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**Hygroscopic Properties of Aerosol Particles
in the Temperature Range $-11^{\circ}\text{C} < T < 20^{\circ}\text{C}$**

HYGROSCOPIC PROPERTIES OF AEROSOL PARTICLES IN THE TEMPERATURE RANGE $-11^{\circ}\text{C} < T < 20^{\circ}\text{C}$

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BACKGROUND

The ability of hygroscopic atmospheric aerosols to absorb water is of great importance. A change in relative humidity (RH) may significantly change the size of aerosol particles which in turn affects their light scattering efficiency, and influences aqueous phase reactions on the aerosol particle surface (heterogeneous chemical reactions). In the field, hygroscopicity measurements are commonly performed using a Hygroscopicity Tandem Differential Mobility Analyzer (H-TDMA). Recent investigations (McMurry and Stolzenburg, 1989; Svenningsson *et al.*, 1992, 1994; Zhang *et al.*, 1993; Covert and Heinzenberg, 1993; Pitchford and McMurry, 1994) have observed two distinctively different types of particles: so-called "less" and "more" hygroscopic particles with diameter growth factors of ≤ 1.1 and $1.3 - 1.5$, respectively, at RH = 80 - 90 %. Most of these measurements were performed at ambient temperatures ($20 - 30^{\circ}\text{C}$).

The aim of this study is to measure aerosol hygroscopic properties at remote locations and to compare the results with polluted air masses. The high-alpine site Jungfraujoch at 3580 m asl in Switzerland is an ideal place for such studies because this site is often exposed to the planetary boundary layer as well as to the free troposphere. Average ambient temperatures at the station are low (-1°C in summer and -14°C in winter). At these temperatures, semi-volatile compounds (such as nitrate or lower-molecular-weight organics) may be adsorbed and considerably alter aerosol hygroscopic properties. To minimize artefacts due to volatilization, aerosol hygroscopic behavior should be measured at ambient conditions.

INSTRUMENTAL SETUP

Figure 1 shows the experimental setup of the H-TDMA. Briefly, the aerosol is dried with a diffusion dryer and fed into the first DMA where particles of a certain "dry" size (frost point temperature $< -22^{\circ}\text{C}$) are selected. After exposure to higher RH, the new particle diameter is determined with a second DMA combined with a condensation particle counter (CPC). To ensure a constant temperature within the entire H-TDMA system, both DMAs, as well as the humidifier, are submersed in a water/ethylene glycol bath which can be cooled down to -12°C . RH is derived by measuring temperature and dew/frost point temperature with an accuracy of 0.1°C . The humidifier works without warming-up the gas-stream, hence ensuring that the aerosol composition is unaltered. Both DMAs are operated using a re-circulating or "closed-loop" sheath air arrangement, which ensures that air properties within the instrumentation are similar to those under ambient conditions.

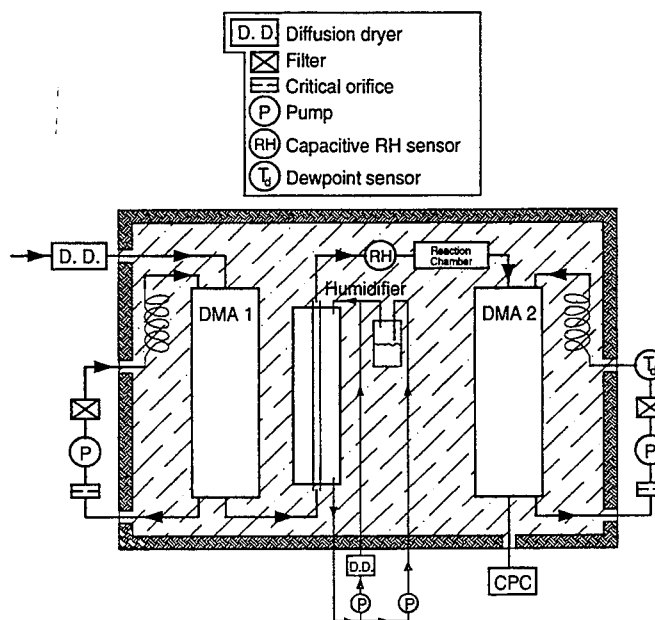


Figure 1: Sketch of the experimental setup.

