Response to Reviewer 2

Zhou et al. present an instrument characterization of a humidified cavity-enhanced albedometer (H-CEA) for simultaneous measurements of light extinction and scattering up to 88% RH. The instrument's performance was evaluated with ammonium sulfate, sodium chloride, and nigrosin aerosol particles. The manuscript is well written and I recommend this manuscript to be published in AMT 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. I recommend adding a section comparing the versatility and accuracy of this setup with cell-reciprocal nephelometer (Mulholland and Choi, 1998; Mulholland and Bryner, 1994; Abu-Rahmah et al., 2006) and cavity ring-down techniques (Strawa et al., 2003) equipped with cosine sensor, which also allows simultaneous measurement light extinction and scattering in dry and humid conditions (Mikhailov et al., 2006).

The main advantages of integrating sphere (IS) based albedometers over cosine sensor nephelometers are:

(1) Uniform Lambertian reflector: The collection of scattered light is not dependent on the viewing direction and thus does not depend on the location of the scattering detector on the surface of the IS.

(2) Smaller truncation angle: An IS coupled with two truncation reduction tubes can reduce the forward (backward) truncation angles to 1°.

These advantages will improve the versatility and accuracy of this setup. In this work, however, we focused our discussion on the cavity-based albedometers. A comparison between our cavity-enhanced albedometer and other cell-reciprocal nephelometers is not part of this study, but reference to these nephelometers has been added to the text:

These instruments include <u>the cell-reciprocal nephelometer (Mulholland and Choi,</u> <u>1998; Mulholland and Bryner, 1994; Abu-Rahmah et al., 2006; Mikhailov et al.,</u> <u>2006), the CRDS-reciprocal nephelometer (Strawa et al., 2003), and other optical</u> <u>cavity-based albedometers, including</u> the CRDS-albedometer (Thompson et al., 2008; Ma et al., 2012), the BBCES-albedometer (Zhao et al., 2014; Xu et al., 2018a) and the cavity attenuated phase shift spectroscopy (CAPS)-albedometer (Onasch et al., 2015), which combine CRDS/BBCES/CAPS with integrating spheres (IS). These albedometers are suitable for operating under high RH conditions and have <u>sampling</u> advantage<u>s</u> over independent measurements of different parameters with different instruments (Wei et al., 2013; Zhao et al., 2014; Xu et al., 2018a). References:

- Abu-Rahmah, A., Arnott, W. P., and Moosmüller, H.: Integrating nephelometer with a low truncation angle and an extended calibration scheme, Meas. Sci. Technol., 17, 1723-1732, https://doi:10.1088/0957-0233/17/7/010, 2006.
- Mikhailov, E., Vlasenko, S., Podgorny, I., Ramanathan, V., and Corrigan, C.: Optical properties of soot-water drop agglomerates: An experimental study, J. Geophys. Res., 111, 1-16, https://doi.org/10.1029/2005JD006389, 2006.
- Mulholland, G. W., and Bryner, N. P.: Radiometric model of the transmission cellresiprocal nephelometer, Atmos. Environ., 28, 873-887, https://doi.org/10.1016/1352-2310(94)90246-1, 1994.
- Mulholland, G. W., and Choi, M. Y.: Measurement of the mass specific extinction coefficient for acetylene and ethene smoke using the Large Agglomerate Optics Facility, Proceedings of the 27th Symposium (International) on Combustion, 27, 1515-1522, https://doi.org/10.1016/S0082-0784(98)80559-6, 1998.
- Strawa, A. W., Castaneda, R., Owano, T., Baer, D. S., and Paldus, B. A.: The measurement of aerosol optical properties using continuous wave cavity ring-down techniques, J. Atmos. Ocean. Technol., 20, 454-465, https://doi.org/10.1175/1520-0426(2003)20<454:TMOAOP>2.0.CO;2, 2003.

2. Section 3.1.1 How particle losses were evaluated? Please specify in detail.

We added the following description in the revised text.

The particle loss was characterised based on the difference in concurrent CPC measurements at the inlet and outlet of the sample tube or cavity, after accounting for dilution inside the cavity.

3. Section 3.1.3 A single DMA, in addition to selected particles, transmits large multiply charged particles. How was this taken into account in the uncertainty analysis?

Multiply charged particles were characterized by a tandem DMA (TDMA) method (*Bueno et al., Aerosol Sci. Technol. 45, 1217-1230, 2011; Zhao et al., Anal. Chem. 85, 2260-2268, 2013*). In this work, by carefully adjusting the flow rates of the sample and sheath gas of the first DMA, the fraction of the multiply charged particles can be minimized. Larger multi-charged particles were effectively removed by inertial impaction at the inlet of the classifier, which have little effect on the measurement of f(RH).

