E. Weingartner, S. Nyeki, U. Baltensperger
Paul Scherrer Institut
Villigen / Switzerland

Seasonal Variation of size distributions and fine particle formation of continental aerosol at a high alpine site (Jungfraujoch, 3454 m a.s.l.)

SEASONAL VARIATION OF SIZE DISTRIBUTIONS AND FINE PARTICLE FORMATION OF CONTINENTAL AEROSOL AT A HIGH ALPINE SITE (JUNGFRAUJOCH, 3454 m a.s.l.)

E. Weingartner, S. Nyeki, and U. Baltensperger

Paul Scherrer Institut, 5232 Villigen, Switzerland

Aerosol size distribution of particles with diameters D in the size range 18 nm - 750 nm were measured on a continuous basis (every 30 minutes) with a Scanning Mobility Particle Sizer (SMPS) (Wang and Flagan, 1990) at the high alpine research station Jungfraujoch from March 1997 - May 1998. Because of its elevation this site is suitable for investigating the lower free troposphere. The measurements presented in this paper were performed in addition to those of Nyeki *et al.* (1998) where optically relevant aerosol parameters (scattering, back scattering and absorption coefficient) as well as surface area and number concentrations have been measured. A high seasonality (about an order of magnitude higher concentrations during the summer months) is found in aerosol parameters that are influenced by surface or volume concentration. These higher concentrations are mainly due to a stronger influence from planetary boundary layer air (convective transport) during summertime rather than to increased aerosol formation (by homogeneous and heterogeneous nucleation) (Lugauer *et al.* 1998; Baltensperger *et al.*, 1997).

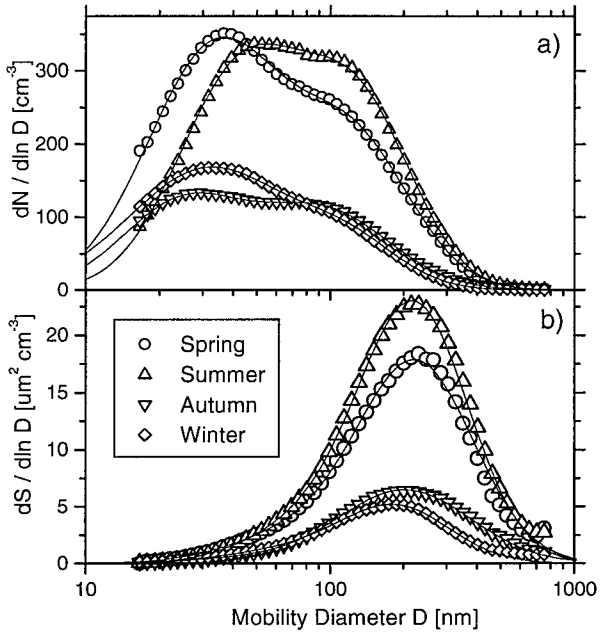

Figure 1. *Seasonal variation of the number (a) and surface (b) size distributions. The points are measured data and the solid lines represent fitted bimodal curves.*

A different mechanism is seen to govern the number concentration. The shape of the number size distribution also undergoes a seasonal variation (Fig. 1a): The number size distribution in summertime is characterized by higher number concentrations (summer: $N_{10} \approx 900\ cm^{-3}$; winter: $N_{10} \approx 500\ cm^{-3}$) as well as larger diameters. The shift towards smaller

particles in wintertime is less distinct if higher moments of the distribution are considered: As an example, Fig. 1b shows the temporal evolution of the normalized surface distribution which was deduced from the measured mobility diameter assuming spherical particles. The invariance of the shape of higher moments of the size distributions supports the idea of a relatively stable size distribution, at least in the accumulation mode (100 nm < D < 1000 nm). Furthermore, it explains the good correlation found between aerosol surface area concentration and optical aerosol parameters (Nyeki *et al.*, 1998). The significant shift towards smaller particles is responsible for the lower seasonality observed in the number concentration compared to surface area or volume concentrations. We suggest that nucleation mode particles (D < 20 nm) are formed by homogeneous nucleation and subsequent growth. This hypothesis is also corroborated by the fact that the number concentration of particles in the size range D = 10 - 18 nm exhibits a diurnal variation which is most probably due to photochemical processes (Fig. 2).

