Atmospheric Measurement Techniques Discussions

Atmos. Meas. Tech. Discuss., referee comment RC2
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Comment on amt-2021-108

Anonymous Referee #2

Referee comment on "A dual-droplet approach for measuring the hygroscopicity of aqueous aerosol" by Jack M. Choczynski et al., Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2021-108-RC2, 2021

The authors demonstrate a dual-droplet approach for hygroscopicity measurements of micron size aerosol droplets in a linear quadrupole electrodynamic balance. One of the droplets serves a relative humidity sensor; the other is the particle under study. The authors use a spectroscopic method (Mie resonance spectroscopy) for measuring size and refractive index of the particles. They need to switch the two particles alternatively into the measuring volume by adjusting the DC voltage compensating the gravitational force. They illustrate the method by showing several examples of hygroscopicity for different aqueous model systems and show that the accuracy in RH reaches 0.5% at 50% RH and better than 0.1 % at 90% RH. This is significantly better than what can be achieved with traditional RH sensors in electrodynamic balances.

The paper is well written, the topic timely and of interest for the readers of AMT and I recommend publishing but ask the authors to take the following comments and suggestions into account.

The main weakness of the presented approach is in my opinion that the technique allows measuring the size growth factor but not directly the mass growth factor. The main advantage of the electrodynamic balance technique is that the inverse of the mass growth factor is the concentration (mass fraction of solute) of the aqueous droplet as long as only water partitions between gas and condensed phase in the experiment. As the authors explain, going from size growth to concentration requires knowledge of the density, which is typically not available for unknown samples. This makes the method less attractive for practical applications where no independent density data exist.

Even more restrictive may be any application to multicomponent particles in which one of the compounds has low solubility and hence the droplet spherical symmetry is no longer conserved. This will make sizing with Mie resonance spectroscopy difficult if not impossible. In their paper describing the setup in detail (Davies, 2019) they make use of the electrostatic force between two particles in the balance to calculate the characteristic constant of the EDB and measure density of a particle by a combination of the optical measurement and the DC voltage to compensate gravitational force. Could this be applied to the two-droplet method here? Please add a discussion.

Minor comments:

In Section 3.1. a more extended discussion on why there is an upper limit in RH measurements because of sizing problems for large particles would be helpful.

Lines 265-274: Lienhard et al. (2012) provide experimental density data and refractive index data for citric acid. May be you could compare with those? Fig. 4 would provide an opportunity.

Line 339: How well is the temperature dependence of water activity really known for the reference systems you have in mind? For NaCl there are data available, but the situation is probably not as good for LiCl, correct? For example, Lienhard et al. (2015) reported water diffusivities at temperatures as low as 210 K.

Technical comments:

Line 322: Typo TEG

Fig. 2 and Fig.3: May be you add a residual panel (E-AIM and the continuous NaCl probe data) to illustrate the agreement.

Fig. 5: I recommend using a solid line for the simulated spectra instead of the dashed one; it makes it easier to read. Typo in figure caption: use (a) instead of (A) etc.

References:

Davies, J. F., Aerosol Science and Technology, 53, 309–320, (2019).

Lienhard, D. M. et al., J. Phys. Chem. A 116, 9954-9968 (2012).

Lienhard, D. M. et al., Atmos. Chem. Phys. 15, 13599-13613 (2015).