

## **Response to Reviewer 1**

*This paper described a new humidified cavity-enhanced albedometer (H-CEA) consisted of a broad-band cavity-enhanced aerosol extinction spectrometer (BBCES) and an integrating sphere (IS) for investigating the optical hygroscopic parameters including light extinction, scattering, absorption and single scattering albedo at 532 nm. The performance of instrument was evaluated by three standard chemicals (ammonium sulfate, sodium chloride and nigrosine), the measured  $f(RH)_{ext, scat}$  agree well with literature reported values and model calculations and the estimated uncertainties were lower than 25%. The manuscript is well written and presents a valuable method in the field of aerosol hygroscopicity measurement. I recommend this manuscript to be published after the following issues to be addressed and modified.*

We thank the reviewer for the thoughtful and thorough reviews. Point-by-point responses to the comments are attached below. We have made corresponding modifications, and these changes are marked in the revised manuscript.

*1. Page 1, line 21: “from 10% to 90% RH” should be “from 10 to 88% RH”.*

Done. We changed the value 90% RH to 88% RH in the revised manuscript.

*2. Page 1, line 26-27: please cite some references to support this claim (Moise et al., Chem Rev, 2015; Tang et al., Chem Rev, 2016; Shrivastava et al., Rev Geophys, 2017; etc.).*

Done. We added these references in the revised manuscript.

### References:

- Moise, T., Flores, J. M., and Rudich, Y.: Optical properties of secondary organic aerosols and their changes by chemical processes, *Chem. Rev.*, 115, 4400–4439, <https://doi.org/10.1021/cr5005259>, 2015.
- Shrivastava, M., Cappa, C. D., Fan, J., Goldstein, A. H., Guenther, A. B., Jimenez, J. L., Kuang, C., Laskin, A., Martin, S. T., Ng, N. L., Petaja, T., Pierce, J. R., Rasch, P. J., Roldin, P., Seinfeld, J. H., Shilling, J., Smith, J. N., Thornton, J. A., Volkamer, R., Wang, J., Worsnop, D. R., Zaveri, R. A., Zelenyuk, A., and Zhang, Q.: Recent advances in understanding secondary organic aerosol: Implications for global climate forcing, *Rev. Geophys.*, 55, 509-559, <https://doi.org/10.1002/2016RG000540>, 2017.
- Tang, M. J., Cziczo, D. J., and Grassian, V. H.: Interactions of water with mineral dust aerosol: water adsorption, hygroscopicity, cloud condensation and ice nucleation, *Chem. Rev.*, 116, 4205-4259, <https://doi.org/10.1021/acs.chemrev.5b00529>, 2016.

*3. The first paragraph of Introduction is not logical and coherent, and should be rewritten. For example, adjusting the order of sentences.*

We disagree, but have modified the paragraph as follows to improve clarity:

Atmospheric aerosols directly influence global climate forcing by absorbing and scattering solar radiation (IPCC 2013; Moise et al., 2015; Tang et al., 2016; Shrivastava et al., 2017). The extinction (sum of absorption and scattering) capacity of aerosol particles strongly influences visibility, especially at high relative humidities (RH) (Massoli et al., 2009a; Liu et al., 2012). Hygroscopic particles can take up water from the surrounding atmosphere, modifying their composition, size, complex refractive index (CRI), and mixing state, and thereby altering their optical and radiant properties (Covert et al., 1972; Tang and Munkelwitz, 1994; Zhang et al., 2008; Bian et al., 2009; Kuang et al., 2015). Research on the hygroscopicity of aerosols is therefore crucial for assessing their climate and environmental impacts (Pitchford et al., 2007; Cheng et al., 2008; Bian et al., 2009).

*4. As shown in Figure 6, 7 and 8, the measured  $f(RH)_{ext, scat}$  agree well with literature reported values and model calculations, however, experiments were conducted in range of 10-88% RH, not up to 90% RH. How about the performance of the H-CEA at high RH, especially at 90% RH?*

Measuring optical properties at RH greater than 90% would be valuable but are technically challenging. Our current humidigraph system is limited by the retention time and can only humidify the dried aerosol samples ( $\sim 5\%$  RH) to 88% RH. We did not conduct experiments at a higher RH. Instead, Monte Carlo simulations were used to analyze the uncertainties of  $f(RH)$  measurement at different RH (Section 3.1.3). For RH between 20% and 95%, the uncertainties in  $f(RH)_{ext}$  and  $f(RH)_{scat}$  ranged from 7% to 25% for moderately hygroscopic aerosols ( $\gamma \sim 0.5$ ).

*5. Stability is a key factor to evaluate the application of the H-CEA, especially for field observation measurements. However, only the RH control of humidifier system had been checked for a relatively long time (8 h) in this manuscript. As to the influence of water vapor on measurements, only 40 min data were presented in Figure 4; furthermore, the performance of optical devices, including laser source, CCD and PMT may also change in a long-time measurement. More details about this aspect need to be given.*

The stability and performance of the optical system have been evaluated and discussed in our early papers (Xu et al., 2016; Fang et al., 2017). By using a high-performance temperature and current controller to control the LED light source, high stability was achieved. No obvious drifts in the transmitted light intensity and PMT intensity were observed over several hours. The influence of the intensity fluctuation arising from the optical devices (including light source, cavity, CCD and PMT) on the measurement uncertainty is negligible. In this work, the hygroscopic growth of optical parameters

was achieved by periodically changing the sample RH. For each 20 min RH ramp cycle, Figure 4 fully demonstrates the effect of the water vapour on optical signal.

We revised the text of the discussion in Section 2.1 to clarify these points.

The albedometer was periodically flushed with particle-free zero air to obtain the BBCES reference spectrum and reference scattering intensity. High optical stability was achieved by using a high-performance temperature and current controller for the LED light source. No obvious drifts in the transmitted light intensity (fluctuation  $\sim 0.1\%$ ) and PMT intensity (fluctuation  $\sim 0.5\%$ ) were observed over several hours (Xu et al., 2016; Fang et al., 2017).

#### References:

- Fang, B., Zhao, W., Xu, X., Zhou, J., Ma, X., Wang, S., Zhang, W., Venables, D. S., and Chen, W.: Portable broadband cavity-enhanced spectrometer utilizing Kalman filtering: application to real-time, in situ monitoring of glyoxal and nitrogen dioxide, *Opt. Exp.*, 25, 26910–26922, <https://doi.org/10.1364/OE.25.026910>, 2017.
- Xu, X., Zhao, W., Zhang, Q., Wang, S., Fang, B., Chen, W., Venables, D. S., Wang, X., Pu, W., Wang, X., Gao, X., and Zhang, W.: Optical properties of atmospheric fine particles near Beijing during the HOPE-J<sup>3</sup>A campaign, *Atmos. Chem. Phys.*, 16, 6421-6439, <https://doi.org/10.5194/acp-16-6421-2016>, 2016.