Investigation of Atmospheric Processes and Optical Properties of Nanoparticles by Size Distribution Measurements

Marc Schröter, Andreas Obermeier, Dieter Brüggemann

Lehrstuhl für Technische Thermodynamik und Transportprozesse (LTTT)
Universität Bayreuth, Universitätsstr. 30, 95440 Bayreuth, Germany
E-mail: marc.schroeter@uni-bayreuth.de

Introduction

The Intergovernmental Panel on Climate Change (IPCC) reports two key points to improve the scientific knowledge of aerosol radiative forcing and climate models: systematic ground-based measurements of size-segregated concentrations of particle numbers as well as characterisation of particle processes that influence the size-segregated concentration. To address these requirements, measurements of particle size distributions with a Scanning Mobility Particle Sizer (SMPS, TSI model 3639) have been carried out during 2004 at a roadside, urban and rural station, investigating size-dependent particle processes and calculating optical properties from particle size distributions to study their climate effects.

Results

At all stations most total number concentrations are between 1000 and 7000 cm^{-3}, with an average of 4500 cm^{-3} at the roadside and 2500 cm^{-3} at the rural station, respectively. This is within the range of other European measurements (e.g. Van Dingenen et al., 2004). Results show a size-dependent coherence of particle number with temperature and SO_2 concentration, probably caused by size-dependent condensation processes. The more a particle surface is curved, the higher the vapour pressure and therefore the growth of bigger particles is preferred. A size-independent exponential dependency of particle number with wind speed is observed. For all particle sizes we found the higher the wind speed, the lower the number concentrations. This effect is due to a more turbulent mixing at higher wind speeds leading to lower particle number concentrations at ground level. NO_2 concentrations show a size-independent linear influence on particle number, being a precursor gas for secondary formed particles.

To calculate optical properties of particle size distributions, we have to know or assume their average refractive index firstly. Results from measurements reported in literature and the database "Optical Properties of Aerosols and Clouds (OPAC) indicate the usage of a mean refractive index of atmospheric particles as adequate. Estimating the influence of the refractive index on the optical parameters results in the highest gradients with variations of the imaginary part. Consequently, knowledge of the black carbon fraction of atmospheric particles is needed, because of its highest imaginary part and therefore dominating role within an average refractive index.

Secondly, using a numerical correct and highly efficient source code by Mishchenko (2002) the fitting of common distribution models to the measured data is necessary. Here, the fitting of a bimodal lognormal size distribution results the best goodness of fit statistics. Consequently, the average of measured distributions shows a first mode in the diameter range of 30 - 45 nm and a second one around 80 - 90 nm. Although there are uncertainties within the necessary estimation of parameters for calculating optical properties of particles a comparison experiment to Lidar measurements reveals a good agreement.

Conclusions

The particle size distributions of our year-long measurements are best described analytically with a bimodal distribution, consequently showing two modes within the Aitken size range on average. Analysing the correlation of the particle size distribution with meteorological and air quality parameters shows a size-dependency of the number on temperature and SO_2 concentration. A size-independent exponential decrease in particle number with increasing wind speed and a linear rise with increasing NO_2 concentration is also found. These coherences indicate the occurring atmospheric processes, like dilution and condensation.

Estimating optical properties of particle size distributions by assuming an adequate average refractive index and using the Mie theory gives results which are in agreement with literature data and own Lidar measurements. This allows the application of the method within future closure studies to estimate optical properties of atmospheric particles.

References

Mishchenko, M. I., Travis, L. D., Lacis, A. A., 2002. Scattering, absorption, and emission of light by small particles. Cambridge University Press, Cambridge.

Van Dingenen, R., Raes, F., Putaud, J.-P., Baltensperger, U., Charron, A., Facchini, M.-C., Decesari, S., Fuzzi, S., Gehrig, R., Hansson, H.-C., Harrison, R. M., Hüglin, C., Jones, A. M., Laj, P., Lorbeer, G., Maenhaut, W., Palmgren, F., Querol, X., Rodriguez, S., Schneider. S., ten Birk, H., Tunved, P., Torseth,K., Wehner, B., Weingartner, E., Wiedensohler, A., Wahlin, P., 2004. A European aerosol phenomenology - 1: physical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe. Atmospheric Environment 38, 2561-2577.

Investigation of Atmospheric Processes and Optical Properties of Nanoparticles by Size Distribution Measurements

Marc Schröter, Andreas Obermeier, Dieter Brüggemann

Motivation

Knowledge of size-segregated particle processes and optical properties are fundamental for radiative forcing and health effects of particles. But so far, little data on long-term particle size distribution variations and their optical properties are reported.

Methods

- Particle size distributions measurements with a Scanning Mobility Particle Sizer (SMPS, TSI model 3936) at an urban and a rural station (Arzberg and Tiefenbach).
- Investigation of size-dependent processes by classification of particle numbers into parameter categories and standardisation of each size class.
- Calculation of optical parameters from particle numbers fitting a bimodal lognormal-distribution, assuming an adequate average refractive index and using the Mie theory.

Results (Particle Processes)

An exponential correlation of particle number with wind speed and linear dependency with NO_2 concentrations is observed for all detected particle sizes (15-750 nm). This can be explained by an increasing dilution at higher wind speeds and the formation of secondary particles by the precursor gas NO_2, respectively.

