Comprehensive Measurement of Atmospheric Aerosols with a Wide Range Aerosol Spectrometer

L. Keck, M. Pesch and H. Grimm
GRIMM Aerosol Technik GmbH & Co. KG, Dorfstrasse 9, D-83404 Ainring, Bayern, Germany
Email: lk@grimm-aerosol.com

Abstract. A wide range aerosol spectrometer (WRAS) was used for comprehensive long term measurements of aerosol size distributions. The system combines the results of an optical aerosol spectrometer with the results of a Scanning Mobility Particle Sizer (SMPS) to record essentially the full size range (5 nm – 32 µm) of atmospheric particles in 72 channels. Measurements were carried out over one year (2009) at the Global Atmospheric Watch (GAW)-Station Hohenpeißenberg, Bavaria. Total particle number concentrations obtained from the aerosol size distributions were compared to the total number concentrations measured by a Condensation Particle Counter (CPC). The comparison showed an excellent agreement of the data. The high time resolution of 5 minutes allows the combination of the measured size distributions with meteorological data and correlations to gaseous pollutants (CO, NOx and SO2). A good correlation of particle number and CO concentrations was found for long distance transported small particles, which were probably mainly soot particles. Correlations to NOx were observed for aerosols from local sources such as traffic emissions. The formation of secondary aerosols from gaseous precursors was also observed. Episodes of relatively high concentration of particles in the range of 2-3 µm were probably caused by pollen.

1. Introduction
Comprehensive measurement of particle size distributions plays an important role in aerosol research. Examples are studies on particle source apportionment, on effects of atmospheric aerosols on human health, and on the role of particle in the climate system [1-3]. Nowadays people pay more and more attention to the airborne nanoparticles particularly because there is the suspect that these nanoparticles are, despite of their small contribution to PM Mass concentrations, relevant for the adverse health effects of atmospheric aerosol. The relatively large surface area and the higher concentration of adsorbed or condensed toxic material per unit mass [4] could contribute to the adverse effects.

Measurement of the full size range of atmospheric particles is a challenge and there is no single measurement principle, which can detect both nanoparticles and coarse particles. The most promising approach for the detection of the complete size range is the combination of (1) a Scanning Mobility Particle Sizer with Condensation Particle Counter (CPC) as detection system (SMPS+C) and (2) an optical aerosol spectrometer (OPC). The SMPS+C covers the size range of 5 – 350 nm in 44 size channels, the OPC covers the size range of 250 nm – 32 m in 31 channels. This instrument enables: (1) Continuous and reliable aerosol measurements over with low maintenance, (2) High time resolution of 5 minutes, (3) High dynamic range (Mass concentrations 1 µg/m³ to 5000 µg/m³; number concentrations 1 P/cm³ to 10⁹ P/cm³), (4) Low modification of the size distribution during sampling by...
diffusion or impaction, agglomeration, and losses of semi volatile components (SVC), (5) Simultaneous measurement with meteorological sensors (6) Operation as a stand alone system even under rugged conditions due to stainless steel weather housing and built-in data logger.

2. Methods
The WRAS system consists of a scanning mobility particle sizer with condensation particle counter (SMPS+C) and an optical aerosol spectrometer (OPC). The GRIMM SMPS+C includes a CPC and a differential mobility analyzer (DMA). The classification occurs in the electrostatic field in the annulus between inner and outer electrodes of the DMA. A particle size distribution (electric mobility diameters) in the range of 5.5 nm – 350 nm can be obtained by changing the DMA voltage stepwise. The GRIMM OPC works on the basis of light-scattering for single particles. The scattered light is detected with a PIN-diode. The signals are amplified and classified in 31 size channels. Thus the particle size number distribution (optical diameters) in the range of 250 nm – 32 \( \mu \text{m} \) is measured. The two datasets from the SMPS+C and OPC are automatically synchronized and combined using the GRIMM software, there is however no fitting or adaption to smooth the transition from mobility to optical diameters. In this way the WRAS system measures particle number size distributions from 5.5 nm up to 32 \( \mu \text{m} \).

The whole system is mounted in stainless steel weather protection housing with air conditioning system. The sampling probe features a Nafion dryer to reduce the humidity to ~40%, thus the particles feature a well defined diameter in the detection systems independent of the ambient humidity. Unlike heated sampling probes, this drying system causes no loss of volatile components. Meteorological sensors for wind speed, wind direction, temperature, humidity, and rain are also integrated in the weather housing.

Data were stored on a notebook in the weather casing. The newest version of the WRAS system features however also a data logger for online data transfer to the internet. All data can now be downloaded by the user from a password protected web-site.

Figure 1. Photo of the WRAS system during regular maintenance on the Hohenpeissenberg.

In this study, a GRIMM WRAS system was applied for comprehensive measurements of atmospheric aerosols at the GAW Station Hohenpeissenberg (47°48' N, 11°02' E, 985m a.s.l.), located on a hill with about 300 m elevation above a rural area in Bavaria, Germany. Measurement started on January, 13th 2009 and lasted essentially one year.
3. Results and Discussion

The typical number size distributions featured a count median diameter of 51 nm. Mass size distributions were calculated from the measured number size distributions, these mass size distribution featured a clear bimodal shape of accumulation mode and coarse mode.

To verify the results from the WRAS system, we have calculated total number concentrations from the measured size distributions and we have compared these values to the total number concentrations measured with a CPC. The two total number concentrations agreed well, the concentrations from the WRAS were however slightly lower than those from the CPC. This small difference is however partly caused by the different minimal detected particle size, being 5.5 nm for the WRAS system and 3 nm for the CPC.

![Figure 2. Comparison of total particle number concentrations (1) calculated from the aerosol size distributions measured by WRAS system and (2) measured by a TSI CPC](image)

The size distributions showed pronounced diurnal variations. Typical features were high concentrations of very small particles right after sunrise, which show the particle formation by photochemical processes. A second feature is the increasing concentrations of rather large particles in the afternoon, probably due to the onset of convection. Figure 3 shows the aerosol size distributions on January 23-24, 2009. Although the measurement site is located on a hill with about 300 m elevation above a rural area, large numbers of ultrafine airborne particles were measured. As evident from Fig.3a and Fig.3b, large differences between the diurnal variations of size distributions occur. The high particle concentration at January 24, 21:31 (nanoparticles with CMD of 35.6 nm) result from combustion aerosol particles.
Figure 3. Particle number concentrations on a) January 23, b) January 24.

The total particle number concentrations showed a good correlation to gaseous CO, indicating that CO and particles originate both from the same combustion source. Figure 4 shows exemplarily the time series of gaseous SO$_2$ and total number concentrations. The strong SO$_2$ peak was accompanied by a nearly simultaneous small peak in the particle number concentrations and a second, much higher peak in the particle concentration occurred with approximately 1 h of delay after the SO$_2$ peak. We speculate that the first peak in the particle concentrations was due to the combustion particles which were released together with the SO$_2$, the second larger peak in the particle concentrations might consist of secondary sulfate particles, which were formed from the gaseous precursor with the consequence that the gas concentrations were lower in that air parcels. Such sulfate particles feature, unlike nitrate particles, only a accumulation mode.
4. Conclusions
The Wide Range Aerosol spectrometer employed for this test made it possible to measure the complete size range of atmospheric particles over one year with a size resolution of 72 channels and a time resolution of 5 min.

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