

Long term measurements of optical properties and their hygroscopic enhancement” by M. Hervo et al.

Answers to Referee #1

The authors would like to thank Referee #1 for his detailed review. To fully answer Referees comments and to make them easily accessible, a supplementary material was written.

The comments from the reviewer are underlined.

1. Trajectory analysis.

- a. 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.
 - b. Please add a figure on the spatial surface residence time retrieved from the trajectory analysis.
 - c. What exact life time (here called “decreasing weight”) was assumed?
 - d. Was precipitation and thus wash-out taken into account?
 - e. 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.
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- a. We do not quite understand the question on clustering since we did not calculate any backtrajectory clusters (maybe the referee can reformulate his concern about this specific point). Concerning the BL/FT segregation, no distinction between residence in the BL or the FT was made along the backtrajectory, for several reasons. First, at Puy de Dôme, we have a mixture between BL and FT air masses but it is difficult to assess where the station exactly lies due to topographic effects that are not well captured by models such as Hysplit. In fact, we believe that the Puy de Dôme measurements can be representative of the atmospheric column,
 - b. Please find below a figure showing the “spatial surface residence time”. This map shows the number of back trajectory points per 5*3° rectangle. Results are shown in log scale and interpolated. The back-trajectories are weighted to give more influence to back-trajectories close to the station. The weight is varying linearly from 1 for the first point of the back trajectory, to 0.5 for the last one. This method was already used and described in Asmi et al. 2012.

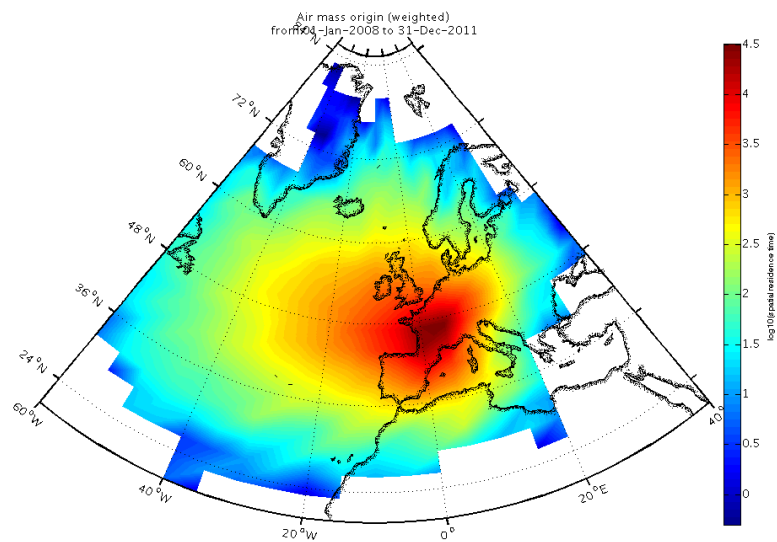


Figure 1: Number of back trajectories points

As all back trajectories converge to the measurement point, the maximum value is around the PdD. Most of the back-trajectories comes from the west and are classified as oceanic air masses.

- c. The following sentence was added in section 3.2 to describe more precisely the weighting process: “ *The weight is varying linearly from 1 for the first point of the back trajectory, to 0.5 for the last one as already described in Asmi et al. 2012*”
- d. Air masses that had experienced washout along the backtrajectory path were not separated from the rest of the backtrajectories. On one hand, the prediction of rain along the trajectory path is not completely trustworthy with Hysplit and on the other hand, we want to characterize aerosols within different air mass types including the processes that are inherent of these air masses. Filtering air masses that have experienced rain would include a bias in the statistics toward anticyclonic situations compared to low-pressure systems. Moreover, we believe it is important to stay consistent with the seasonal analysis and no filtering on the rain occurrence was applied to the data discussed in the previous paragraphs of the paper. At last, a rapid study suggest that this washout effect does not change the main conclusions of the paper:

When filtering all measurements where the integrated rain along Hysplit backtrajectories is higher than 2mm, the absorption coefficient distribution according to the air masses origin is very similar compare to the one without rain filtering.

