sequential mobility particle sizer (SMPS $0.0055\ \mu\text{m} < D < 0.3\ \mu\text{m}$) and modelled particle size distribution ($0.0055 < D < 20\ \mu\text{m}$) calculated for four lognormal size modes, namely nucleation-mode, accumulation-mode 1, accumulation-mode 2 and coarse particle-mode (Onyeneke-Edwards H. C. 2006)

The results in figure 3 and 4 show, that the optical particle counter and the SMPS system measure in the overlap size range around $0.3\ \mu\text{m}$ in the same concentration range, also completely different techniques are compared (e.g. optical latex-equivalent diameter and electrical mobility diameter). This means that the used calibration methods for number and size are suitable and lead to precise and reproducible results. The measured size distribution easily can be explained by combined log normal distributions.

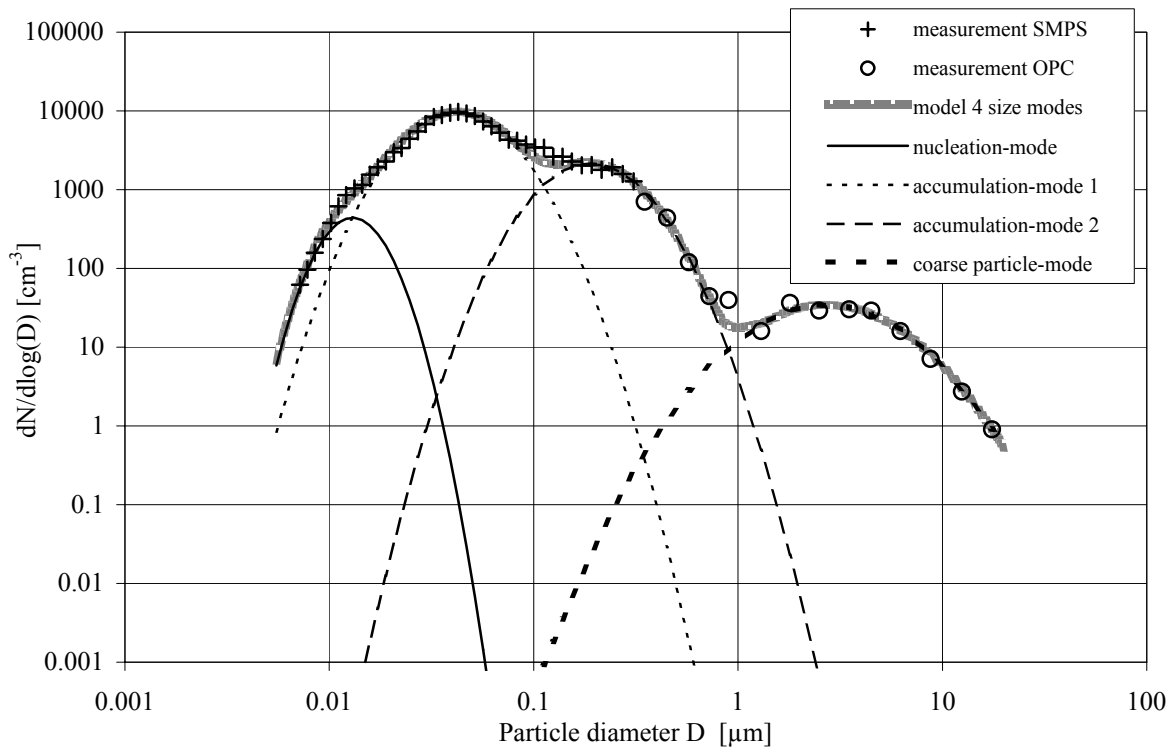


Figure 4: Normalized particle size distribution in a swine stable measured with an optical particle counter (OPC $0.3 \mu\text{m} < D < 20 \mu\text{m}$) and a sequential mobility particle sizer (SMPS $0.0055 \mu\text{m} < D < 0.3 \mu\text{m}$) and modelled particle size distribution ($0.0055 < D < 20 \mu\text{m}$) calculated for four lognormal size modes, namely nucleation-mode, accumulation-mode 1, accumulation-mode 2 and coarse particle-mode (Mahmoud-Yasin N. 2006)

