

Interactive comment on “Effects of relative humidity on aerosol light scattering: results from different European sites” by P. Zieger et al.

Anonymous Referee #1

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General comments:

The manuscript is fairly clear though figure 3 and related discussion. After that a lot more detail will be needed to describe the methods use to model and estimate ‘closure’. Much of the latter half of the manuscript is difficult to read and interpret.

Visually there is very little to be gained from figures 7 and 8 even when magnifying them by 300%. A table quantifying the main effects of dust and coarse mode seasalt as described in the text would be adequate.

A definition of closure and a means of quantifying the closure for each of the modeling parameterizations and locations is needed. Without a definition of closure and inclusion of and propagation of the uncertainties in the parameters that are compared,

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“achievement of closure” is a hollow claim.

The Köhler kappa parameter is often invoked. It is not always clear how that kappa value was determined.

The OPAC parameters were determined to be inadequate for many atmospheric aerosol types and recent literature values were recommended. A table of the original OPAC values and component types should be compared to values of recent literature values as mentioned but not cited or presented quantitatively in text. This would be of value to the community to adopt or further revise.

I recommend that the manuscript be shortened to cover the observations and analysis thereof or that a major revision of the second half be made to include more detailed explanations of the methods and results and conclusions.

Specific comments:

Abstract line 2

“...to 3.41 for Arctic aerosol.” Strictly, ‘to 3.41 for Arctic marine aerosol.’

P 8943, line 15 This period was characterized by very low particle concentrations and distinct sea salt transports transport to the station

P 8994 line 14 Henne et al. (2010) categorized the CAB site as agglomeration. . . This last word doesn’t make sense. Simplify to: Henne et al. (2010) characterized the CAB site as one impacted by large and varied pollution burdens from multiple sources.

P 8945 line 5 . . .in the humidogram humidograph mode. In the latter one mode . . . Humidograph is the proper term for the instrument. Humidogram is the record or graph made by the humidograph. I know this syntax is counterintuitive but there it is. Revise here and throughout the manuscript.

Section 3.1 A block diagram of the humidograph system and an example of the RH time profile through the instrument would be valuable here.

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Line 22 Delete “e.g.”, not needed.

P 8948 line 2 droplet converges to the one that of water.

P 8948 paragraph 2 The model $f(RH)$ results presented in Fig. 1 and described here are of value to the extent they have been used in calibration (QA tests) of the humidograph though that is mentioned only peripherally and has been published already and adequately referenced. The data does not represent the atmospheric $f(RH)$ data presented in figure 3 or table 1 except in the diameter range 100 to 1000 nm. The plot and discussion should be limited to that range. Your points about chemistry vs. size in the text can still be made without unrepresentative, misleading, high $f(RH)$ values.

P 8949 line 22 The inverse calculation needs explanation or a reference.

line 24 “At MHD, all relevant aerosol measurements except of for chemical measurements ...”

P 8950 line 15 Figures refer to this as SDE. Saharan Dust Events? Mention this acronym here and in figure legends or captions.

line 23 “. . . behavior as one would expect from relatively pure inorganic salts like NaCl.”

P 8951 several places. I understand how hysteresis could be measured with the humidograph system as described. I fail to see clear evidence of hysteresis in the data, e.g., Figure 4 where it is mentioned. Clear evidence would be the difference between a humidogram of increasing RH from $RH < \text{efflorescence}$ and a humidogram of decreasing RH from $RH > \text{deliquescence}$.

Page 8951 line 10 The main catchment area at NYA 10 was the open oceans and ice shields of the Arctic, bringing, e.g. clean sea salt to ... Catchment is not the right term, geographically it refers to river basins. Use the term ‘source area’. What is “clean” seasalt?

P 8952 line 1 “settings”? More properly termed ‘limitations’, perhaps.