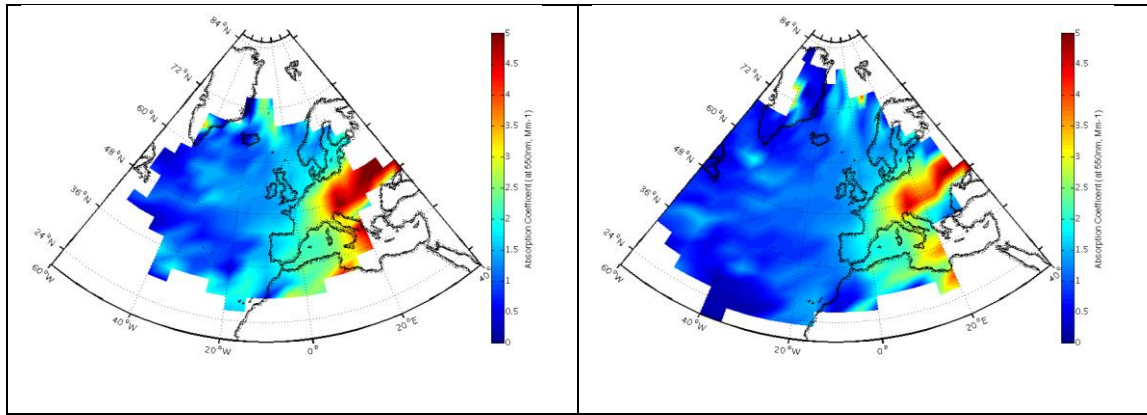


Figure 2: Map of the influence of the air mass origin on the absorption coefficient at the PdD. Left with rain filtering, Right without.

- e. Again, we do not claim to provide BL marine aerosol characteristics, but the whole atmospheric marine column (FT aerosol over the Atlantic ocean are still marine aerosol in our opinion). The low values of single scattering albedo and asymmetry factor can be linked to the low aerosol concentration transported at PdD. Andrews et al. (2011) and Delene and Ogren, (2002) clearly shows a bias at low scattering coefficient on the single scattering albedo or high hemispheric backscatter coefficient (used to calculate the asymmetry factor). Similar results are measured in Sherman et al. (2014). Andrews et al. (2011) suggests that the low ω_0 values for low aerosol concentration are induced by an aerosol mixture in which large scattering aerosol particles have been preferentially removed (e.g., by cloud scavenging and/or deposition), leaving behind a more absorbing aerosols.

2. Retrieval of the refractive index.

- a. 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.
 - b. 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 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.
 - c. 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.
- a. If the Real refractive index is changing but complex refractive index is stable, the single scattering albedo will change. It is what was observed at PdD: the scattering aerosols are more variable than the absorbing ones.

- b. An example of the surface size distribution merged from SMPS and OPC GRIMM measurements is presented in the supplementary material. All diameters from the SMPS are used [10 400nm]. The two first diameters of the OPC Grimm are ignored (350nm and 450nm), because they were considered less accurate than the SMPS size distribution over the common size range. The other diameters are used [575-1750nm]. The uncertainty on the OPC GRIMM measurements are difficult to find in the literature, and no calibration procedure exists in term of number particle concentration. In the supplementary material, we tentatively attribute uncertainties on the OPC number concentration and size and use known uncertainties on other parameters to calculate a propagated uncertainty on the calculated refractive index and hygroscopic enhancement of the scattering coefficient. Moreover, in answer to the next comment, we compare calculated and measured wet scattering coefficients for marine air masses and find a good agreement, giving us confidence on our calculation and on the accuracy of the size distribution measurement. According to Burkart et al. (2010) the number concentration derived from a SMPS and an OPC Grimm in a common range (300-1000nm) agrees within 10%. At PdD, there is only one diameter in the common range. Therefore this measurement cannot be used for uncertainties estimation.
- c. The reviewer is right, the comparison with AERONET was removed and will be detailed more deeply in another study. The goal there was to show that we had good orders of magnitudes for the retrieved m .

3. Calculation of wet optical properties and importance of the coarse mode.

- a. 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.
- b. In addition, it has to be clarified if an internal mixture was assumed and which refractive index is taken for the calculations.
- c. Why was only one dry diameter of the HTDMA being used?
- d. 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 separated.
- e. 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 longrange 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 -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.

- a. As suggested by Referee #1, an analysis on the validity of our method was missing. In the supplementary material, we now add a section describing the errors associated with our methodology. The sensitivity study suggested that the uncertainties are lower than 26%. Additionally, we now add in the supplementary material a comparison of our calculated wet scattering coefficient with direct measurements. It was performed during a short period at the station in 2011. The comparison showed that the difference between measurements and calculations was lower than 17%, even in case of oceanic air masses.
- b. As now better described in section 2.2, neither mixing state nor aerosol refractive index was assumed. The mixing state was measured by the HTDMA. If the aerosol is mixed internal, only one hygroscopic mode was used. In case of external mixing, up to 3 modes was used. The refractive index was retrieved by an iterative method as described by Raut and Chazette (2007) and shown on the schematics of the method in the supplementary material.
- c. The following statement was added in section 2.2:

“The HTDMA was measuring at the six dry diameters: 25,35,50,75,110 and 165nm. Lower sizes (25 to 75nm) have a limited impact on optical properties and were not used. At PdD, GF at 165nm is, in average, very close to the GF measured at 110 nm (respectively 1.39 and 1.43 Holmgren et al. 2014). In 2010 and 2011, more measurements were available at 110nm than at 165nm. It can probably be explained by the larger number concentration at 110nm (Venzac et al., 2009).”