FIRST RESULTS

The H-TDMA was tested with NaCl aerosol which was generated by atomization of an aqueous solution. Measurements were performed at 20°C, 0.5°C and -9°C and RH was calculated with respect to water. Figure 2 shows that NaCl particles are characterized by an abrupt increase in diameter at the deliquescence humidity (DH), which is due to the phase transition of particles changing from "dry" crystals to saturated solution droplets.

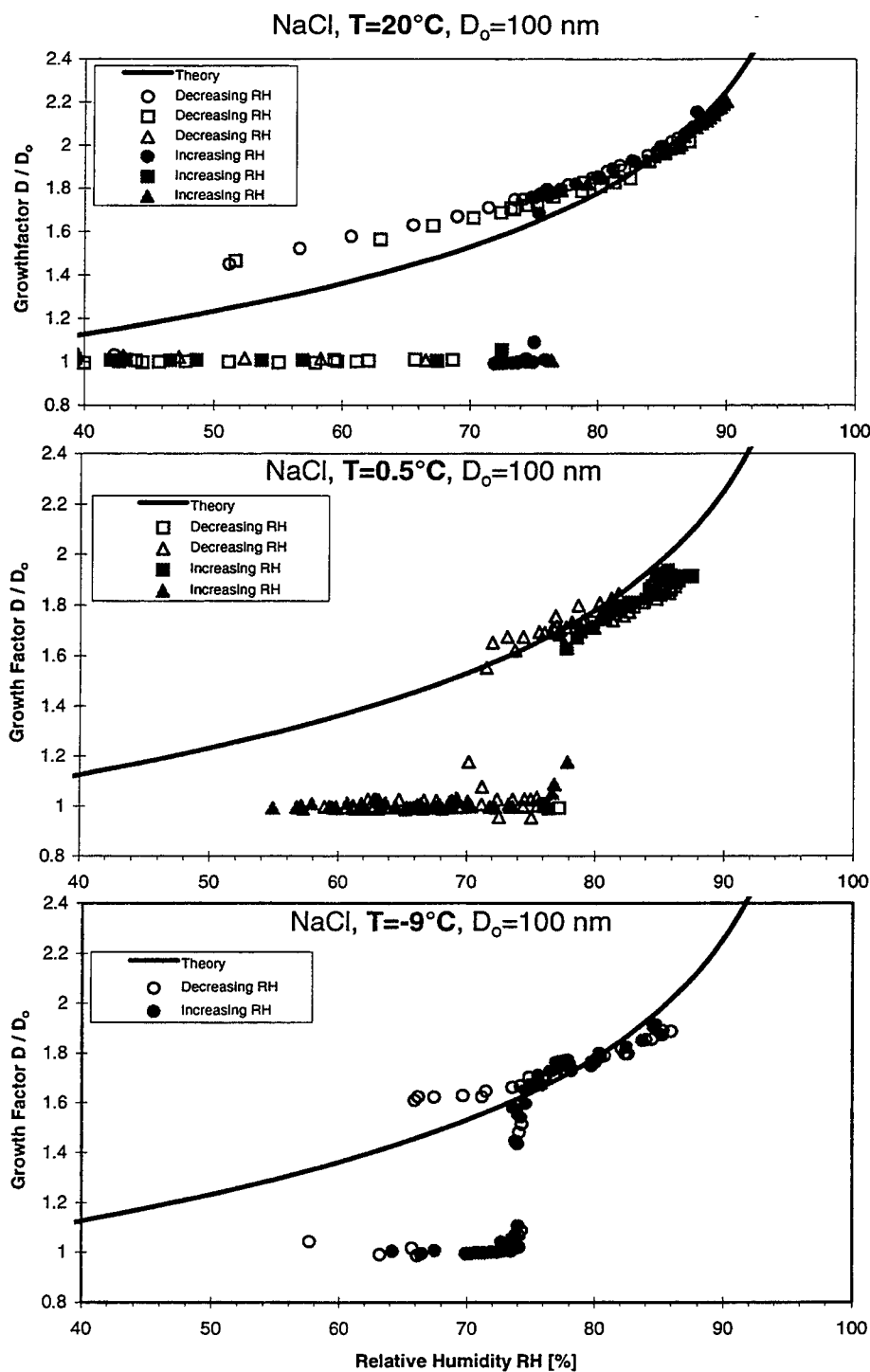


Figure 2: Humidograms of NaCl test aerosol at different temperatures. The diameters were calculated by fitting the size distributions measured by the second DMA with a sum of lognormal functions. Since a fraction of the aerosol particles is temporarily subjected to higher RH in the humidifier, more than one diameter is often found for a fixed RH. Solid lines are theoretical curves calculated with a simplified Köhler theory.

A few milliseconds after exposure to higher RH the solution droplets are in new equilibrium with water pressure. The diameter of the droplet is described by the Köhler theory (solid lines in the Figures) which