Response to referees for manuscript "Aerosol optical properties within the atmospheric boundary layer predicted from ground-based observations compared to Raman lidar retrievals during RITA-202" by Xinya Liu, Diego Alves Gouveia, Bas Henzing, Arnoud Apituley, Arjan Hensen, Danielle van Dinther, Rujin Huang, and Ulrike Dusek. (manuscript ID: EGUSPHERE-2023-2262)

Referee #2 comments:

The authors present a comparison of backscatter and extinction coefficients as inferred from ground-based in-situ measurements and lidar observations. The manuscripts describe the steps taken to convert in-situ measurements to optical properties and shows findings for a five-month measurement campaign in 2021. Given these specifics, the manuscript would potentially qualify as a Measurement Report. However, I don't find the work to be within the scope of ACP and there are several issues that lead me to recommend rejection:

1. The authors fail to motivate why the scientific community should be interested in this work. Will the method be applied in future analyses of CAELI observations? Can it be adapted to other sites? How is it superior to the traditional approach of just assuming a lidar ratio?

   In response to the referee's feedback regarding the motivation behind our work, we have taken steps to more clearly state the significance and relevance of our study in the revised manuscript, particularly in the abstract. Here, we wish to emphasize several key points why such a study is interesting and worthwhile:

   a. **Novelty:** The methodology developed in this study provides a valuable approach for obtaining vertical profiles of the lidar ratio through in situ measurements and readily accessible ECMWF meteorological data. This is particularly valuable for atmospheric research sites lacking advanced lidar techniques. We show that for realistic aerosol size distributions the lidar ratio is rather insensitive to relative humidity and can be used to evaluate typical lidar ratio assumptions more quantitatively.

   b. **Integration**: Our research contributes significantly to bridging the gap between in situ measurements and remote sensing lidar observations. The methodology provides a good connection from the ground to the lowest lidar profiles, effectively helping solve the overlap issues that are often a challenge in lidar systems.

   c. **Broad Applicability**: One of the key advantages of our approach is its only reliance on routine in situ measurements to derive aerosol vertical optical properties. This principle is applicable to other sites. As part of our future research efforts, we aim to collect and analyse data from additional sites to conduct broader and more comprehensive studies.

   d. **Validation**: While our primary aim was not to apply this method directly to CAELI observations, it nevertheless offers a significant reference values for validating observations made by CAELI or similar instruments.

   Moreover, while the literature suggests that traditional methods can provide reasonably reliable lidar ratio values, the effectiveness of these traditional approaches is inherently limited to sites equipped with Raman lidar measurement capabilities. This limitation is a significant constraint, as not all atmospheric research sites possess such advanced instrumentation. Our methodology offers a valuable alternative for deriving lidar ratio through conventional in situ measurements, significantly broadening the applicability of aerosol research to sites without advanced lidar technology.

2. The presentation is rather unfocussed with an additional 42 figures in the supplement. The authors also mention instruments like the microwave radiometer and the ceilometer that are not really used later in the work. I suggest to identify key messages and trim the presentation accordingly.

We have undertaken a thorough revision of our manuscript to address this issue. We have now put some key information from the appendix to the main text, ensuring that the most critical data and findings are readily accessible. Additional detailed profiles and supplementary data (previously Figure S21-42) have been relocated to a publicly accessible repository (https://doi.org/10.5281/zenodo.11174465), providing an opportunity for interested readers to explore this information further.

Furthermore, we have clarified the role of the ceilometer in our study, specifically its utility in identifying boundary layer heights, which is mentioned on line 193-195 in the revised manuscript.

While the microwave radiometer data were not a primary focus of our investigation, the meteorological data, particularly relative humidity measurements, play a crucial role in our research. These data were incorporated for comparison purposes, underscoring that ultimately, utilizing data from ECMWF is a superior choice, which also broadens the applicability of our model. However, in the revised version of the manuscript, we have chosen to retain only 3 profiles (Figure 6-8) to avoid overwhelming the readers with excessive detail.

3. The presented measurements seem to be quite specific focussing almost exclusively on clean conditions. It would have been nice if there had been an effort to put the aerosol conditions during RITA into a long-term perspective, e.g., using long-term sun-photometer measurements.

In our original manuscript, we discussed two relatively polluted cases and two clean cases to present a balanced view of aerosol conditions. In the revised version, we continue to showcase results under both polluted (originating from the continent, 1st case and 2nd case) and clean conditions (originating from the ocean, 3rd case). While the pollution levels in these cases might seem mild when compared to the more heavily polluted regions globally, they are representative of the pollution encountered in the Netherlands. This is demonstrated in Figure R1. The boxplots below compare $PM_{2.5}$ concentrations from May to October 2021 to the conditions encountered during the two RITA campaign periods (Figure R1(b)) and the lidar case studies (Figure R1(c)). The cases discussed in the manuscript are indicated by dashed grey lines.

