AN INSTRUMENT FOR STUDIES OF THE RELATION BETWEEN CLOUD DROPLET SIZE AND DRY RESIDUAL PARTICLE SIZE – THE DROPLET AEROSOL ANALYSER

Göran P. Frank and Bengt G. Martinsson
Department of Physics, Lund University, Lund, Sweden

1. INTRODUCTION

The Droplet Aerosol Analyser (DAA) is an instrument especially developed for studies of the interaction between aerosol particles, and cloud/fog droplets and interstitial particles. The DAA measures the ambient size of individual droplets and interstitial particles, the size of the dry residual particle after evaporation of the water vapour, and the number concentration of dry residual particles. This gives a unique three-parameter data-set (ambient diameter, dry residual particle diameter and number concentration).

Having access to these parameters, a number of related aerosol/cloud parameters can be determined:

- Number size distribution of ambient droplets/particles
- Number size distribution of dry residual particles
- The relation between ambient diameter and dry (residual) diameter on a single droplet/particle basis
- Characterisation of the droplet activation as defined by the Köhler equation
- The size dependent scavenging of particles due to activation
- Concentration of soluble matter in the individual droplets (solute concentration)
- Liquid water concentration

The DAA is based on a concept where the aerosol is processed in several steps by aerosol charging mechanisms, diffusion drying, and electrostatic aerosol spectrometry (using Differential Mobility Analysers, DMAs). The first version of the instrument (Martinsson, 1996; Cederfelt et al., 1997; Frank, 2001) was developed during the 1990-ies, and used in a number of ground based cloud and fog experiments (Martinsson et al., 1997; Frank et al., 1998; Martinsson et al., 1999; Martinsson et al., 2000). After a period of low activity, a second generation instrument is currently under development. The improved version will be more suited for long term measurements, which are favourable in order to obtain results with high statistical confidence. The time resolution of the instrument will be improved as well.

Here, we will present the features and capabilities of the DAA, by showing examples of previously obtained results. In addition, the technical details of the improved instrument will be described, and the planned scientific projects will be outlined.

2. FEATURES AND CAPABILITIES OF THE DAA

Results from a cloud event during the ground based cloud experiment at the mountain summit of Great Dun Fell in northern England 1995 (Bower et al., 1999; Martinsson et al., 1999) are used as examples. Average values of the event 1995-03-23, 00:00-05:00 (local time) can be seen in the figures. The total droplet number concentration was 1500 cm^{-3}, and the aerosol particle concentration (D_p>0.1 µm) was 2500 cm^{-3}. It must be emphasized that the results presented here are measured with the DAA only, and show the unique capabilities of the instrument.

Contact:
G. Frank: goran.frank@pixe.lth.se
B. Martinsson: bengt.martinsson@pixe.lth.se

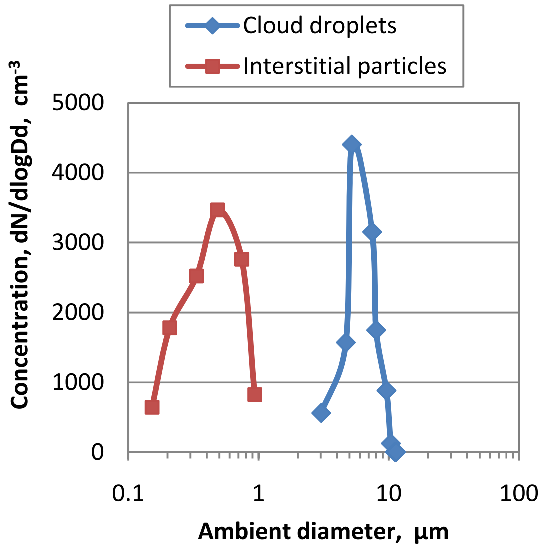

Figure 1. Number concentration of cloud droplets and interstitial particles

Figure 1 presents the ambient number size distribution of cloud droplets and interstitial particles. The separation between the two modes is clear.

In Figure 2, the dry (residual) aerosol particle number size distributions are presented for cloud droplet residuals, interstitial particle residuals and the sum of these (total). The total size distribution can be compared to a size distribution measured by a Differential/Scanning Mobility Particle Sizer, placed for example at an upwind station, in order to validate the results. This was indeed done, showing good agreement (Martinsson et al., 1999).

Figure 3 shows the cloud droplet scavenging ratio as a function of dry particle diameter, calculated from the results presented in Fig. 2, by dividing the cloud droplet residual concentrations by the total concentrations.

