Is the aerosol particles hygroscopicity only surface-driven ?

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INTRODUCTION

In recent years, much attention has been directed towards understanding the effect of aerosols on a variety of processes in the earth's atmosphere. Atmospheric aerosol particles play an important, but still poorly understood role in cloud formation. The Kohler theory is often used to describe the critical super saturation at which a Cloud Condensation Nuclei (CCN) of known size is activated, i.e. grows into a cloud droplet.

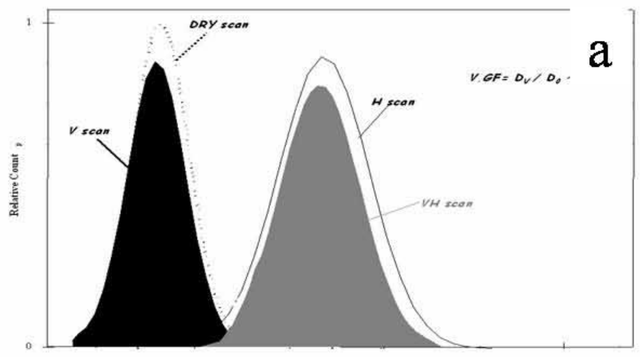
Theoretical calculations of sub-saturation growth mainly involves the solubility of the particle bulk, however it is not yet known how the surface of the particles influences their growth rate when they are exposed to a humid environment.

METHODS

A novel instrument based on a Tandem Differential Mobility Analyser (TDMA) combining volatilisation and humidification conditioning of particles was developed to test the effect of surface properties on their hygroscopic growth, and it has been named **A**erosol **S**urface and Internal **A**nalyser (ASIA).

The system can be broken down into three main steps (Figure 1): **1**-particle selection, **2**-particle conditioning and **3**-measurement of modified particle number size distribution.

In order to study the role of the surface of these particles, the volatilisation should be gentle enough so that the size of the heated particles do not change significantly. A complete characterization of a particle of a specific diameter (Dp) consists in first measuring the change obtained after slight heating (close to T=100°C) (Volatility-Scan), then the size change due to exposure to a humid flow (RH=90%) (Humidity-Scan), and finally the size change due to heating followed by humidifying (Volatility-Humidity-Scan). In Figure 2 a,b,c are summarised three possible results from ASIA samples.



Case a: The hygroscopicity of the surface and the core are similar.

Case b: After heating, the particle surface layer is removed or decomposed and reveals a less hygroscopic mode.

Case c: When the surface is modified a new class of more hygroscopic cores appears. The explanation can be illustrated Figure 3.

Figure 2 (a, b and c) Schematics of the size distributions obtained after a particle size selection (dry), a heating treatment (V-scan), a humidifying treatment (H-scan) and a heating followed by humidifying treatment (VH-scan).

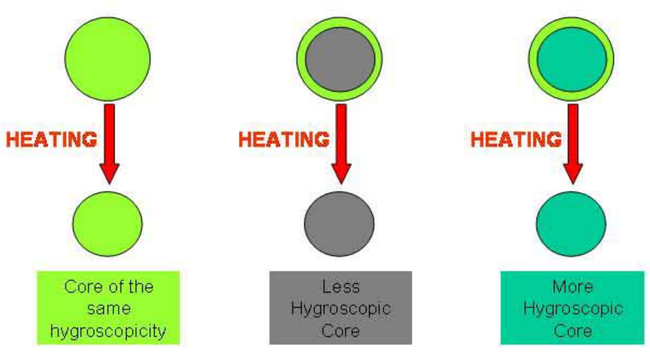


Figure 3. Illustration of the possible explanation of the apparition of a less or more hygroscopic property after heating.

