Comprehensive Measurement of Atmospheric Aerosols with a Wide Range Aerosol Spectrometer

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In this paper, the measuring principle of a wide range aerosol spectrometer (WRAS) with more than 70 size channels from $5.5~\rm nm$ to $32~\rm \mu m$ is introduced. Comprehensive measurements of atmospheric aerosols were performed with a GRIMM WRAS system at the Global Atmosphere Watch (GAW) Station Hohenpeissenberg in Germany. These wide range data were combined with simultaneous monitoring of meteorological parameters and gaseous pollutants. Total particle number concentrations obtained from the aerosol size distributions measured with the WRAS system were also compared to the TSI CPC counts and TEOM results, showing a good correlation. The correlation between these data allows the source apportionment of particles, the investigation of aerosol transport, transformation and mixing processes, as well as the research on the effect of atmospheric aerosols on the human health and climate.

1. Introduction

Comprehensive measurement of atmospheric aerosols, e.g. aerosol size distribution, plays an important role in the atmospheric aerosol investigations, such as particle source apportionment, effect of atmospheric aerosols on human health and climate (Anderson, 2000; Birmili et al., 2009; Oberdörster et al., 1996). Nowadays people pay more and more attention to the airborne nanoparticles due to the relatively large surface area, i.e. airborne nanoparticles have higher concentration of adsorbed or condensed toxic material per unit mass (Sioutas et al., 2005). 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 300m elevation above a rural area in Bavaria, Germany.

2. Methods

As illustrated in Fig.1, this mobile WRAS system consists of a scanning mobility particle sizer with condensation particle counter (SMPS+C) and an optical aerosol spectrometer (OPC). The whole system is mounted in stainless steel weather protection housing. The sampling probe has a Nafion dryer inside, which causes no loss of volatile components during the measurement. Optional meteorological sensors are integrated.

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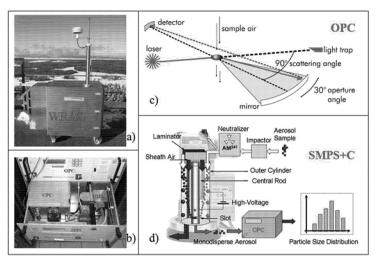


Fig.1 a) WRAS system mounted in a stand-alone stainless steel weather housing; b) Configuration of the WRAS system consisting of OPC,CPC and DMA; c) Measurement principle of the GRIMM OPC; d) Measurement principle of the GRIMM SMPS+C.

2.1 Measurement Principle of SMPS+C

The GRIMM SMPS+C includes a CPC and a differential mobility analyzer (DMA), as shown in Fig.1d. During the measurement larger particles are removed firstly by an impactor at the inlet of the DMA, and then fine and ultrafine particles are classified with a DMA after they pass through a bipolar charger (Neutralizer Am-241), which establishes a well defined charge distribution of the particles. The classification occurs in the electrostatic field in the annulus between inner and outer electrodes of the DMA. Only particles of a certain size or mobility reach a narrow slit at the bottom of the inner electrode and are measured with a CPC. Thus, a particle size distribution (PSD) in the range of 5.5 nm – 350 nm can be obtained by changing the DMA voltage stepwise.

2.2 Measurement Principle of OPC

The GRIMM OPC works on the basis of the light-scattering technology for single particle counting. As shown in Fig.1c, a semiconductor laser serves as the light source. The scattering pulses from the particles passing the laser beam are collected by a mirror with an accurately defined aperture angle and transferred to a recipient-diode positioned under 90° to the incident laser beam. The detected signals are further preamplified and classified in 31 size channels, resulting in a PSD in the range of 250 nm - 32 μm . Finally, these two datasets from the SMPS+C and OPC are automatically synchronized and combined using the GRIMM software. In this way the WRAS system allows the measurement of wide range particle size distributions from 5.5 nm up to 32 μm .

