Journal of Aerosol Science 101 (2016) 1–9

Contents lists available at ScienceDirect

Journal of Aerosol Science

journal homepage: www.elsevier.com/locate/jaerosci

Relative humidity non-uniformities in Hygroscopic Tandem Differential Mobility Analyzer measurements

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ARTICLE INFO

Article history:
Received 30 December 2015
Received in revised form
4 July 2016
Accepted 5 July 2016
Available online 12 July 2016

Keywords:
Aerosol Particle Hygroscopicity
HTDMA
Particle Tracking Model

ABSTRACT

Hygroscopic Tandem Differential Mobility Analyzers (HTDMAs) are widely used to measure the water uptake characteristics of aerosol particles. As has been shown experimentally in the past, potential differences in the relative humidity (RH) between the aerosol and the sheath flow of the second Differential Mobility Analyzer (DMA) can lead to erroneous estimates of the apparent hygroscopic behavior of the sampled particles. A prompt phase transition, for example, may become smeared and be erroneously interpreted as non-prompt. Using a particle-tracking model, here we simulate the trajectories and the state of the particles classified in a DMA with non-uniform RH and temperature profiles. Our simulations corroborate earlier observations proving that such an experimental artifact can induce particle growth within the second DMA. Given the importance of maintaining uniform RH and temperature inside the second DMA of HTDMA systems and the limitations of existing RH and temperature sensors, we further provide suggestions for their operation.

1. Introduction

The extent to which atmospheric particles can take up water (i.e., their hygroscopicity) affects their optical behavior and in turn the radiative properties of the atmosphere (Haywood & Boucher, 2000; Ogren & Charlson, 1992). In addition, hygroscopicity can influence the health effects of inhaled particles by changing their size and therefore their deposition patterns in the human respiratory system (Chen & Lee, 1999). Knowledge of the hygroscopicity of airborne particles is thus of particular importance for determining their environmental impacts (Anastasio & Martin, 2001).

Many techniques have been employed to measure the hygroscopic properties of aerosol particles (cf. McMurry, 2000). Electrodynamic Balances (EDBs; Peng & Chan, 2001) have been developed and used to measure changes in the mass of micron-sized particles as a function of relative humidity (RH). The water uptake characteristics of smaller particles can be probed by Hygroscopic Tandem Differential Mobility Analyzers (HTDMAs; Rader & McMurry, 1986), which can measure

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http://dx.doi.org/10.1016/j.jaerosci.2016.07.004
0021-8502/

changes in the electrical mobility of the particles with high precision when exposed to different conditions. Apart from the amount of water that the particles can take up at any given RH, these systems can also determine the deliquescence and the efflorescence RH (DRH and ERH, respectively; Martin, 2000) of the sampled particles (e.g., Biskos, Russel, Buseck & Martin, 2006a, 2006b; Mifflin, Smith & Martin, 2009; Cheng, Su, Koop, Mikhailov & Pöschl, 2015).

The hygroscopic properties of inorganic aerosol particles having diameters larger than 50 nm have been determined in several laboratory studies (e.g., Tang, Munkelwitz & Davis, 1977, 1978; Tang and Munkelwitz, 1993; Tang, Tridico & Fung, 1997). In these studies, the observed phase transitions were prompt (i.e., spontaneous), while the measured hygroscopic growth factors agreed within experimental uncertainty with theoretical predictions. Hämeri, Väkevä, Hanson, and Laaksonen (2000), who were the first to measure the hygroscopic properties of sub-50-nm $(NH_4)_2SO_4$ particles using an HTDMA, reported that these did not spontaneously transform from the solid/dry to the aqueous solution state but instead exhibited a gradual increase of their size when exposed to increasing RHs close to their DRH value. Mirabel, Reiss, and Bowles (2000) provided a theoretical explanation of the non-prompt deliquescence of inorganic salt nanoparticles by assuming coexistence of the solid with the aqueous phase during the transition. The non-prompt particle deliquescence, however, did not agree with predictions by thorough theoretical models developed later by Djikaev et al. (2001), Russell and Ming (2002), and Topping, McFiggans, and Coe (2005). In view of this disagreement, Biskos et al. (2006b), Park, Kim, and Miller (2009), Hu et al. (2010) and Cheng et al. (2015) provided new sets of HTDMA measurements with non-agglomerated sub-50-nm $(NH_4)_2SO_4$ particles, which in contrast to the observations reported by Hämeri et al. (2000) showed that their phase transitions are prompt. The main difference between the measurements mentioned above and those conducted by Hämeri et al. (2000), was that the RH of the monodisperse aerosol and that of the sheath flow of the second DMA were matched in order to ensure that the RH conditions were fixed along the path of the particles downstream the humidifier. Although matching the RH of these two streams is important, potential temperature non-uniformities along the path of the particles during classification can also induce RH variability, resulting in an apparent non-prompt particle deliquescence, even for $(NH_4)_2SO_4$ particles larger than 50 nm as suggested by Duplissy et al. (2009).

