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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Long term measurements of optical properties and their hygroscopic enhancement" *by* M. Hervo et al.

Anonymous Referee #1

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Hervo et al. present in their study the analysis of optical measurements at the Puy de Dôme site for a time period of seven years. This includes the discussion on temporal (annual, seasonal and diurnal) and spatial (trajectory analysis) variation. In addition to direct measurements of particle light scattering and absorption coefficients, the discussion is also extended to calculated intensive parameters (single scattering albedo, complex refractive index and asymmetry parameter). A focus is set on the calculated scattering enhancement factor (2 years), its variability and possible parametrization.

While the topic of the paper is of general interest to the scientific community, it has some major methodical limitations and also suffers from some presentation errors so that significant doubt arises as to the overall scientific quality of the work. A proper assessment of this study is thus not fully possible due to limitations in the methodology

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and the data analysis. The major points are described below (in arbitrary order) and have to be answered thoroughly in order to achieve the quality standards of ACP (major revisions).

Major comments:

1. **Trajectory analysis.** I strongly doubt the result of the trajectory analysis, for the following reasons. It is not clear to the reviewer what exact criteria were taken to cluster the trajectories. It is especially unclear if the time of the air parcel residing within the planetary boundary layer was taken into account. Please add a figure on the spatial surface residence time retrieved from the trajectory analysis. What exact life time (here called "decreasing weight") was assumed? Was precipitation and thus wash-out taken into account? The unreasonably low values (down to 0.7) of the single scattering albedo over the Atlantic could be as result of this (e.g. when free tropospheric air was categorized as marine). They are also clearly in contradiction with observations (see e.g. Quinn et al. 1998 or Russel et al. 2002). The same is valid for the asymmetry parameter g: I would expect higher values of g for coarse mode dominated aerosol (see Andrews et al. 2006 or Fiebig and Ogren, 2006) coming from the Ocean or the arid regions. Instead the opposite is observed here.

2. **Retrieval of the refractive index.** It is astonishing that the single scattering albedo shows a clear seasonal variation, while the imaginary part (related to the absorption) does not (see Fig. 1) and is not distinguishable for the different air mass types. Therefore, doubts arise on the informative value of this analysis. The retrieved complex refractive index depends strongly on the quality and the agreement of the different size distribution measurements (SMPS and OPC; electrical mobility vs. optical equivalent diameter). The authors should proof the agreement between SMPS and OPC by showing median and percentile values of

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the entire surface size distribution (important for the optical properties) by assuming spherical particles. This should be done for the different air mass sectors and seasons separately. The quality of the agreement and the limitations (and uncertainty) of this approach should be discussed. Last, the diameter where SMPS and OPC were merged has to be stated in the text.

The comparison of the retrieved refractive index to AERONET retrievals presented here is questionable since a columnar value is compared to a point measurement. A profile of the RH and the particle size distribution would be needed to do this properly.

3. **Calculation of wet optical properties and importance of the coarse mode.** The calculation of the scattering enhancement factor f(RH) is not done properly and thus the entire section is questionable. As pointed out in Zieger et al. (2013) and Zieger et al. (2014), the coarse mode is an important factor contributing to f(RH). On the one extreme and simplified, it can be mineral dust (non-hygroscopic) or sea salt (very hygroscopic). The authors themselves show in the trajectory analysis that the site is influenced by sea salt and desert dust influenced air masses (Page 27744, Line 1: "Air masses sampled at the PdD station originated in majority from the oceanic sector (37.8% over the calculation period), while African, continental and oceanic modified air masses represented 27.1, 18.4 and 20% of the air mass types, respectively."). Therefore this has to be taken into the account for the f(RH)-predictions, e.g. by presenting a detailed sensitivity study or by assuming different hygroscopic growths factors and refractive indices for the different air mass sectors. In addition, it has to be clarified if an internal mixture was assumed and which refractive index is taken for the calculations. Why was only one dry diameter of the HTDMA being used?

Within this part of the work, the authors often confuse "observation" with "prediction" or "calculation". The scattering enhancement factors at PdD are a prediction or calculation based on different assumptions and this should be clearly sepa-

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rated. At Jungfraujoch, for example, Fierz-Schmidhauser et al. (2010) compared a direct measurement of f(RH) to Mie predictions. Jungfraujoch also offers the simplification of the coarse mode being predominately mineral dust.

The deliquescence behaviour was also observed for polluted marine and long-range transported sea salt particles (e.g. at Melpitz where it was transported approx. 450 km to the site, see Zieger et al., 2014). The authors themselves define two large air mass sectors which are called oceanic and oceanic modified (see Fig. 5), thus one would expect that these air masses could exhibit deliquescence behaviour.

The proposed polynomial parametrisation is therefore highly questionable, since a modelled (and not validated!) quantity is parametrized (which later is being advertised to be used by models). In addition, the points in Fig. 5 were extrapolated assuming the validity of the κ-approach which, as shown in the way here, is definitely questionable for the marine cases, because they do not capture the course of the upper branch of the hysteresis curve correctly. Therefore this parametrization should be removed from the manuscript, because their is the potential risk that these errors keep on propagating into future studies.