3. Results

By using this WRAS system, aerosol size distributions were continuously measured at the GAW Station Hohenpeissenberg for one year. Total particle number concentrations obtained from the aerosol size distributions measured with the WRAS were compared to the TSI CPC counts and TEOM results. These wide range data were also combined with simultaneous monitoring of meteorological parameters and gaseous pollutants.

3.1 Aerosol Size Distributions

Figure 2 shows the aerosol size distributions measured with the WRAS at the GAW Station Hohenpeissenberg on 1st-4th Feb, 2009. It is seen that large numbers of ultrafine airborne particles have been monitored though this WRAS system ran at about 300m elevation above a rural area in Bavaria, Germany. Aerosol size distributions measured with the GRIMM WRAS system have a high size resolution with over 70 size channels ranging from 5.5nm to 32μm as well as a high time resolution of the scans (3-4 minutes). From these wide range data one can observe the airborne particle number concentrations change with the measuring time. The diurnal variations of 3D aerosol size distributions reveal the concentration and size range of the measured airborne particles, i.e. source variations. This is very helpful for particle source apportionment. For example, a peak occurred at about 16:00 in Fig.2d and this high particle concentration resulted from combustion aerosol particles, which flowed over the GAW station.

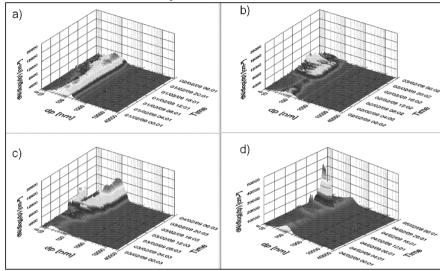


Fig.2 Continuous measurement of aerosol size distributions with the WRAS system at the GAW Station Hohenpeissenberg on 1st-4th Feb, 2009.

3.2 Comparison with Total Number Concentrations and TEOM Results

Based on the WRAS measurement of aerosol size distributions dN/dlog(dp), as illustrated in Fig.2, total particle number concentrations can be obtained by integral of all these size channels. Corresponding particle counts measured with the WRAS system on 1st-4th Feb, 2009 were plotted with hollow dots in Fig.3. A comparison with the results by a TSI CPC (plotted with solid dots) was made in Fig.3a. A quite good agreement between GRIMM WRAS and TSI CPC is visible, though both measuring systems work on the different principles, respectively. The TSI CPC results are somewhat larger than those measured by the GRIMM WRAS due to their different measuring ranges of aerosol size. The GRIMM WRAS system measures the particle size distributions ranging from 5.5nm upward while the TSI CPC measures the particles larger than 3nm. In this case more small particles are measured with the TSI CPC.

Furthermore, total particle number concentrations obtained from the aerosol size distributions measured by using the WRAS were compared to the TEOM results, also showing a good correlation, as shown in Fig.3b.

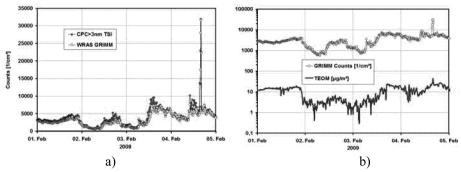


Fig.3 Total particle number concentrations obtained from the aerosol size distributions measured with the WRAS system at the GAW Station Hohenpeissenberg on 1st-4th Feb, 2009, corresponding to Fig.2. a) Compared with the results by a TSI CPC; b) Compared with TEOM results.

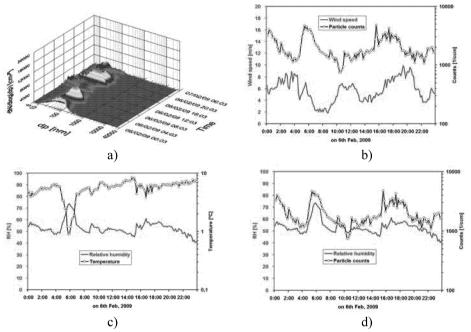


Fig.4 Wide range results correlated to meteorological parameters. a) Diurnal variation of aerosol size distribution on 6th Feb, 2009; b) WRAS result vs wind speed; c) Relative humidity vs temperature; d) WRAS result vs relative humidity.

