

We thank the two reviewers for their thoughtful comments and recommendations for improving the manuscript. Below we have listed each specific comment (in italics) followed by our corresponding response to that comment (in bold). We hope that through the expansion of our discussion, additional references, and further analysis we have addressed the general comments of the two reviewers. We feel the manuscript has been strengthened significantly with this substantial revision.

Reviewer 1)

*Major comments*

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*Section 2.1: Please precisely describe what kind of common inlet (any size cut?) was used at the station and if driers were used before the aerosol was measured in the nephelometer. The losses within this inlet have to be determined using particle loss calculations and this information should be included in the paper. I have doubts that you will measure PM 10 particles if for example you are using 1/4-inch tubing in front of the nephelometer even with just a few bends.*

**We added more specificity to the inlet design and it's calculated cutoff (10um). We had stated previously that the air was heated before the nephelometer measurements to dehydrate the aerosols. More detail to this is provided in the response to Reviewer 2 who asked more extensive questions about the RH.**

*Section 2.2.3. and Equation 2: It should be clarified if the AOD is calculated by using the number size distribution taken from the same AERONET retrieval, as described here (also I can't find the actual discussion in the results section). From my point of view, it does not make sense to calculate an extensive parameter using the columnar size distribution which was retrieved from the same (extensive) measurement to which the result will be compared to. This is a circular path and is therefore not valid.*

**We did calculate the scattering AOD as described. The calculation was done so that we could size segregate the total AOD into the portions due to particles greater than and less than 1um, to create an "intensive parameter" for the column aerosols. We added a statement to clarify this point. We then compared this parameter with the surface scattering as measured with the nephelometer. Nowhere in the paper, is this calculated value compared directly with the AERONET AOD, which we agree would be circular.**

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*Page 1796, Line 10: As stated here, the absorption coefficient was also measured at the site, but is neglected when comparison is made to the ambient extinction coefficient. This is not justified and the absorption should be accounted for when comparing the in-situ value to the ambient extinction coefficient. Within the Mie calculations, I also assume that you have included the imaginary part of the complex refractive index. Please clarify.*

Because our site typically had very low aerosol optical depth, we never had sufficient aerosols to accurately derive the imaginary part of the index of refraction from the AERONET inversions (this requires optical depths >0.4). Since the imaginary part of the index of refraction determines the absorption coefficient of the particles, the calculated absorption coefficients for the column are associated with large uncertainties. Consequently, comparisons between these calculated coefficients and those measured at the surface would have provided no relevant insight (as stated in the paper). In general, by combining our absorption measurements with the nephelometer measurements, we have found that at this site, our single scattering albedo (SSA, the portion of extinction that is attributed to scattering) is almost always greater than 0.90. While absorption is an important characteristic of aerosols, ignoring its contribution to extinction causes a relatively small (<10%) effect. But we have mentioned this possible bias in the relevant sections.

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*Sect 2.3.2.: A discussion on the uncertainty of the MPL system and the Klett-inversion is missing and should be added.*

**We have added a sentence pointing to a reference where the uncertainties and limitations for the lidar inversion have already been discussed. (Welton et al. 2000). We also added a sentence about the possible errors because it is a column averaged lidar ratio.**

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*Please precisely describe what kind of linear regressions were performed within this work. The presented regressions do not appear to account for the uncertainty in the x-direction or include any weights/uncertainties of the different data points. I have the impression that the negative slope of the linear fit in Fig. 6 is mainly driven by a few outliers, which is underlined by the low correlation coefficients. In general, it would be better to show squared correlation coefficients within the entire paper. The correct regression-technique is especially important for the fit presented in Fig. 1b. Here, it seems that the slope is as well biased by a few large outliers. A bivariate weighted fit has to be performed here as well.*

**We have added  $r^2$  values throughout the paper. We describe the type of fit we now have done, where it allowed weighted uncertainties for each point in both x and y. When this was done, the old figure 6 made little sense so we took it out.**

*The conclusion demonstrates in its shortness the moderate scientific gain of this manuscript. It is not clear to me what we can actually learn from this work and what the real take-home message is. Quotes like "highly correlated" are not justified if you consider that the best  $R^2$  observed in the work is about 0.42.*

**We added  $r^2$  throughout the paper, and have significantly rewritten the conclusions. Most  $r^2$  were  $>0.5$ .**

*The lidar data and radiosonde data could be used more intensively. For example by systematically looking for elevated layers or by investigating the effects of hygroscopic growth at elevated layers. Thus, the radiosonde data should be used during the analysis of the lidar data.*

**We did not launch our own radiosondes during this experiment, but had to use archived data from a nearby site (as described in the paper). While we could use this data to estimate a lapse rate and evaluate the associated stability, only a few points were available within the lower km. The vertical resolution was really not sufficient to add anything significant to the lidar data. Even with higher resolution radiosonde data and the lidar profiles, differentiating between hygroscopic growth in an elevated layer and a simple increase in aerosol density would be problematic.**