Here, we investigate the effects of RH and temperature non-uniformities in HTDMA measurements. To do so we calculate temperature and RH profiles within the second DMA the HTDMA and use a particle-tracking model, coupled with a module for predicting the phase (solid or droplet) and size of the particles along their migration paths within the DMA. The results of the simulations are compared with experimental observations reported by Biskos et al. (2006b) using $(NH_4)_2SO_4$ particles.

2. The Hygroscopic Tandem DMA

Figure 1 shows the main components of the HTDMA system used to collect the data reported by Biskos et al. (2006b). It consisted of two Differential Mobility Analyzers (DMAs) for sizing particles in the nanosize range (Chen et al., 1996), a humidification system, and an Ultrafine Condensation Particle Counter (UCPC; Stolzenburg & McMurry, 1991). Particles produced by atomization were initially dried and passed through a bipolar charger before entering the first DMA (DMA-1). The monodisperse aerosol flow downstream DMA-1 was then exposed to elevated RH conditions inside the humidifier. The second DMA (DMA-2), which was also operated with a sheath flow of elevated RH, and the UCPC were used for measuring the size distribution of the particles downstream the humidifier. Consequently, any changes in the size of the particles caused by water uptake could be detected and quantified (cf. Sections 3.1 and 3.2). It should be noted that the RH values of the aerosol and the sheath flow were measured upstream of DMA-2. More details on the experimental setup and the procedures followed are available in Biskos et al. (2006b).

To avoid changes in particle size due to water evaporation or condensation during the measurements, the RH of the aerosol should remain constant downstream the aerosol humidifier. Without special care, however, the RH of the aerosol (RH_a) and that of the sheath (RH_{sh}) can become mismatched, yielding a non-uniform RH profile within DMA-2. RH non-uniformities can also be caused by temperature variations within the second DMA. To minimize non-uniformities in the

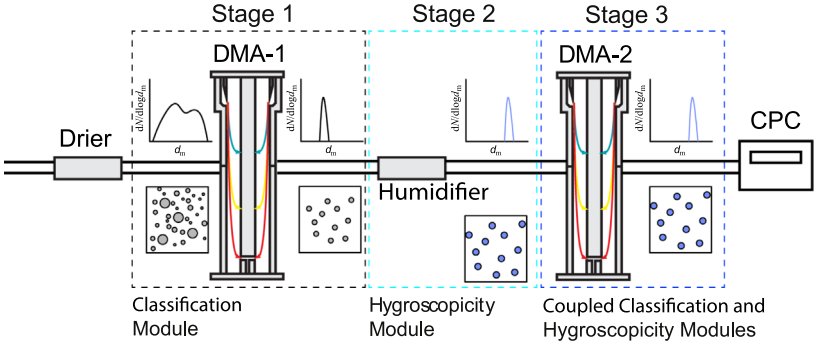

Fig. 1. Schematic layout of the HTDMA system and the associated modules of the numerical model. Examples of particle size distributions at each stage are provided for particles of uniform chemical composition (i.e., an internally mixed aerosol).

Please cite this article as: Bezantakos, S., et al. Relative humidity non-uniformities in Hygroscopic Tandem Differential Mobility Analyzer measurements. *Journal of Aerosol Science* (2016), http://dx.doi.org/10.1016/j.jaerosci.2016.07.004