applies only to sufficiently diluted droplets. For 20°C and 0.5°C, DH is about 75% and agrees well with measurements reported by other investigators (see Seinfeld and Pandis, 1998; and references therein). For all presented temperatures a good agreement with Köhler theory is found. The agreement at $T = -9^\circ\text{C}$ supports the idea that NaCl particles remain liquid at this temperature.

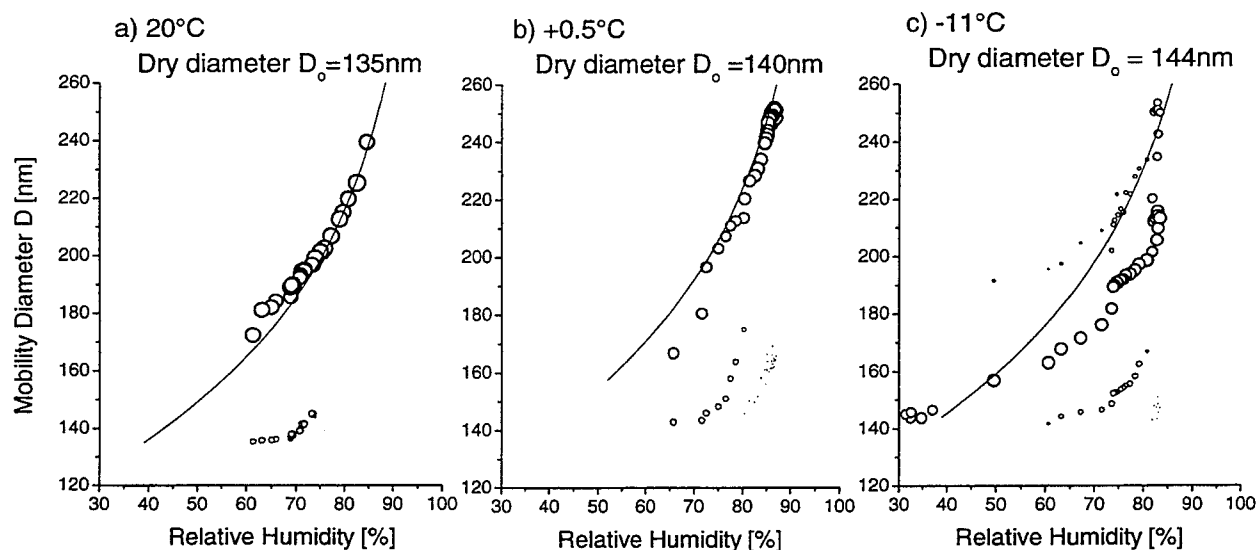


Figure 3: Humidograms of NaNO_3 test aerosol at different temperatures. The area of the points is proportional to the number concentration in each mode.

Figure 3 shows the measurements of NaNO_3 particles. For $T > 0^\circ\text{C}$ a good agreement is found between experimental and predicted theoretical data. DH is found to be 76 and 80% for 20°C and 0.5°C, respectively. In contrast to NaCl particles, the measured humidogram of NaNO_3 particles is different at $T = -11^\circ\text{C}$. For a fixed RH three different modal diameters are measured indicating another phase transition, which is currently the subject of further research.

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