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*As mentioned above, the amount of figures is quite excessive, especially the lidar profiles (Fig. 2 and 3). Further, the many unneeded time series (Fig. 5a, Fig 7a and especially Fig. 8) can be combined and/or removed. They are often just discussed with a single sentence.*

**We have reduced the number of figure panels from 13 to 7 and eliminated most of the time series figures.**

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*Figure 9: Could the differences between MPL and in-situ be explained by systematic errors in in-situ (particle losses) or the lidar (Klett inversion)? The rather constant ratio for the low RH below 70% is striking and would point towards this direction. As mentioned in the manuscript, one would expect an increase of the shown ratio with increasing RH, however, a decrease is seen from 80 to 90% RH. I would guess that this is caused by the low amount of comparable cases (as given in the Figure). At high RH, the MPL retrieval could also be influenced by small and thin clouds.*

**We no longer have this figure in the paper.**

*The work could be improved by adding a substantial amount of further analysis. A few ideas:*

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*Further and more intensively using the radiosonde and lidar data to investigate the effect of hygroscopic growth within the entire planetary boundary layer (PBL) and not just the lowest point at 400 m.*

**As mentioned above, without having an independent measurement of the scattering properties for dry aerosols, distinguishing between a hygroscopic growth effect and a difference in aerosol concentration at that height is problematic. That is why we**

**chose the lowest point in the lidar profile, as close as possible to where we have the nephelometer measurement at a different (dry) RH.**

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*The trajectory analysis could be used to investigate the differences found in Sect. 3.4 when the lidar extinction coefficient is being compared to the in-situ value.*

**We used the trajectory analysis much more extensively in the revised version.**

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*A marine hygroscopicity of  $f(RH)$  could just be assumed or calculated (if chemical proxy data is available) and included in the analysis.*

**We evaluated the sensitivity of results to assumed hygroscopicities but this approach did not in reduce the noise or increase  $r^2$  values significantly.**

*Minor comments*

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*The section heading 2.2.1 is not needed and can be deleted, since no 2.2.2 is following.*

**This has been taken out.**

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*Section 2.2.: Please mention the mean RH inside the nephelometer.*

**Now mentioned (<40%).**

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*Page 1804, Line 25: Please state the slope and intercept.*

**Done throughout manuscript**

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*Equation 1: Using the absolute value when calculating the correct scattering coefficient can bias the true scattering coefficient towards larger values. For example, if the nephelometer will result in negative scattering coefficients (which can happen at very low concentrations due to the uncertainty of the internal nephelometer calibration (dark current, offset, etc)). In addition, it should be  $b$  size–samp in the Equation and not only  $b$  size–surf*

**This is correct, we did not use the correct brackets when writing Eq. 1. On the second point, the nephelometer only gives surface values for submicron or bulk scattering. That is why we have said  $b$  size–surf, where  $size$  can be substituted with the appropriate size class, but all measurements with the nephelometer are surface measurements.**

Reviewer 2:

Specific Comments:

1. *The abstract is concise and provides a reasonably complete summary but contains some misleading (although not completely inaccurate) phrases regarding the absorption measurements and the complex refractive index. Absorption measurements were made but only at one wavelength and they were not used in the study so the reference to 'spectral absorption measurements' should be re-phrased or better yet, omitted. Furthermore, there were no days where the optical depth was sufficient for reliable complex refractive index retrievals so the reference to column-averaged Angstrom exponent derived using a column-averaged size distribution and complex refractive index should also be re-worded to more accurately describe the study.*

**While the absorption measurements were done spectrally (350-1100 nm, every 10 nm, see Aryal et al., 2014, J. Atm and Ocean Techn), we agree that they were not used, because we did not have a corresponding column measurement with which to compare, so we removed mention of absorption in the abstract. We also removed the word “complex” as this was not reliably retrieved because of the low aerosol optical depth at the site.**

2. *The Introduction section is brief and consists primarily of a few generic comments regarding well-known aerosol impacts on climate and the utility of co-located surface in situ and remotely-sensed aerosol measurements. No mention is made of results from any of several similar studies conducted in other regions to assess the representativeness of the surface-based aerosol measurements. A few examples are (i) Quinn et al, JGR-2004; (ii) Sheridan et al, ACP-2012; (iii) Andrews et al, JGR-2004. A reference to the Voss 2001 paper and a few others is included along with obvious comments regarding paired measurements of optical properties but no mention is made of any conclusions from these studies or how the current work will add to these results. For these reasons, the Introduction section requires significant revisions to place the current work in the context of previous works and to emphasize the novel contributions of the study to the current state of knowledge.*

**The introduction has been expanded to include additional information from previous investigations of column vs surface measurements.**

**(The following comment has many parts, and we will address them separately)**

3. *The discussion of near-surface aerosol light scattering measurement (section 2.1) and the air sampling was lacking in some key areas. (a) The internal RH of the nephelometer used to make the scattering measurements was never mentioned and is critical to*