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line 5 Define optical closure quantitatively and how that was calculated. Is that $f(\text{RH})$ closure, specifically or optical closure, i.e., the intensive cf. the extensive variable?

line 7 At JFJ, the best closure was achieved, if the. . .

line 8 and fig 5 The g factors in the legend are not explained other than in cursory fashion here; please expand and add to figure captions perhaps only by reference to text section.

line 12 “showing that a mean chemical composition is sufficient to predict $f(\text{RH})$. This result is in accordance with findings of Jur’anyi, et. al. (2010) who showed that for a prediction of the cloud condensation nuclei (CCN) number concentration, using measured size distribution and hygroscopicity measurements (H-TDMA), a mean chemical composition is sufficient.” The accordance you mention is not strictly relevant since CCN activation occurs at a much higher RH and water content per particle. The more relevant reason, briefly, is that the integral (over size) chemical composition is dominated by the majority of the particulate mass being in the accumulation mode to lower coarse modes, the size ranges that contribute most to light scattering and control $f(\text{RH})$. You should define the size range of your mean chemical composition, total mass, sub-10, sub-2.5, sub-1 μm ; filter, AMS.

line 19 The modal value of the Nessler-based predicted to measured is an overestimate, i.e., >1 . But there is a significant tail of underestimates in the PDF. How do both of these relate to Ångström and dust? “Validation” is the wrong term. ‘A seasonal comparison’ would be better. So far Nessler’s model is not validated for the aerosol at this location

Page 8953 line 1 and following Its not clear how these g values were obtained from size distribution, scattering, absorption and volume ratios.

line 17 “. . .because the aerosol origin showed larger fluctuations.” . . .because the back trajectories showed a large fluctuation in transport vectors and likely aerosol sources.

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line 21 "...largest dry diameter of the deployed H-TDMA was 165 nm. The larger particles in the coarse mode accumulation and coarse modes were therefore missed by the H-TDMA." Its not just the coarse mode that was not sampled.

line 25 Again, what is the proposed parameterization?

6.3 "recipe" 'formula' or 'analytical method' would be better here. This is basically a sensitivity study.

P 8955 line 11 "width" is OK but standard deviation is the more scientific, statistical term.

line 16 It hard to put these number ratios in context given the steep slope of the number distribution and where the fine-coarse size cut is made. A volume ratio would be more intuitive and comprehensible.

Page 8956 lines 19 through 21 Terms such as "predominantly", "will fail", "reliable" are not useful in this discussion. The discussion needs to be quantified.

Page 8957 The OPAC model often used as a basis for hygroscopic growth in global chemistry models but other aerosol classes are added and/ or heavily modified. I'm not convinced this exercise is relevant. It could be improved by a table of original and modified OPAC parameters.

Page 8958 Modal terminology here and elsewhere is not consistent or defined, accumulation ,fine, coarse.

Page 8961 line 15 and following The modifications that were made to OPAC components is not clear.

Page 8962 The first recommendation (and the earlier description of the data reduction process for the nephelometer) does not mention the illumination, truncation correction also needed for a precise measurement. The internal, Ångström-based correction has large uncertainty when coarse particles are present. A better correction requires size

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distribution measurements, ideally at the sensing volume RH of the nephelometer.

For a given aerosol, kappa changes depending on the RH range over which it is applied. Specify recommended range limits.

Strictly speaking the third fourth and fifth points are not recommendations but only observations or conclusions.

Fig 4 The color coding is not different enough to distinguish the traces for individual locations.

Fig. 8. “Same as Fig. 7 but for varying imaginary parts of the refractive index and varying width of the fine mode.”

Add that” Indicated values of sigma and m_{imag} as indicated in the legend are compared to the reference case given in figure 7.

Fig 10 Not all parameters in the legend are evident in the plots, e.g., Melpitz. Are they overplotted or not in the data set? If the latter delete from legend.

Fig 11 Several of the measured distributions look unreasonable. Is this due to poor sampling statistics.

Fig 13 This includes a range of uncertainties in OPAC-based $f(\text{RH})$. What is the range of uncertainty in measured $f(\text{RH})$?

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