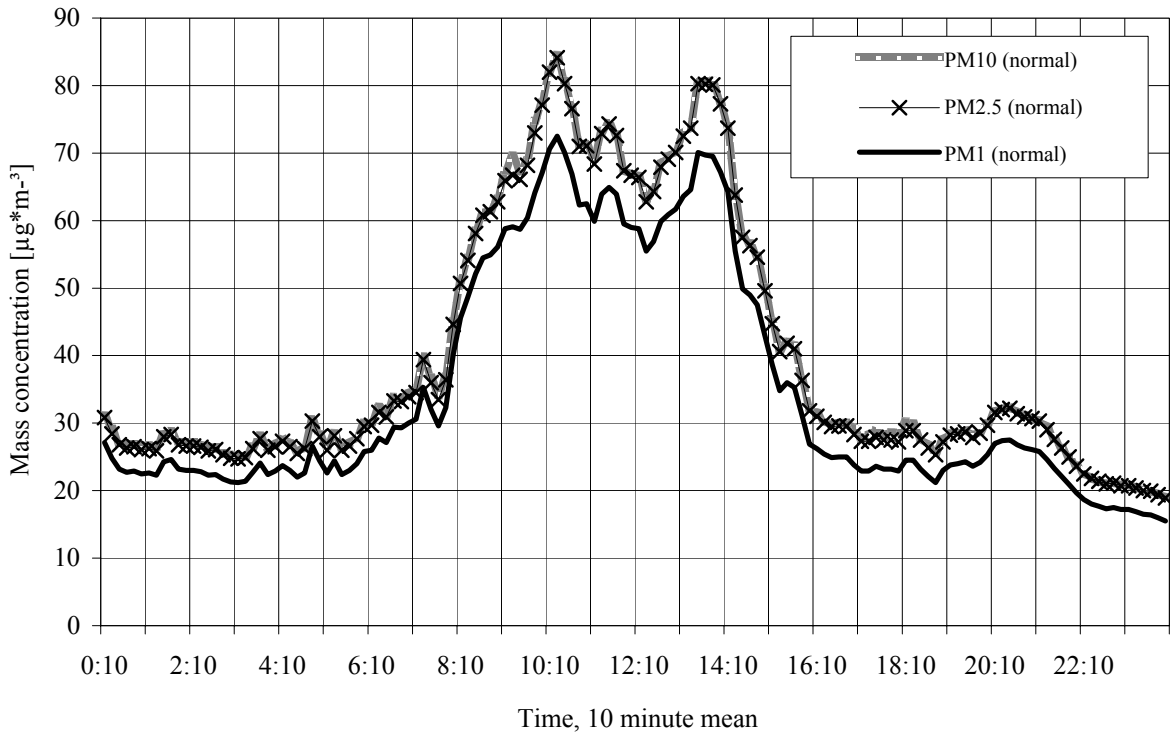


Figure 5: Mass fraction PM10, PM2.5 and PM1 during a winter day, measured with a standard sampling inlet

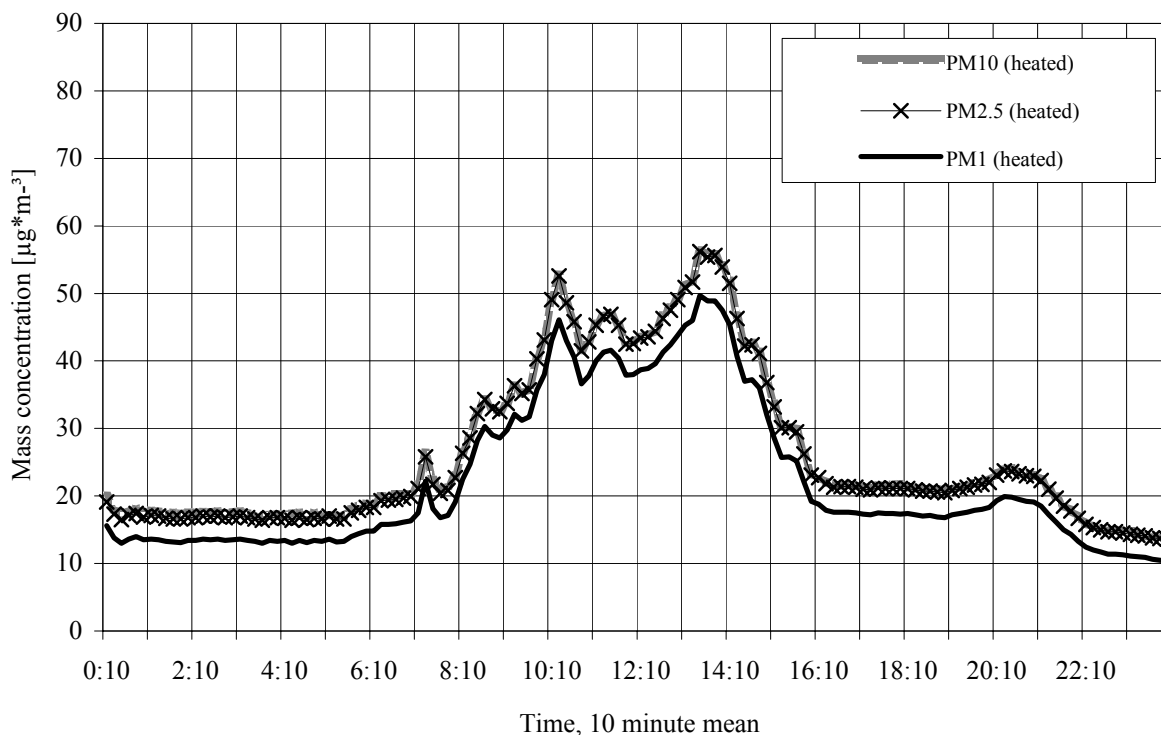


Figure 6:

Mass fraction PM10, PM2.5 and PM1 during a winter day, measured with a heated sampling inlet

As indicated in the graphs in figure 5 and 6 it can be seen, that PM10 and PM2.5 fraction are almost the same. Looking on the effect of the heated sampling inlet, it is obvious, that about 1/3 of the particulate matter is semi volatile compounds, which are lost, if the sampling inlet is heated. This measurements have been compared with mass fractions collected with standard filter samplers, running at the same location simultaneously and chemical analyses. The Grimm 107 was used in many international campaigns with very good results, e.g. Gorny R. L. et al. 2002, Liao C.-M. et al. 2003, Putaud J.-P. et al. 2003, Querol X. et al. 2004.

The further development of the model 107 is the Grimm model 180 environmental dust monitor. This stationary 19" rack version has been approved (Umweltbundesamt, 2006) as an equivalent PM10 measuring system, according to EN 12341! It runs with a included Nafion dryer, which enables simultaneously determination of PM10, PM2.5 and PM1 mass fraction, while the PM2.5 mass fraction includes the semi-volatile species, but excluding water. This is a big advantage in comparison to other environmental dust samplers as described e.g. by Delbert J. E. et al. 2007.

As shown above, aerosol spectrometers successfully can be used in applications, where particle number, size or mass has to be determined.

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