4. Section 3.2.1 Since E-AIM is an accurate thermodynamic model and can be used as a reference standard, I recommend first compare the measured optical coefficients with these calculated from E-AIM-based values. The obtained difference should be discussed and indicated in Table 1.

DONE. We added a new table in the revised text.

Table 2. Comparison between the measured and E-AIM model calculated extinction and scattering cross sections for the size-selected ammonium sulphate and sodium chloride particles under three selected RH conditions.

Species	RH (%)	Particle	Cross section (×10 ⁻¹⁰ cm ²)		
		diameter	Measured		Calculated
		(nm)	σ_{ext}	σ_{scat}	$\sigma_{ext} = \sigma_{scat}$
Ammonium sulphate	<40	200	1.97±0.03	1.96±0.02	2.06±0.23
		250	4.34±0.12	4.17±0.10	4.60±0.30
		300	9.56±0.40	9.34±0.29	9.70±0.90
		350	18.61±0.64	17.64±0.59	18.25±1.22
	80	200	6.22±0.09	6.22±0.11	6.81±0.62
		250	14.66±0.69	14.29±0.61	15.13±0.76
		300	27.68±0.89	26.40±0.97	28.04±2.39
		350	49.31±1.92	44.88±1.64	48.60±2.77
	85	200	7.81±0.18	7.81±0.18	8.61±0.74
		250	17.85±0.48	17.35±0.46	18.73±0.90
		300	32.62±1.08	31.15±1.00	34.06±2.82
		350	58.87±3.75	53.55±3.13	58.69±3.16
Sodium chloride	<40	200	1.85±0.03	1.81±0.02	1.99±0.07
		250	4.05±0.08	3.94±0.07	4.03±0.25
		300	9.04±0.18	8.53±0.20	9.33±0.43
		350	20.38±1.13	17.97±1.26	19.76±0.93
	80	200	17.08±0.29	16.58±0.27	18.53±0.36
		250	36.53±0.66	34.63±0.82	33.74±1.44
		300	65.61±2.63	60.54±1.86	63.75±2.24
		350	108.91±6.58	94.39±4.67	110.93±3.58
	85	200	21.18±0.47	20.51±0.40	24.19±0.46
		250	45.80±0.93	43.06±0.79	43.93±1.79
		300	79.47±3.57	72.80±3.17	80.58±2.71
		350	135.75±6.19	117.60±4.70	136.83±4.00

The corresponding discussion was added in the revised text.

The measured extinction and scattering cross sections agreed with values calculated using the E-AIM model for different particle diameters and RH conditions (Table 2). The overall consistency between H-CEA measurements and other measured and calculated values in the literature indicate the reliability of the H-CEA instrument.

5. Section 3.2.1, Fig.6 and Fig.7 As the RH measured with T/RH-sensor-2 is lower than actual RH by ~2% (Amm. sulfate measured DRH =77-78% vs. 80%), the experimental f(RH) values must exceed the model coefficients especially at high RH, which contradicts the data presented in Fig.6 and partially given by Fig.7. What is the RH difference between T/RH sensor-2 (input) and T/RH sensor-3 (output)? Due to water vapor sorption on the huge setup surface, the RH difference will be time-dependent. I recommend checking out the RH difference vs. time at least at RH>85%. According to Fig. 2, a full measurement cycle was 20 min. If so, then it is likely that in this short time, the thermodynamic equilibrium was not reached, and real RH was lower than that measured by T/RH sensor-2. As a compromise, the average RH can be used for data plotting. Please consider the issue outlined above.

The measured deliquescence RH (DRH) of ammonium sulphate was ~ 2% lower than the value reported in the literature and broadly in line with the accuracy of the T/RH sensor ($\pm 1.5\%$). After considering the dilution inside the cavity by the mirror purge gas, the difference between T/RH sensor-2 and T/RH sensor-3 was within the accuracy of the sensors. Since the T/RH sensor-2 was closer to the inlet of the cavity, it was only necessary to consider the purified air near the front mirror to obtain the sample RH.

As can be seen from Fig. 2, the RH control of the humidifier system has a fast response speed. Each humidification cycle was well controlled. The sample humidity can be increased rapidly from 10% to 90%. Furthermore, the repeatability was good. Therefore, the thermodynamic equilibrium should have been reached.

6. The data should be made available in a FAIR aligned repository. Making data "available upon request to the author" is inconsistent with the AMT data policy (https://www.atmospheric-measurement-techniques.net/about/data_policy.html).

Done. We added the data access method in the revised text.

Data availability. The data used in this study can be obtained from https://pan.baidu.com/s/1hIFEJQPwKX8gJH9ZRs0KMw. The extraction code is m7ub (last access: 22 April 2020).