*interpreting the results. The authors do state that the air was heated to temperatures of  $28\pm 50^{\circ}\text{C}$ , which could lead to a very wide range of RH values. Was there any sort of RH control employed? My guess is that at these temperatures and at ambient temperatures and RH typical of the region, the internal RH could often be significantly above 50% and at the very least can take on a wide range of values. This in turn results in a poor assumption of dehydrated aerosols for several aerosol types, especially for air masses with continental influence, where aerosols often exhibit a slower (but non-negligible) scattering versus RH portion of the growth curve for RH  $\sim 30\text{-}60\%$ .*

**RH was measured within the nephelometer and all but 9 individual RHs were less than 40%. Removing these data yielded no significant change in any of the results. A statement about this has been added to the paper.**

*(b) Insufficient information was provided regarding the sampling infrastructure. Unlike NOAA-GMD sites, which all employ the same instruments (TSI nephelometers, Radiance Research PSAPs), tubing diameter/type, impactors, etc., the setup at the site of the study does not appear to follow standard protocols with well-estimated particle losses and uncertainties. For this reason, details regarding air sampling should be provided.*

**As described in the manuscript, the sun photometer and the micro-pulsed lidar were deployed and operated as part of the AERONET and MPLNET programs, respectively. Performance of these instruments and the quality of resulting data are well established as described in the cited literature. We have added two references relevant to the nephelometer. Li-Jones et al. (1998, JGR) report additional information regarding performance and calibration of an identical instrument operated at Barbados. Hygroscopic growth factors for mineral and marine aerosol inferred from measurements with this instrument are consistent with published results based on measurements using other instruments, which implies that performance is comparable. In addition, Moody et al. (2014), report comparisons of scattering measured at Bermuda using the same measurement technique with published measurements reported elsewhere in the North Atlantic region. Results are reasonably consistent although differences in reference RHs, wavelengths, space and time preclude direct intercomparisons. Additional details regarding the design and passing efficiency of the plenum have also been added to the text.**

*4. The methodology for Mie-based scattering calculations in section 2.3.3 is mystifying. The authors applied size distributions from AERONET to the Mie calculations to calculate column-averaged scattering contribution to AOD, which would be acceptable if not for the fact that the resulting scattering was used for comparison with AERONET AOD. The use complex refractive indices derived by AERONET that are highly uncertain at the measured AODs is questionable, at best. The authors did acknowledge the high uncertainty but the benefit of the results from the Mie calculations are debatable*

**Obviously this portion on the explanation of the mie calculations was unclear since both reviewers mistakenly inferred that we compared the Mie calculation (resulting from the AERONET inversions) with the AOD (from AERONET). As indicated above, we have rewritten the explanation of the Mie scattering to clarify this point.**

*5. The analysis is often incomplete and in some cases prone to misinterpretation. Take for example the discussion of AOD versus in situ-measured bulk aerosol scattering coefficient in section 3.1. In general, the results exhibit some degree of correlation but this will nearly always be the case. The authors gave one example (March 23) of poor agreement and illustrated the likely reason on for the poor agreement on that day using vertical profiles of aerosol extinction coefficient. This should have been supplemented with a vertical profile of RH derived from the radiosonde to provide context for the lidar-measured aerosol profiles (i.e. Was the deviation due to an elevated aerosol layer or perhaps hygroscopic growth present in a moist layer?). Furthermore, such results were then extrapolated to other periods of poor agreement, using only references to a few other studies conducted in different regions and with little substance. The level of agreement could have been investigated in more detail using, for example, comparisons of the level of agreement for days with no upper level aerosol layers versus that for days with upper-level aerosol layers measured by the lidar. The degree of correlation will also depend on RH inside the nephelometer and the vertical profile of ambient RH. neither of which was discussed, despite the availability of radiosonde-measured RH profiles. These omissions make interpretation of the correlations extremely difficult.*

**As stated above, our radiosonde data was sparse both in time (2 times a day) and in vertical resolution (2-3 points/km). It is difficult to use this for much other than to estimate the ambient RH at the lidar height, which we did and report. We tried various f(RH) formulations using this RH, and none increased r<sup>2</sup> or reduced the noise significantly. Also as indicated above, the reviewer's concerns regarding RH within the nephelometer has been addressed. More generally, we tried to improve the analysis through out the paper.**

*6. The Conclusion section is brief and weak. The main result emphasized was reasonably good correlation between variables that are expected to have a fair degree (on average) of correlation. Deviations were simply attributed to vertical structure in the lidar profiles. The final sentence "The generally good agreement between the paired measurements suggest that, in most cases, aerosol optical properties measured at the surface can be extrapolated with reasonable confidence to the overlying atmosphere" overstates what can be concluded based on measurements of only size-segregated aerosol light scattering at a single wavelength, which may or may not have been conducted at well-known and controlled RH values (not stated).*

**The conclusion section has been rewritten and the RH issue clarified.**

Technical Corrections:

1. Figure(s) 1 are difficult to read, when viewed at 100%. I needed to magnify to 200-300% to clearly see the figure details. Please consider using larger font and perhaps making the font bold to enhance readability, or else increasing the size of the figure.

**The size of Figure 2 (the old Figure 1) has been increased (by reducing from 3 panels to 2), if not sufficient we can work on changing fonts...**