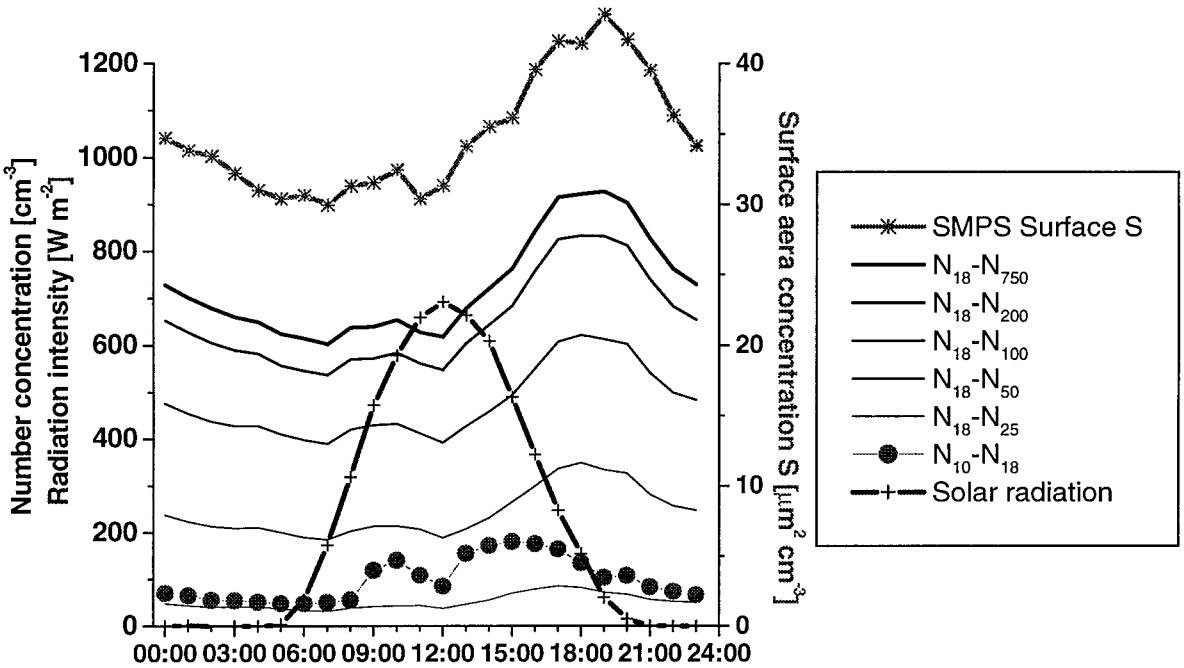

Figure 2. *Average diurnal variation during Summer 97 of the surface area concentration measured by the SMPS (bold line with stars, right hand scale) and number concentration in different size bins (see legend, left hand scale). N_x is the number concentration of particles with diameters D > x nm. The SMPS measurements are represented as solid lines. From the CNC signal the concentration of even smaller particles (N_{10}-N_{18}) was deduced and is represented as open circles. The average solar radiation intensity during the measurements is also shown (line with crosses, left hand scale).*

With the onset of solar radiation in the morning, photochemical reactions which result in the production of condensable species (e.g., $SO_2 + OH \rightarrow H_2SO_4$) are started (Weber *et al.*, 1997). If the pre-existing aerosol surface area concentration is low, homogeneous nucleation is favored and new particles are formed. These nucleation mode particles are not detected with the CNC until they grow into the "visible" size range of the instruments. This delay is ~3 hours for particles with 10 nm < D < 18 nm and ~4 hours for particles with 18 nm < D < 25 nm. At noon the precursor vapor gas reservoir is exhausted which results in lower

production rates. Then, the fraction of Aitken mode particles (20 nm < D < 100 nm) increases again due to thermal convection which provides new precursor gases from the planetary boundary layer. The decrease in new particle formation between 15:00 and 18:00 is most probably due to the increase in the surface concentration and the decrease of sunlight. The concentration of ultrafine particles is significantly higher during Wintertime which is most probably due to the fact that the nucleation rate depends on the surface area concentration which is lower in Winter. In addition, the lower temperatures during Winter time may also favor the formation and growth of new particles. Nevertheless, since the formation of new particles also depends on the concentration of precursor gases, the solar intensity and even on the concentration of ions, the detailed mechanisms which lead to the observed new particle formation are by far not understood.

ACKNOWLEDGEMENTS

This study was run under the auspices of the WMO GAW Project and was supported by the Swiss Meteorological Institute. The support of the staff and facilities at the Jungfraujoch Research Station are greatly appreciated.

REFERENCES

Baltensperger U., Gäggeler H.W., Jost D.T., Lugauer M., Schwikowski M., Weingartner E., Seibert P. (1997) *J. Geophys. Res.* **102**, 19707-19715.

Lugauer M., Baltensperger U., Furger M., Gäggeler H. W., Jost D. T., Schwikowski M. and Wanner H. *Tellus*, **50B**, 76-92, 1998.

Nyeki S., Baltensperger U., Colbeck I., Jost D.T. and Weingartner E. (1998) *J. Geophys. Res.*, **103**, 6097-6107, 1998.

Wang S.C. and Flagan R.C. (1990) *Aerosol. Sci. Technol.*, **13**, 230-240.

Weber R.J., Marti J.J., McMurry P.H., Eisele F.L., Tanner D.J. and Jefferson A. (1997) *J. Geophys. Res.*, **102**, 4375-4385.