Figure 4 shows the relation between the ambient and dry residual diameters. In addition the critical diameter for activation according to the Köhler equation, as a function of dry diameter, is presented for particles consisting of ammonium sulphate and an insoluble core, for three different compositions. Although the particle chemical composition has been assumed, it can be concluded that cloud droplets smaller than 10 µm were activated, and that the largest droplets were possibly not activated.

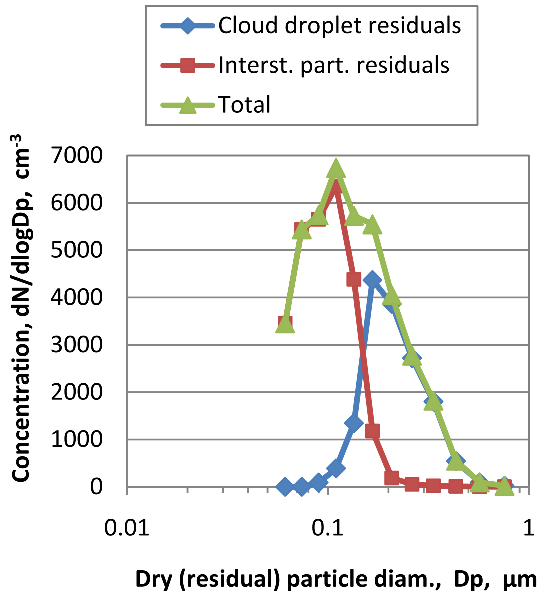

Figure 2. Number concentration of dry (residual) particles

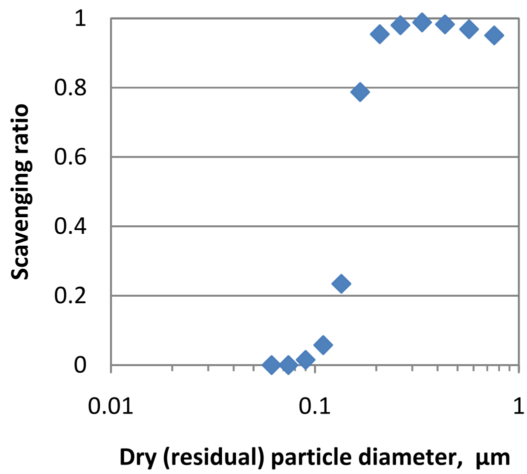

Figure 3. Cloud droplet scavenging ratio as a function of dry particle diameter

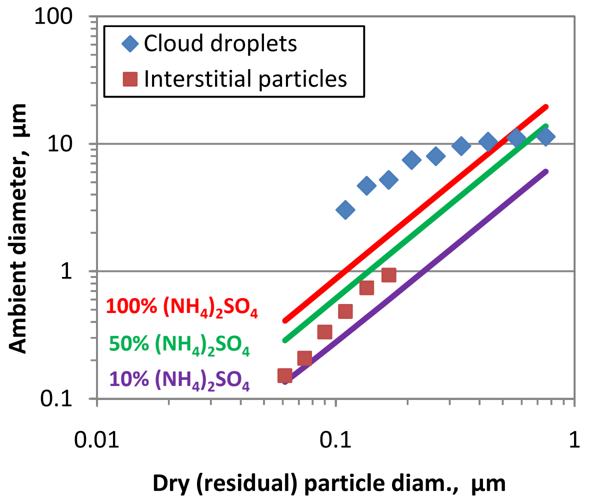

Figure 4. Relation between ambient and dry residual diameter, as well as critical diameter for activation. See details in the text.

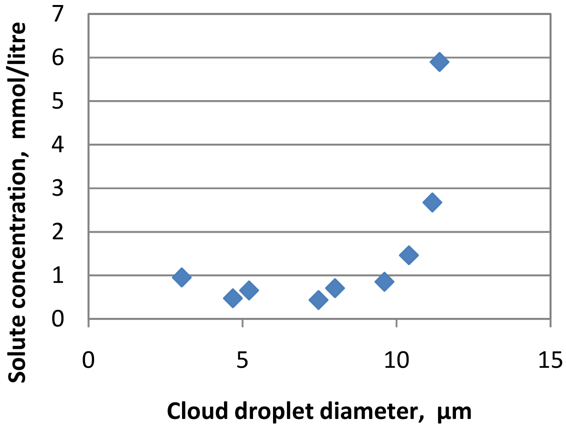

Figure 5. Solute concentration as a function of droplet diameter. See details in the text.