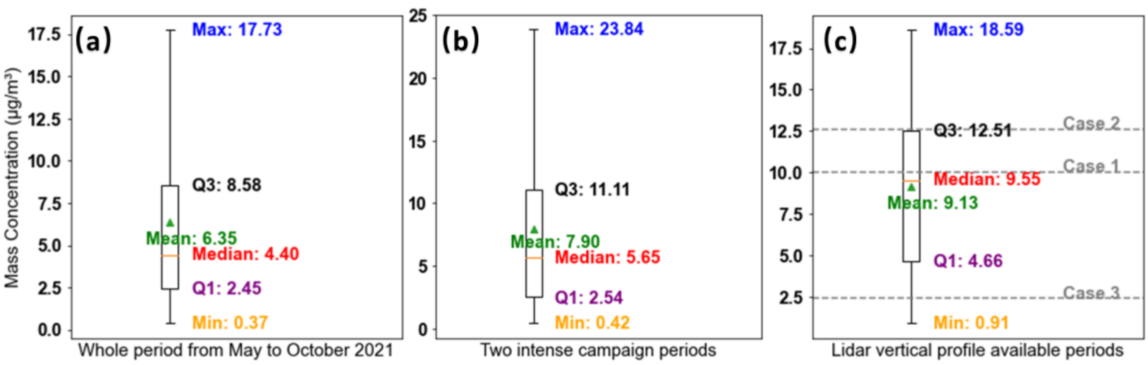

Figure R1: Boxplot of $PM_{2.5}$ mass concentration (a) during the entire period from May to October 2021 with outliers excluded; (b) Two intensive campaign periods (11th May - 24th May and 16th September - 12th October) with outliers excluded; (c) lidar vertical profile available periods with all data included. The three gray dashed lines represent the mass concentrations for the three cases discussed in the revised manuscript.

Among all the lidar measurement periods, there was only one case where considerably higher pollution values were recorded at the surface than in case 1 and 2, namely 18.6 µg m^{-3} as shown in Figure R1(c). This corresponds to the profiles showing below in Figure R2. However, the layer above 1 km was very clean, and only a very small region with a valid lidar ratio retrieval was available, therefore we did not focus on this case. However, within the small region, predicted and retrieved lidar ratio agreed well. This case further underscores the benefits of using in situ measurements to validate the reliability and accuracy of lidar observations.

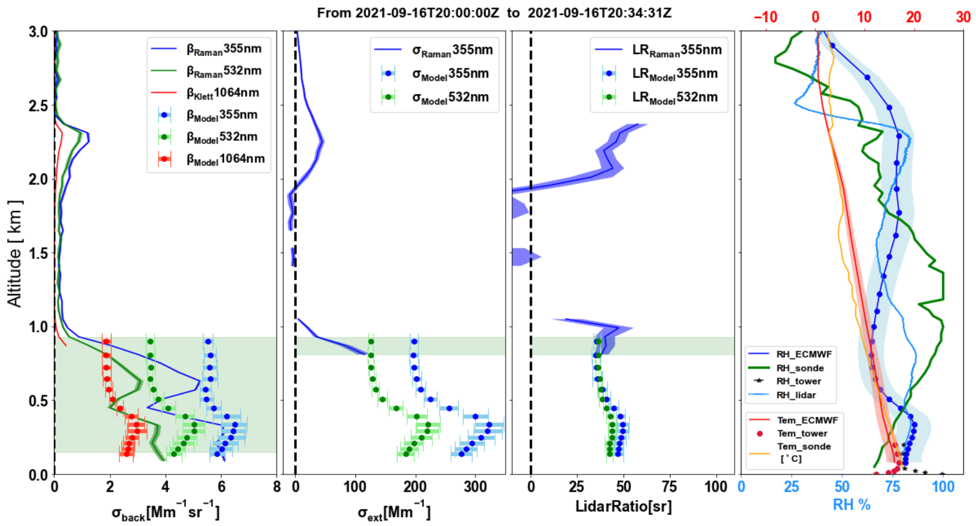

Figure R2: Profiles from 20:00 to 20:34 at UTC time on 2021-09-16.

Regarding the incorporation of long-term measurements to provide a broader perspective on aerosol conditions during the RITA campaign, we acknowledge the value of such an analysis. However, the availability of sun-photometer measurements during the RITA-2021 campaign period was very limited, which constrained our ability to conduct a more extended analysis. Despite this limitation, integrating long-term data into our research is indeed part of our future plans.

4. The authors admit that coarse-mode aerosols have a large impact on the scattering calculations. While this is somewhat minimised by the clean conditions considered in their work, it is likely to be of huge importance during other conditions. In that context, it would have been nice to get some long-term perspective on the occurrence of coarse-mode aerosols. The authors should also mention that Mie theory is inadequate to infer optical properties of dust particles.