Additionally, observed corrrelations with temperature and SO_2 concentration are size-dependent, probably caused by size-dependent condensation processes.

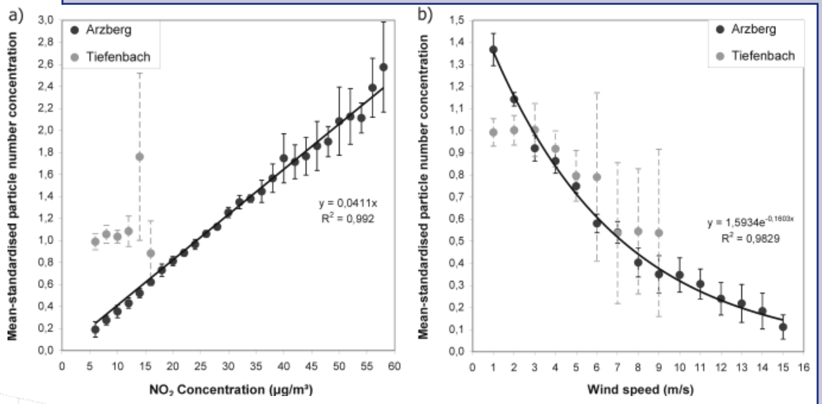

Fig. 1: Dependency of standardised particle number concentrations on NO_2 concentration (left) and wind speed (right).

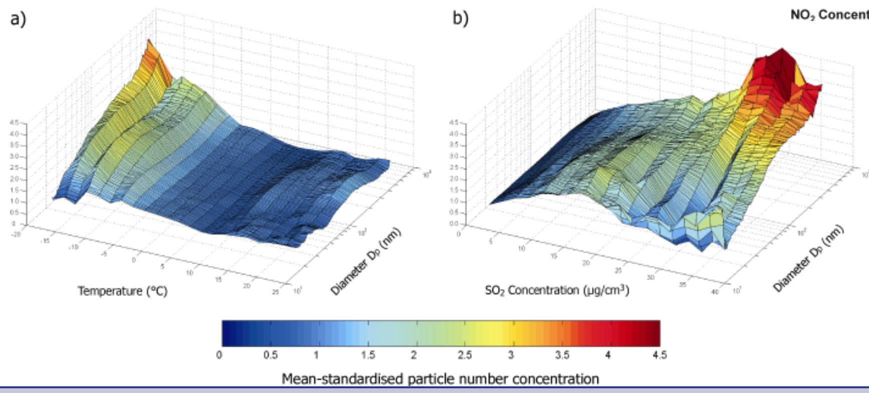

Fig. 2: Correlation of standardised particle number concentrations with temperature (left) and SO_2 concentrations (right) at Arzberg.

Results (Optical Properties)

Estimating the influence of the refractive index on the optical parameters results in the highest gradients with variations of the imaginary part. Consequently, knowledge of the black carbon fraction of atmospheric particles is needed, because of its highest imaginary part and therefore dominating role.

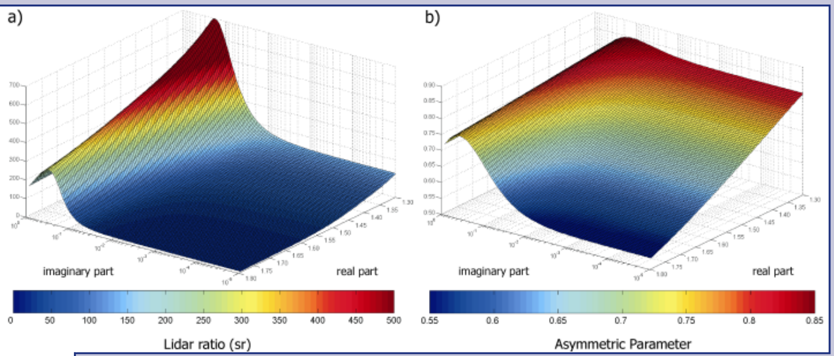

Fig. 3: Calculated Lidar ratio (left) and asymmetric parameter (right) as function of the refractive index.

Using a numerical correct and highly efficient source code from Mishchenko et al. (2002) to calculate optical properties the fitting of common distribution models to the measured data is necessary. Here, the fitting of a bimodal lognormal size distribution gives the best goodness of fit statistics. Consequently, the majority of measured size distributions shows two modes within the Aitken size range.

A comparison with Lidar measurements reveals a good agreement. This allows the application of the method within future closure studies to estimate optical properties of atmospheric particles.

Conclusions

- Correlations of particle size distributions with wind speed and NO_2 concentrations are size-independent; with temperature and SO_2 concentrations they are size-dependent.
- Estimating optical properties of particle size distributions gives results which are in agreement with literature data and own Lidar measurements.

Acknowledgements

Supported by the European Community Initiative INTERREG III A. We acknowledge the Bavarian Environmental Protection Agency for providing infrastructure and data.

UNIVERSITÄT BAYREUTH

LEHRSTUHL FÜR TECHNISCHE THERMODYNAMIK UND TRANSPORTPROZESSE
PROF. DR.-ING. D. BRÜGGEMANN