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AMTD

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Interactive Comment

Interactive comment on "Measurement of relative humidity dependent light scattering of aerosols" by R. Fierz-Schmidhauser et al.

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We thank Anonymous Referee #1 for all of his valuable suggestions and comments, which certainly helped us to improve this paper and make it more concise and clear. In the following we give detailed answers and explanations to the issues raised.

Comment: The manuscript presents laboratory and ambient data of light scattering coefficient from two integrating nephelometer systems in which the relative humidity was measured, controlled and scanned to yield the hygroscopic, deliquescence and hysteresis properties of the aerosols. The fundamental light scattering ratio at 80% RH, f(RH), presented for quasi-monodisperse test aerosol of known composition, is new and reasonably good agreement is found between measured and modeled f(RH) for the test aerosol though the error bands are large. However, the agreement between





measured and modeled scattering coefficient results does not seem to be within the range of experimental uncertainty. This is contrary to the statements in the text (see figure A2 and A3). Again the uncertainty ranges are large.

Response: The measurement of the scattering coefficient divided by the number concentration of particles is in the range of the experimental uncertainty for all particle sizes except for those with a dry diameter of 300 nm. The measurement and model of the 300 nm particles do show the same deviation at dry conditions. Therefore, we can assume that the discrepancy is due to uncertainties in the measurement of the number size distribution, which influences the scattering coefficient strongly via the doubly and triply charged particles.

Comment: Some aspects of the data analysis remain unclear, particularly the truncation and RH corrections. The narrative is succinct to the point of being laconic.

Response: The scattering coefficients are truncation error corrected. We will add this missing information in the new manuscript.

Comment: Figures 2 through 4 focus on f(RH) while figure 5 presents light scattering coefficient as a function of RH. It would be clearer if the data in figure 5 were normalized to present f(RH) as well.

Response: It is not possible to present the data in Figure 5 as f(RH) since the DOE/ARM dry nephelometer changed between PM10 and PM1 every half an hour. That means the PSI humidified nephelometer can be only normalized for half of the RH cycle (either hydration or dehydration), and with showing only half of the cycle the important information of this figure would be lost.

Comment: Page 2163, line 20, Some added discussion of the connection between hysteresis, atmospheric aerosol hysteresis and metastability would be useful to the reader and would support the subsequent results and discussion thereof.

Response: We will add the following sentences to the new manuscript on page 2163:

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"The exact knowledge of the DRH and ERH points is of great importance because they define – based on the RH history of an air parcel – which fraction of the atmospheric aerosol is present as purely liquid droplets. This fraction is scattering much more light than the fraction that is solid."

Comment: line 17 The correction is small but easily calculated and applied. How does this error compare to that derived from the T and RH uncertainties?

Response: This is true, the error is small, but for the lab measurements it was very important that the flow stayed constant. The uncertainty of the RH is assumed to be +/-2% (see page 2168, line 21).

Comment: What truncation correction scheme was applied to the data? Given the quasi-monodisperse nature of the laboratory aerosol, what added uncertainties are there if the usual polydisperse correction schemes are applied?

Response: see above.

Comment: Page 2166, line 21 Just to be perfectly clear: Since we refer to this lower RH in the nephelometer in all our figures, the apparent deliquescence step change appears at a lower RH than the true literature value,

Response: We will change that to apparent.

Comment: Page 2167, line 3, I think the proper term is: monotonically

Response: The sentence will be changed, due to other reviewer comments.

Comment: line 9, Put the residence time in context of the equilibration time of hydration and dehydration for submicrometric particles. Ditto for the DOE system.

Response: The residence times are put into context on page 2173, line 27 (and following). The different residence times of the two humidified nephelometers is only a hypothesis, so we don't want to extend the discussion on this possibility further than done until now. AMTD

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Comment: Page 2169, line 10, Quantify "minimal". Is the same IR blocking filter and cooling system as in the PSI nephelometer used?

Response: No, no IR blocking and cooling system is used in the DOE system. The aerosol enters the nephelometer at a higher T (heated from the humidifier). Delta T=0.13°C, standard deviation: 0.53° C.

Comment: Section 2.3 What were the uncertainties in the DOE system including RH, T, losses?

Response: Carrico et al., 1998 describes that agreement between different RH measurements during scanning is within 3% RH, 6-7% losses between dry and humidified nephelometer (see p. 2172, line 16)

Comment: Page 2170, line 19, Even though the

Response: We will change it to "though"

Comment: line 21 A previous experiment showed differences in the measured vs. Mie scattering anomaly for monodisperse aerosols at different wavelengths, Heintzenberg et. al. Was that not the case here?

Response: No it was not. We observed for all wavelengths a similar behavior. This is mentioned on page 2170, line 19.

Comment: Page 2171, line 18, The measurements are compatible with the model. I don't know how to interpret the term "compatible". What is this quantitatively?

Response: We will change the sentence to: "The measurements agree with the model prediction within their rather large uncertainties." We calculated the differences between theory and measurements and will add this in the manuscript.

Comment: Line 20 Similarly, define "very good agreement".

Response: We calculated the difference between theory and measurement and will

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add this. (Here, between 4.5 and 7% difference between theory and measurement).

Comment: line 21, Figures A2 and A3 Can the difference between the scattering per particle at 100 and 150nm cf. 240 and 300nm be explained? One set of results is at the upper limit of theory the other at the lower.

Response: We cannot really explain this. But Heintzenberg et al., 2006 also observed a tendency of systematically higher calculated signals as compared to measured ones, in particular at lower particle concentrations. At 300nm the particle concentration was lowest and the calculated compared to measured values were highest.

Comment: Page 2172, line 15, How was the empirical correction determined? How does it compare to the loss determined for the PSI system?

Response: We divided the dry scattering coefficient of the dry nephelometer by the dry scattering coefficient of the humidified nephelometers. On average the dry scattering coefficients of the humidified nephelometer were 5.8 (PM10) or 6.6% (PM1) lower. PSI: 3.1%.

Comment: line 21, On most days the f(RH) obtained by the PSI humidified nephelometer was slightly higher... The result should be quantified more completely than the graphical presentation, the range of f(RH)s and "slightly". This result is the crux of the experiment. What are the mean values, mean difference and standard deviations thereof, the mean difference for the 4 selected cases? This is the result needed in tabular form for easiest understanding and as a reference for subsequent experiments and for modelers to use, for this aerosol case anyway.

Response: The f(RH=80%) of the PSI humidified nephelometer was for the PM10 size fraction on average 1.54 (+/- 0.19) and for the PM1 size fraction on average 1.46 (+/- 0.19). The DOE/ARM humidified nephelometer measured on average f(RH)s of 1.27 (+/-0.16) (PM10) and of 1.25 (+/-0.10) (PM1). We will change the sentence to "The f(RH) obtained by the PSI humidified nephelometer was on average 21% (PM10) and

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16% (PM1) higher ..."

Comment: Page 2182 The model values shown in figure A3-a should also be presented in figure 2.

Response: We will not do this, because the figure would become too busy.

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