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Interactive comment on "A global model-measurement evaluation of particle light scattering coefficients at elevated relative humidity" *by* María A. Burgos et al.

Anonymous Referee #2

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This paper discusses an important aspect of aerosol modelling: the growth of aerosol with increasing humidity. This wet-growth has important implications for aerosol transport and aerosol optical properties (including aerosol-radition-interactions). The authors conduct a process study by comparing the light scattering enhancement factor f(RH) from models with that from several observation sites. The observational data is novel and the evaluation of models highly relevant to numerous modelling groups, including the AEROCOM community.

General comments: The study itself seems properly conducted and the paper is mostly well-written. However, it seems the authors were only interested in a very narrow objective: numerically comparing model data with observations. There is very little interpretation of the results. While I appreciate that a full interpretation might be a study in itself (and the authors suggest they are working on it), this is rather unsatisfying. It prevents the reader from tapping in to the combined expertise of the authors (observers & modellers) and a better understanding of the limitations and opportunities associated with the current study.

In particular, I missed a discussion of why models might have a different f(RH) from the observations. I suppose there are at least three reasons: 1) incorrect wet-growth of individual species (e.g. incorrect kappa); 2) incorrect internal mixing rule for wet-growth; 3) incorrect internal and external mixing states. The advantage of a process study is of course that the models do not need to accurately simulate mass loads themselves.

If possible, it would be useful to provide more information on per-species wet-growth in individual models, especially because of the finding of substantial wet-growth at low RH.

In addition, I found important information on e.g. observational errors and methodology to be missing. Yes, the authors refer to Burgos et al 2019 but it would be good if brief (!) summaries of relevant sections in Burgos et al 2019 are provided.

Specific comments:

Abstract: the abstract contains several conclusions without any attempt at interpretation.E.g. "An important finding is that the models show a significantly larger discrepancy with the observations if RHref =0 % is chosen as the model reference RH compared to when RHref =40 % is used" might become "The definition of dry conditions is difficult from an observational point-of-view, which affects our model evaluation negatively as several models exhibit significant and unexpected wet-growth between RH=0 and 40%.". One interesting finding (also supported by a recent Gliss study) is not included in the abstract.

Introduction: I miss a discussion of the importance of correctly simulating wet-growth in models. How does wt-growth affect different aspects of simulation? E.g. emissions are unaffected but both wet and dry deposition are. Consequently, so is transport. At the same time, optical properties (important for ARI) are affected. Anything else (e.g. chemistry)? While wet-growth ultimately leads to activation of aerosol as cloud droplets, models usually disconnet these processes.

Two papers that consider impact of f(RH) on modelling of biomass burning: Johnson, B. T., Haywood, J. M., Langridge, J. M., Darbyshire, E., Morgan, W. T., Szpek, K., . . . Bellouin, N. (2016). Evaluation of biomass burning aerosols in the HadGEM3 climate model with observations from the SAMBBA field campaign. Atmospheric Chemistry and Physics, 16, 14657–14685. https://doi.org/10.5194/acp-16-14657-2016 Reddington, C. L., Morgan, W. T., Darbyshire, E., Brito, J., Coe, H., Artaxo, P., Scott, C. E., Marsham, J., and Spracklen, D. V.: Biomass burning aerosol over the Amazon: analysis of aircraft, surface and satellite observations using a global aerosol model, Atmos. Chem. Phys., 19, 9125–9152, https://doi.org/10.5194/acp-19-9125-2019, 2019.

An introduction should end with a: - a brief description of the paper's methods and goals, and why/how they adbvance the field (i.e. add to the existing body of work) - a short description of the content (sections listing). I miss both and the introduction would become substantially stronger if they are added. P 3, l 20-34 does not really provide this.

p 4, sect 2: what I miss here is a brief discussion on expected uncertainties. I have no idea how much can be said about this but admitting to 'uncertainty in the uncertainty'would be acceptable. E.g. at RH=40 or 85%, can we expect measurement uncertainties in f of 5, 10, 15% in individual measurements? Derscribe briefly the main causes of uncertainty. Do you expect errors at a single site to behave like biases or random errors? This discussion would be very useful. If this was discssed in Burgos et al 2019, please provide highlights and the reference.