By measuring or estimating a chemical composition of the aerosol particles, the solute concentration of the cloud droplets can be calculated, since both the ambient and the dry diameters are known. In the example here, we have assumed the particles to consist of 50% ammonium sulphate (by volume) and the rest of an insoluble core. Figure 5 shows the solute concentration, and it can be seen that the dependence on ambient diameter is very strong for the largest droplets.

These were some examples of the results that can be obtained from DAA measurements. To summarize, the DAA instrument can give very detailed information about cloud microphysics, and the aerosol-cloud droplet relation. Together with complementary measurements, for example chemical composition of the aerosol particles, unique data-sets can be obtained, which can be used to improve the understanding of the aerosol-cloud relations.

3. INSTRUMENT PRINCIPLE

Below follows a description of the principle of the DAA, see also Figure 6. The theoretical background of the instrument can be found in Martinsson (1996). A detailed description of the first version of the instrument and a field inter-comparison with respect to six aerosol and cloud characteristics can be found in Cederfelt et al. (1997).

1. *Air inlet.* The inlet collects droplets and interstitial particles. A vane directs the inlet into the wind. The DAA can characterise droplets up to 20 µm, possibly 30 µm in diameter (the upper limit has not been exactly determined). To avoid sedimentation losses the instrument employs vertical flows as much as possible, and there is a bend after the inlet.

2. *Bipolar charging unit.* After the bend, the aerosol particles are charged in a bipolar charger, to give a well defined bipolar charge state and a low number of charges on the individual droplets and particles, when they enter the unipolar charger.

3. *Unipolar charging unit*. In the unipolar charger the droplets and particles, still at their original size, are positively charged. The number of charges acquired by the droplets and particles is

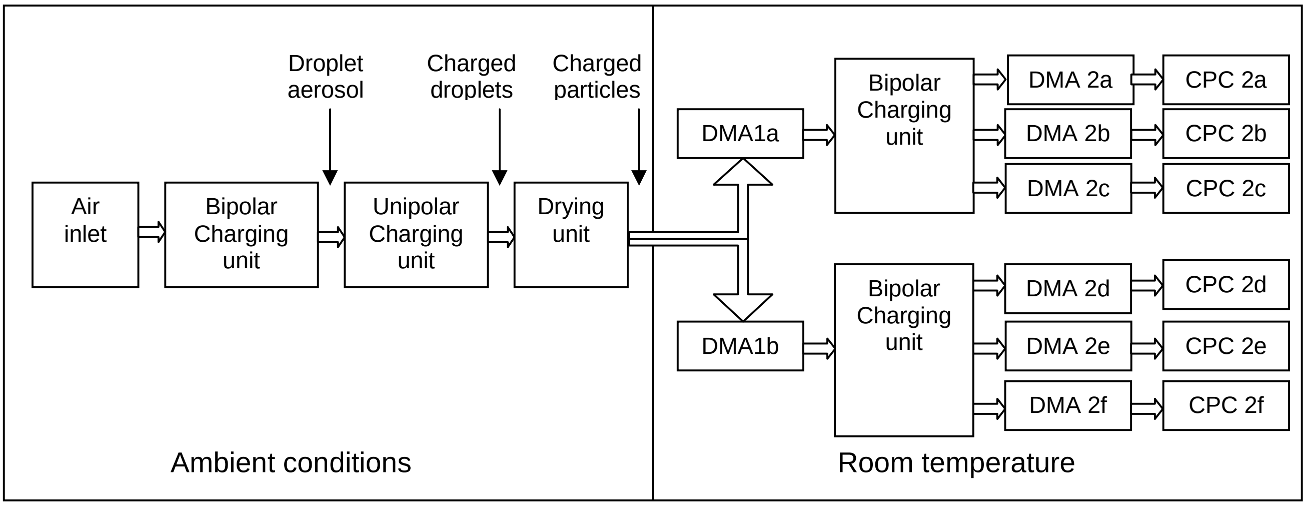

Figure 6. ***The principle of the DAA and the basic units.***

dependent on their size (larger droplets will acquire a higher charge) and the relationship is determined by a calibration. The charging process is a combination of diffusion charging and field charging. Ions are produced with a radioactive α-source and transported to the charging region by an electric field. The development and calibration of the charger used in the DAA are described in Frank et al. (2004).