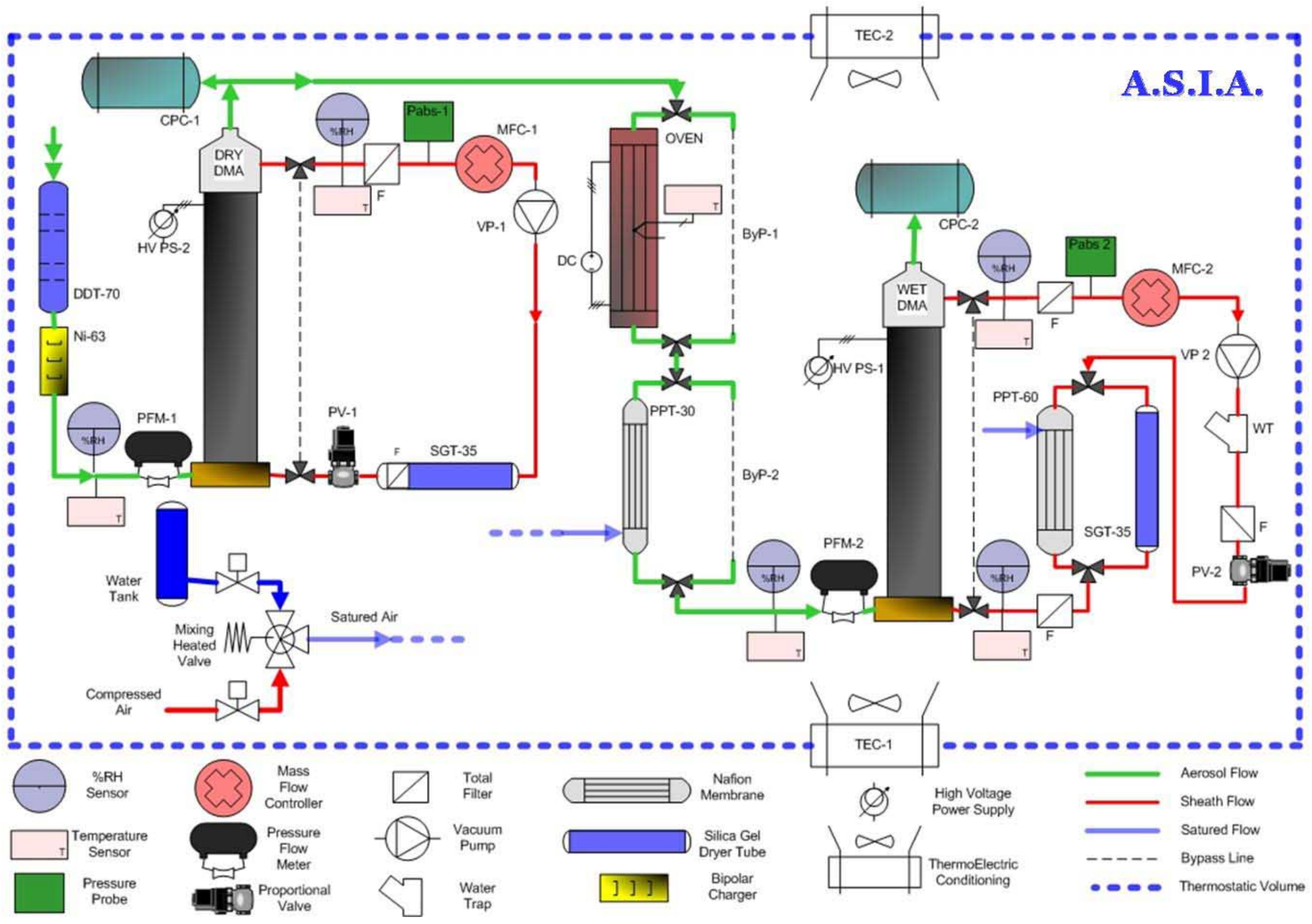


Figure 1. Schematic of the Volatility & Hygroscopicity TDMA.