- d. From our point of view, using the word “prediction” would not represent the accuracy of our methodology. This study is using measurements of 5 state-of-the-art instruments and an accurate optical model. The word “calculation” is intensively used in the paper to avoid confusion with direct measurements.
- e. Indeed our methodology doesn’t take into account deliquescence for marine cases. However:
 - In Melpitz, for sea salt influenced air masses, the maximum effect of deliquescence on f is around 25% after a long-range transport ($f \approx 1.5$ instead of 1.2 at RH=55% according to Zieger et al. 2013, figure 5a). To our knowledge, deliquescence was never observed at altitude stations.
 - Even in case of deliquescence, we assume that the error that is done for RH smaller than the deliquescence RH (75% for sea salt) is very small due to the low HGF measured below this RH. The error is anyway smaller if the parameterisation is used instead of using no parameterisation at all.

- At last, reviewer 2 believes that the parameterisations are useful and one of the main goal of the paper.

Therefore, we believe that the parameterization should still be given for marine air masses, but we now stress the lack of deliquescence effects in the text, in the legend of the figure and in the table, in order to make sure that eventual users are aware of this approximation.

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?)

Figures 2 and 3 present different properties averaged over 3 hours. The values measured between 00:00 and 02:59 are represented at 00:00, and the values measured between 21:00 and 23:59 are represented at 21:00. Thus no values are missing. This approach is common in the literature (e.g. Pandofi et al. 2014). However, the figure's legend and the text have been updated to highlight the 3-hours resolution.

Concerning the percentile values of the BLH, the reviewer is right and there was a bug in the 25th percentile calculation. The figure was updated.

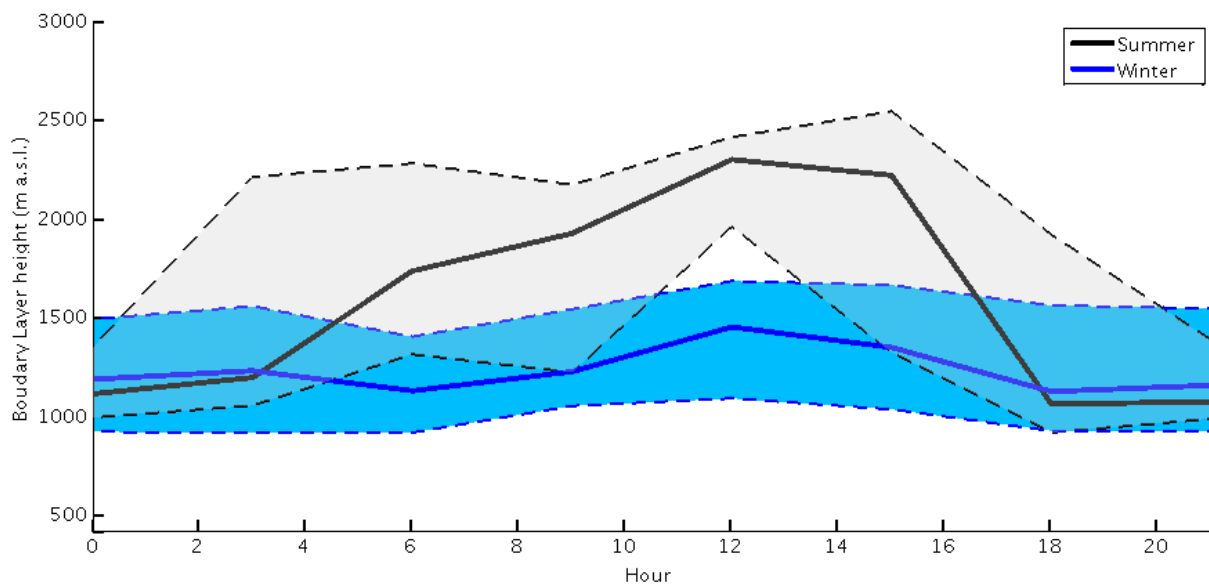


Figure 3: Corrected Mean planetary boundary layer height calculated from Lidar data of 2011.

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 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.

The word “aerosol” was added to the title. From the author point of view, and as suggested by Referee #2, the word “estimated” is not necessary to represent the work done in this paper. However, we now stress more the fact that closure studies indicate that measured f are in fairly good agreement with calculated ones in the literature (e.g.), and also as observed for a short period of f measurements performed at Puy de Dôme.