HYGROSCOPIC GROWTH AND CCN ACTIVITY OF AEROSOL PARTICLES DURING THE 3RD PALLAS CLOUD EXPERIMENT

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INTRODUCTION

An aerosol particle's response to a change in humidity is a function of its size and chemical composition. Soluble particles take on water and grow with increasing humidity, while particles composed of hydrophobic material do not. Aerosol particles become climatically important if they are able to grow to sizes of 50 nm and larger. Particles in this size range can act as cloud condensation nuclei (CCN) and therefore may contribute to the indirect aerosol effect, a series of proposed impacts that include increased cloud albedo due to increases in cloud droplet concentrations (Twomey, 1991). Once particles grow to a size where they can become CCN, their ability to activate into cloud droplets depends on their chemical composition, particle number concentration, and the water supersaturation of the surrounding air parcel. Because both aerosols and clouds have a substantial influence on the climate, the fundamental purpose in looking at aerosol-cloud interactions is to find a relation between the activating aerosol particle and the resulting cloud droplet population.

METHODS

The third Pallas Cloud Experiment (PaCE III), an intensive campaign measuring aerosol and cloud properties, was conducted by the Finnish Meteorological Institute and University of Eastern Finland at the Pallas-Sodankylä Global Atmospheric Watch (GAW) station (Hatakka et al., 2003). This station consists of several different sites, but for this work we concentrate only on the main measurement site Sammaltunturi (67°58′N, 24°07′E, 560 m a.s.l., in Muonio, Finland). The Sammaltunturi measurement site is located on top of a fell, which rises about 300 m above the surrounding area thus being a very suitable site for ground-based aerosol-cloud interaction measurements. In this study a 14-day (21.9.2009–4.10.2009) set of Cloud Condensation Nuclei Counter (CCNC), Hygroscopic Tandem Differential Mobility Analyzer (HTDMA) and Differential Mobility Particle Sizer (DMPS) data has been analyzed to investigate the hygroscopicity and cloud-forming properties of aerosols in this remote continental site.

The DMT CCNC measures the concentration of CCN at a given water vapor supersaturation (SS). Details concerning the characteristics of the DMT CCNC can be found in Roberts and Nenes (2005). In this study we selected five SS values, SS=0.2, 0.4, 0.6, 0.8 and 1.0%, and compared the concentration of activated particles to the total number concentration measured by a Condensation Particle Counter (CPC, TSI 3772) in order to get the average activated fraction (CCN/CN). The HTDMA was running on eight different dry diameters (Dd= 15, 25, 35, 50, 75, 110, 165 and 265nm), with 90% relative humidity. To get the total number size distribution between 7 and 500nm, a DMPS system was deployed. In addition, single hygroscopicity parameter calculations were performed by applying the κ-Köhler-theory (Petters and Kreidenweis, 2007) to the HTDMA and CCN counter data.
RESULTS AND DISCUSSION

Average activation ratio (CCN/CN) ranged from 0 to ~65% depending on the supersaturation of the instrument and most likely on size, origin and composition of aerosol particles. The activated fraction was significantly higher during the first three days of the campaign concurrent with a high total number concentration of particles. Predictably, CCN counts were found to be higher with greater SS values, only on a couple of occasions the average activated fraction of a lower SS value exceeds the higher SSs activated fraction. This is due to the somewhat long SS scanning-time (15 min/SS) of the instrument.

κ-Köhler-theory can be used to predict particle water content in the subsaturated (S<1) regime, as well as to predict the conditions for cloud droplet activation. The hygroscopicity parameter, κ, has an upper limit of ~1.4 for the most hygroscopic species typically found in the atmospheric aerosol (e.g. sodium chloride). Lower values of κ then indicate less-hygroscopic, or less CCN-active, behavior. The time evolution of κ, estimated from the PaCE III CCNC data, is shown in Fig. 1. Here, κ was calculated by the following approximate expression (Petters and Kreidenweis, 2007):

\[ \kappa_{CCN} = \frac{4A}{27D_d^3 \ln S_c}, A = \frac{4\sigma_s M_w}{RT\rho_w}, \]

where \( D_d \) is the critical diameter of the particle and \( S_c \) is the critical supersaturation. The smaller the SS is, the larger the κ value becomes. For example SS=0.2%, κ values are remarkably higher than for the other supersaturations. This indicates that these particles are most likely more aged and larger resulting in higher hygroscopicity. During 21-23 September, κ values did not appear to vary significantly with supersaturation. This can be explained by the fact that on 21.9.2009, at about 15:00, a strong new particle formation event (\( N_{tot,max} \sim 5000 \text{ cm}^{-3} \)) was observed. The growth of the particles lasted for ~24 hours, which was followed by another weaker new particle formation event.

\[ \kappa_{HTDMA} = \frac{GF^3 - 1}{RH} \exp\left( \frac{4\sigma_w M_w}{RT\rho_w D_d GF} \right) + 1 - GF^3. \]

Figure 1. Average κ values derived from the CCNC data for five supersaturations (SS) over the whole study period.

The mean growth factors measured by the HTDMA were also used to derive the respective κ values. The hygroscopicity parameter can be derived from the measured HTDMA growth factor (GF) as follows (Good et al., 2009):
Figure 2 shows the time evolution of $\kappa_{\text{HTDMA}}$. Note that only the five dry diameters with the best data quantity were chosen for this plot. As expected, larger dry diameters of the particles result in a higher $\kappa$ value, however the difference between the dry size dependent $\kappa$ values is not as great as with the SS dependent $\kappa$ values.

![Graph showing time evolution of $\kappa_{\text{HTDMA}}$](image)

Figure 2. Average $\kappa$ values derived from the HTDMA data for five dry diameters ($D_d$) over the whole study period.

Fig. 3 compares the $\kappa$ values calculated from the HTDMA and CCNC data. Only two curves were selected to show consistency between these two instrument’s abilities to estimate the hygroscopicity of the aerosol particles. Selection was performed by estimating the critical diameter of the ambient aerosol according to the Köhler-theory, i.e. SS=0.2% $\sim$ $D_d$=165nm and SS=0.6% $\sim$ $D_d$=50nm. As shown in Fig. 3, CCNC derived $\kappa$ tends to be higher than the HTDMA derived, but overall, the agreement between these two instruments is quite good.

CONCLUSION

In future analyses, we shall analyze other measured data (such as station meteorology, aerosol chemical composition, and aerosol number size distribution) and combine it with CCN data to investigate how the size dependent composition of particles varies during the day, and how the estimated diameter of smallest activated particles depend on the composition of particles. In addition, similar $\kappa$-Köhler-theory estimations will be applied to the Aerodyne High-Resolution Time-Of-Flight Aerosol Mass Spectrometer (HR-TOF-AMS) data to get more detailed information about the applicability $\kappa$-Köhler-theory in this remote continental site.
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REFERENCES