4. **Discussion of the diurnal variations.** The main figures (Fig. 2 and 3) which present the diurnal changes of the different properties are only shown until 21:00 and the night values are therefore missing. These periods should be added (or maybe the axis labels are wrong?) and the argumentation within the manuscript has to be reviewed for validity. The course of the percentile values of the boundary layer height is also strange (e.g. the 25th percentile values of the summer case is very close (and one time even equal) to the median. Is this poor counting statistics or a bug in the presentation?

5. The **title** is still misleading: It should be emphasized that we are talking about aerosol optical properties and that the scattering enhancement is not directly

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measured but rather calculated or estimated. Therefore I suggest to add two words to the title: "Long-term measurements of aerosol optical properties and their estimated hygroscopic enhancement" ... the authors may also add at the end: "at Puy de Dôme" or another appropriate geographic definition.

Detailed comments:

- Title: There should be a hyphen in between long-term.
- Page 27732, Line 2: Please replace "...measured from the GAW..." by "...measured at the GAW...".
- Page 27732, Line 23: The effect on visibility is not just valid for urban areas, but rather everywhere important.
- Page 27732, Line 23: "... over more than more than ..."
- Page 27734, Line 24: To separate the interstitial from the residual (activated) aerosol, a CVI (counterflow virtual impactor) inlet would be needed, which is not of importance for this study. Please clarify.
- Page 27735, Line 3: The Anderson and Ogren (1998) correction does not correct for detection limits. Do the authors mean the illumination correction for the nephelometer?
- Page 27736, Line 1: Please define SD at its first appearance.
- Page 27736, Line 3: At which dry diameters did the HTDMA measure? Why was only the 110-nm diameter selected for the analysis?
- Page 27736, Line 21: Please properly define the scattering enhancement factor (best with an equation) at its first appearance.

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- Page 27737, Line 8-11: This statement is a bit too simple. The same study showed e.g. for Melpitz that the differences (ratio between measured and predicted) can vary between 1.20±0.25 and 1.02±0.22 (mean ± standard deviation) depending on which property of the coarse mode was assumed (NaCl or mineral dust). In addition, Zieger et al. (2013) also gives a general uncertainty estimate for model calculations of f(RH) in their appendix, which might be useful for this study.
- Page 27739, Line 19 (and cont.): To be consistent with the literature, I would suggest to put the zero of the ω_0 as a subscript.
- Page 27738, Line 17: Please define the seasons also by the months.
- Page 27738, Line 21: It should be "At PdD ..." and not "At boundary layer sites ...". Maybe it is also better to replace in the sentence before the term "boundary layer site" by "sites which are continuously situated within the planetary boundary layer".
- Page 27744, Line 28: As mentioned above, the f(RH) is a model prediction and not a measurement. Please replace this word by "calculated" or "predicted". See also Page 27745, Line 27, and Page 27746, Line 3.
- Page 27745, Line 2: The f(RH)-values from Ispra by Adam et al. are no direct measurements, but are rather calculated (taking different assumptions into account; similar to this work). This is should be clearly mentioned (or better removed) here and in Fig. 5.
- Page 27748, Line 1: Repetitive to the sentence before.
- Fig. 1: It should be "autumn" in the legend.

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- Fig. 5: This figure is hard to read and also misleading. The single points were extrapolated assuming the validity of the κ-approach and by applying the Mie code using the measured size distribution and HTDMA data (which, as shown in the way here, is definitely questionable for the marine cases, because it does not capture the course of the upper branch of the hysteresis curve correctly!). The figure could be improved by removing the κ-extrapolations and by just showing (as a box plot for example) the average values for the different sectors in addition to literature values (which should also be improved by adding more recent studies). See comment above.

References:

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- Quinn, P., Coffman, D., Kapustin, V., Bates, T., and Covert, D. (1998). Aerosol optical properties in the marine boundary layer during the First Aerosol Characterization Experiment (ACE 1) and the underlying chemical and physical aerosol properties. Journal of Geophysical Research: Atmospheres (1984–2012), 103(D13), 16547-16563.
- Russell, P., Redemann, J., Schmid, B., Bergstrom, R., Livingston, J., McIntosh, D., Ramirez, S., Hartley, S., Hobbs, P., Quinn, P., Carrico, C., Rood, M., Öström, E., Noone, K., von Hoyningen-Huene, W., and Remer, L. (2002). Comparison of Aerosol Single Scattering Albedos Derived by Diverse Techniques in Two North Atlantic Experiments. J. Atmos. Sci., 59, 609–619.
- Zieger, P., Fierz-Schmidhauser, R., Poulain, L., Müller, T., Birmili, W., Spindler, G., Wiedensohler, A., Baltensperger, U., and Weingartner, E. (2014). Influence

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Interactive comment on Atmos. Chem. Phys. Discuss., 14, 27731, 2014.