3.3 Correlation to Meteorological Parameters and Gaseous Pollutants

Changes in particle size distribution and particle number concentration can be observed simultaneously. High time resolution of the scans allows detailed comparisons with meteorological data (e.g., T, RH, Wind) and gaseous pollutants (e.g. SO₂, NOx, O₃, CO). With some metrological sensors combined, a group of wide range results (aerosol size

distribution and number concentration) are compared with meteorological data on 6th Feb, 2009. A negative correlation between relative humidity and temperature is found in Fig.4c while a positive correlation between relative humidity and particle counts is observed in Fig.4d. Furthermore, this comparison reveals that the rise in temperature enhances the vertical convection of the atmosphere, leading to a reduction in the aerosol number concentration. Aerosol mixing process also affects the particle concentration, because the variation of wind speed and direction at the measuring site can cause the turbulent diffusion or irregular air flow. Such processes can dilute the pollutants at the measuring site. Fig.4b shows the relation between wind speed and particle counts. As the wind speed increases, particle concentration decreases.

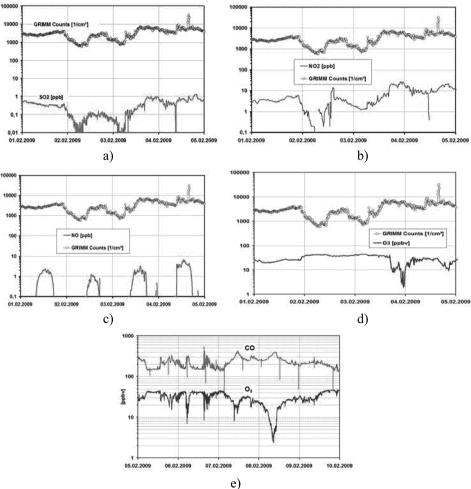


Fig. 5 Wide range results correlated to gas pollutants on 1^{st} - 4^{th} Feb, 2009. a) WRAS result vs SO_2 ; b) WRAS result vs NO_2 ; c) WRAS result vs NO; d) WRAS result vs O_3 ; e) Relation between O_3 and CO.

Continuous observations show that each diurnal variation of aerosol PSD is different due to the complicated influence factors of meteorological parameters, local pollutants, traffic emission, and photochemical reaction. A good correlation between particle counts and the concentration of SO_2 and NO_2 from combustion processes is observed, as illustrated in Figs.5a-b. It means that detected particulates contain large numbers of secondary atmospheric aerosol particles, e.g. sulfate and nitrate, which are formed from the gas pollutants in the atmosphere through the gas-to-particle conversion process. The concentration of NO is high at daytime due to the high sunlight intensity and reaching a maximum at noon, but it is very low and almost undetected at night, as displayed in Fig.5c. This is due to the photochemical reaction, i.e. photolysis and reformation of NO_2 . Other gas pollutants, e.g. O_3 and CO, were also monitored. A negative relation between particle counts and O_3 was found, seen in Fig.5d. From a continuous observation at the GAW station Hohenpeissenberg in 2009, it is found that gas pollutants O_3 and CO were highly negatively correlated. A part of these results was plotted in Fig.5e. However, it is worth pointing out that CO and O_3 might not be always perfectly correlated, depending on the photochemical reaction and the transport process (Zahl and Brenninkmeijer, 2003).

4. Conclusions

Numerous continuous measurements of aerosol size distributions indicate the time variation of airborne particles from various sources, like rush hour, vehicle emissions, power plants out of the measurement site, and chemical components due to photochemical reactions. Furthermore, combination with meteorological data and gaseous pollutants helps study aerosol transport, transformation and mixing processes. Therefore, GRIMM WRAS is a valuable scientific instrument for environmental studies, aerosol research, roadside monitoring, engine exhaust studies, and health effect studies.

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