4. *Drying unit.* The sample flow then passes through a diffusion drier that removes the water. The result is highly charged dry residual particles. Some volatile matter, e.g. HNO_3, may also be removed in the drier.

The aerosol is then transported from the part at ambient conditions (the outdoor part) to the indoor part, where the size, number and charge level of the residue particles are measured. The aerosol is divided into two lines in order to optimize the scanning performance and shorten the time required.

5. *DMA 1.* In the first DMAs, the electrical mobilities of the residual particles are determined. The DMAs are stepping over the interesting ranges.

6. *Bipolar charging unit.* This unit recharges the particles to bipolar charge equilibrium (low number of charges on the individual particles). Bipolar charging is carried out in order to measure the dry size of the residual particles in DMA 2a-f, as in normal electrostatic classification.

7. *DMA 2a-f.* The six DMAs work in parallel, and each of them is set to measure a fixed dry particle diameter. For every voltage step of DMA 1, the number of particles is counted after each DMA 2. By knowing the dry diameter from DMA2, and the electrical mobility from DMA1, the number of charges on each individual cloud droplet can be calculated. The original ambient size can then be obtained via the calibration of the unipolar charger.

8. *CPC 2a-f.* After each DMA2 there are particle counters (CPC = condensation particle counter) to count the number of particles.

The dry particle sizes measured are chosen in order to cover the region where particles are scavenged by droplet formation, i.e. the chosen sizes should be from the smallest particles that do not act as cloud condensation nuclei (CCN), up to particle sizes where the major part of the particles act as CCN. In most cases the number concentration decreases with increasing dry particle size from about 0.2 µm in diameter and the upper dry particle size limit should preferably be where the particle number concentration is negligible.

The six dry particle sizes measured simultaneously can be set to cover the whole region of interest, and one scan with DMA 1 takes ~5 minutes, depending on the particle concentration in the air. The six dry particle sizes are then changed to six new sizes that lie in between the first six, and a new scan is performed with the first DMA, giving a total scan time of 10 minutes. However, the scans can be used independently and the time resolution obtained is thus 5 min.

4. UPCOMING PROJECT

After finishing the instrument development, long-term measurements at the summit of the mountain Brocken (51.80º N, 10.67º E, 1142 m asl) in the Harz region in central Germany will be be performed. The project is in collaboration with the Air chemistry group of the Technical University of Brandenburg (BTU Cottbus), who has a cloud measurement site at mount Brocken since many years. Measurements during a longer time period will provide the possibility for a variety of air masses with different aerosol properties to reach the site, such as more marine type air coming from the west, northwest; relatively freshly polluted air coming from the industrialised regions in Germany and central Europe, and aged polluted air coming from Eastern Europe. Different cloud types with different dynamical properties will immerse at the site, such as orographically induced clouds (high updraught velocity) and stratiform clouds (low updraught velocity). The long-term measurements will lead to many events with several combinations of different aerosol properties, different air masses, and different cloud types. These events will be classified and the results for each main combination can thus be described with high statistical confidence. Focus will be on warm clouds, since the available instrumentation cannot be used for studies of ice or super cooled water clouds.

Descriptions of the aerosol-cloud relation will then be derived, and used for validations of model results, parameterisations, etc. One major goal of the project is to improve existing and/or to develop new aerosol-cloud parameterisations.

5. OUTLOOK

After gaining experience of the improved version of the instrument, and from the first scientific project with the new instrument, the next step would be to study other cloud types at other locations. We have been thinking of both long-term ground based measurements, as well as aircraft measurements. The instrument probably has to be further improved for aircraft measurements, to lower the weight and, if possible, improve the time resolution, but we are confident that this is doable. A DAA version for aircraft measurements would provide an excellent tool for aerosol-cloud interaction studies.

We are interested in future collaborations. Suggestions are most welcome.

REFERENCES

Bower et al., Atmos. Res. 50, 151-184, 1999
Cederfelt et al., Atmos. Environ. 31, 2657-2670, 1997
Frank et al., Contr. Atmos. Phys. 71, 65-85, 1998
Frank, PhD thesis, http://nuclearphysics.pixe.lth.se/theses/gf.asp, 2001
Frank et al., J. Aerosol Sci.,35, 117-134, 2004
Martinsson, J. Aerosol Sci., 27, 997, 1996
Martinsson et al., Atmos. Environ. 31, 2477-2490, 1997
Martinsson et al., Atmos. Res. 50, 289-315, 1999
Martinsson et al., Tellus 52B, 801-814, 2000