Atmospheric Measurement Techniques Discussions

Atmos. Meas. Tech. Discuss., author comment AC2
https://doi.org/10.5194/amt-2021-108-AC2, 2021

Reply on RC2

Jack M. Choczynski et al.

Author 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-AC2, 2021

We thank the reviewer for their comments and suggestions for this manuscript. We will address these fully in the revised manuscript, and present a brief response to selected comments here.

The method is indeed using radial growth factors. Ideally, we would obtain both radial and mass growth factors, however this is technically challenging with the dual-droplet approach. In our earlier work (Davies 2019 AST), we used the separation of two (more or less) identical droplets in the trap to deduce the trap constant, the droplet charge and the density, with mass growth factor inherently obtained. With the dual-droplet method described here, the droplets are different, both in terms of size and charge. It is no doubt possible, given the probe droplet's well characterized hygroscopicity response, that it's mass can be inferred, and a total force balance achieved. However, this would require some assumptions on the trap constant (it must be fixed with particle position, or at least well characterized), and there must be no vertical air flow. The former may be calibrated, but the latter is a technical challenge than would need to be addressed with a new chamber design that eliminates axial air movement. We recently published hygroscopic growth data for model lung fluid particles (https://doi.org/10.1039/D1CC00066G) where we report both radial growth and mass growth (via mass fraction of solute). This was achieved by disabling the gas flow altogether for short periods of time to measure the DC voltage in static air, and works well for single droplets, but with inherently less accurate RH data than the dual-droplet appraoch described here.

For low or non-soluble species in mixed aqueous particles, this would indeed disrupt the Mie resonance spectra. Depending on the morphology and phase of the particle, sizing data can still be obtained with some increased uncertainty. For example, with non-soluble protein aggregates in the aforementioned lung fluid particles, we resolve spectra that show some variation over time reflecting random scattering by aggregates. If non-soluble components become significant, then scattering spectra become incoherent. In this case, using the DC voltage for mass growth factors would be the preferred approach.

In summary, the methods we describe, while suitable across a range of RH, do show significant benefit at very high RH, where solutes are fully in solution and probe droplets provide very accurate RH. While we are currently reliant on radial growth, future developments might allow for this approach to yield mass growth.