MEASUREMENTS

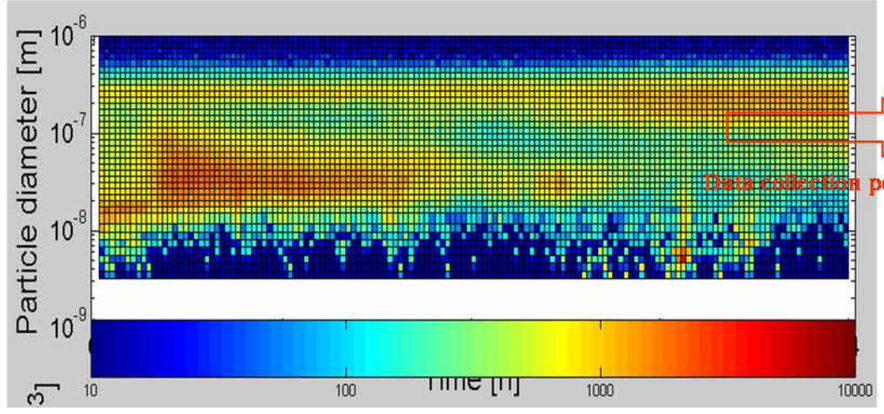


Figure 4. Time evolution of the aerosol size distribution in Hyytiälä on 21th April 2005 (Data acquainted from continuous measurements performed at the SMEAR Station, M. Kulmala)

Results presented in the present work were performed during the BACCI/QUEST campaign in April 2005 at Hyytiälä, Finland. Figure 4 shows the particle size distribution as a function of time during 21th of April obtained from the DMPS of the Research station in Hyytiälä forest.

The examples represented on Figure 5 are relative to a series of hygroscopicity measurement that took place at the SMEAR II Station with ASIA between 21:00 on 21th to 03:00 on 22th of April 2005 with regular intervals.

In these data both volatility and hygroscopicity distributions of 100 nm particles are not changing significantly during sampling. **The "H scan" changes only when the surface is modified.**

Relative increases or decreases in H-GF following thermal conditioning are measurable on several occasions during the campaign for a wide range of particle diameter.

Because the working temperatures are generally less than or equal to 100°C, we can hypothesise that the volatility scan leads to the evaporation of semi-volatile organic material present onto the particle surface. The removal of this surface coating has very little effect on either the size or the mass of the particle but leads to significant change in its H-GF.

The conditions leading to higher or lower hygroscopic growth are still under investigation.. This demonstrates, however, that prediction of hygroscopic growth based on bulk aerosol composition may lead to erroneous results.

These results indicate that surface properties are not sufficiently accounted for by the Kohler theory. Additional studies on the particle surface properties are required to better predict CCN concentrations.

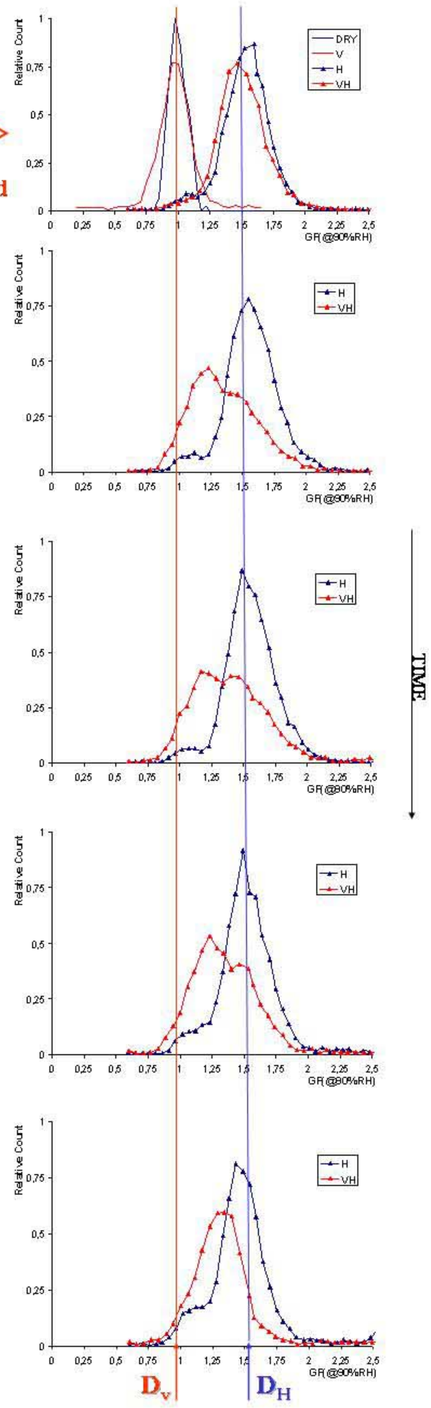


Figure 5. Core and surface hygroscopic time evolution for 100 nm particles after a heating treatment at 90 ° C.

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