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p 4, l 16: given the uncertainty in the analysis at low RH later identified, it would be good to be a bit more precise about low RH measurements? Do the various instruments measure at different low RH? Does a single instrument measure at a single RH, or does it vary for some reason? If it varies, does it vary in a controlled fashion , or not? I see that Table 1 only provides median values and gives not indication of any variation (large or small). p 5, Section models: it seems that the models broadly fall into 2 (3?) categories based on how f(RH) is calculated: either from direct parametrisation (e.g. OPAC), or from Koehler theory (is it fair to include ZSR theory?), or from equilibrium theory (sulphate-nitarte-amoonia, see e.g. Seinfeld & Pandis). I'm not sure the latter category is present in the current family of models but maybe TM5 uses it? Anyway, would it make sense to stress these three broad categories?

p 5, l 3: can the authors say a little about what aspects of data homogenization are considered important, and were different in Titos et al.? It would help make the paper stand out more.

p 5, l 26: So the GEOS family of models are CTMs? Isn't MERRAero an assimilation product?

p 5, l 27: There is no information in Table 2 on hygrospcopic growth of sulphate (most important anthropogenicv aerosol) and organics (to my knowledge, the most uncertain aerosol wrt hygrospcopic growth). Is it possible to include at least these species?

p 5, l 29: is there a name to identify the relevant AEROCOM experiment? Please also provide link to AEROCOM website so interested readers can follow up.

p 5, l 32: I assume surface values were used? PLease state so.

p 6, l 1: Here or at a more appropriate location, it would be good to have a brief discussion of possible impact of the difference between model and observational years.

p 7, l 16: (Bey at al.) -> Bey et al.

p 10, l 7: all hourly output for that month is used, or only at the hours (and presumably days) of observations. Given thge possible importance of representation errors, it may be worthwile to have a short section (Methods) that describes actual collocation procedure in a bit more detail, especially as there are basically two cases: sites that provide data for 2010, and sites that don't.

p 10, l 20: is the gray shaded area for measurements mostly due to measurement error or temporal variability? This has consequences, because if it is due to measurement error, a lot of models greatly overestimate f(RH) variability.

p 10, l 23: this information could be part of the short section suggested above?

p 10, l 27: what is the altitude of these mountain sites? Is it ok to use those measurements for model evaluation when model's orography may not ab able to 'simulate' the mountain?

p 11, l 10-20: ignoring the observations, it appears to me that the models exhibit fairly similar behaviour independent of site. E.g. GOES family rather low f(RH) with small variation, CAM, TM5 and SALSA higher f(RH) with more variation. Can anything be said about that? p 11, l 21: if I'm correct, this is the same data as in Fig 1 but represented in a different way? It may be good to specifically state so.

p 11, l 23: This uncertainty should be mentioned earlier, when describing the measurememnts. Is this the uncertainty in a single measurement, or in an average of measurements?

p 11, l 31: Is it fair to conclude that external mixing reduces variation in f(RH)? And if so, why would that be?

p 12, l 2: are you sure? I thought SALSA uses a combination of internal and external mising.

p 12, l 27: for SGP I also see substantial differences but they seem to their own annual cycle (larger differences in summer).

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p 12, l 28-30: nice analysis, ultimately uncertainties always have to be considered in the context of other uncertainties.

p 13, l 15-17: Dare the authors guess at why even CAM, OsloCTM and TM5 do poorly? They showed pretty strong correlation across the sites. So it appears that these models have skill in predicting spatial patterns but none in predicting temporal evolution. Yet it would appear that observed variation across sites is similar to variation within each site (guesstimating from Fig 1).

p 14, l 26-27: surely the developers of the GOES and SALSA models (who are co-authors) must have an idea where this is coming from? It would be very useful to add such information to the paper. From a modelling perspective, it would be relatively straight forward to present f(RH) curves for each species (i.c. of internal mixtures: predominant species). Such a figure would be a very useful addition to this paper.

p 15, l 10-15: this is an interesting finding that should be in the abstract (there is only an oblique reference to it at the moment).

p 15, Section Conclusions: the current text focusses entirely on techniqcal issues and ignores interesting findings as mentioned above. It would be good if more emphasis is given to lessons learned with regards to possible model deficiencies. There is no speculation why models over-estimate f(RH). Or do the authors believe this is due to remaining technical issues?

p 15, l 17: Define "f(RH)"

p 15, l 19: Define "RH_ref"

p 15, l 27: "at low RH". Did you mean "for low RH_ref"?

p 15, l 30: models should be able to provide f(RH) at multiple RH without any significant CPU or development overhead.

p 16, l 4-6: this gamma parameter has not been mentioned before so it's odd to discuss in the conclusions. Obviously, you may want to mention other possible analysis methods.

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