In our study the mass fraction of coarse mode ($PM_{2.5}$-$_{10}$) vs PM_{10} aerosols was about 49% on average, ranging from 14% to 81% during the two RITA campaign periods (11th May - 24th May and 16th September - 12th October in 2021). This is in line with previous longer-term measurements of $PM_{2.5}$ represented 50 % of the PM_{10} mass fraction at Cabauw from September 2007 to October 2008 (Mensah et al., 2012). The coarse mode accounted for about 13% of the total light scattering on average (ranging from 0.05% to 94.5%) in our study. Under these conditions, our method can still provide reasonably accurate lidar ratio predictions even in the absence of specific chemical composition data for coarse-mode particles, as long as the size distribution of the coarse mode is known. Although in individual cases, there are sometimes significant differences in predicted and retrieved lidar ratios in the original manuscript (as the dates are indicated in the Figure R3(a) below), we found the coarse mode mass fraction was about 10% to 20% during these specific cases, so the difference were not due to the coarse mode. In fact, the comparison has been improved when we applied the overlap correction function to our retrieved data (Figure R3(b)) in the revised manuscript. This suggests that in our study, the mismatches between the model and the lidar measurement were seldom due to coarse mode but rather to the lack of overlap correction.

This is a significant result and we aim to apply this method at other sites and conduct long-term studies in the future to provide a more comprehensive understanding of aerosol conditions, for which this applies. We expect at more inland sites this result might still be valid, even if the coarse mode makes up a bigger mass fraction, because the chemical composition is typically much less variable than the extreme cases (pure sea salt vs pure mineral dust) we consider for the Cabauw site that is sometimes considerably influenced by marine air masses.

We acknowledge the limitations of Mie theory in accurately predicting the optical properties of non-spherical dust particles. This limitation is an important consideration in future research, especially in regions where dust plays a significant role in atmospheric aerosol composition.

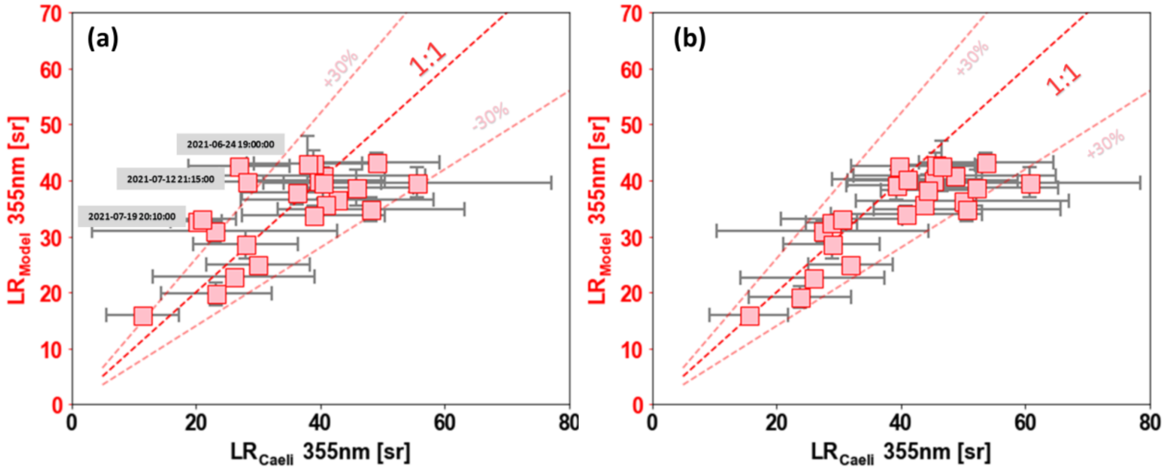

Figure R3: Scatter plot of the lidar ratios from Raman lidar measurements (x axis) (a) original data (b) new data with overlap correction applied, and from calculations (y axis) at 355 nm.

5. The optical profiles inferred from the ground-based in-situ measurements all look like scaled versions of the RH profile. This is not surprising as only RH might give some insight on vertical variation and the authors assume aerosol conditions to be constant with height. It doesn't seem fair that any discrepancies between measured and modelled optical properties are then attributed to errors in the RH profile. The authors should rather find a way to identify a maximum height up to which the vertical extension of ground-based in-situ measurements can give meaningful results. High-resolution sounding profiles could be a source for such an assessment and I wouldn't assume any connection between the ground and above the first inversion - particularly later in the year.

In our methodology and results, relative humidity (RH) indeed plays a significant role in the outcomes of the vertical profiles. However, as stated in our original manuscript, discrepancies between the model and lidar measurements are not solely attributed to variations in RH. Other factors, such as different aerosol layering (line 439-441) and aerosol shape effects (line 426-429 and line 463-466), can also significantly contribute to overestimation or underestimation of the backscatter and extinction profiles. Indeed, these extensive aerosol properties are not directly predicable from ground-based measurements above the mixed layer. Nonetheless, an intensive property, such as the lidar ratio is still comparable between retrievals at around 1 km and the ground, if that height is within the boundary layer or residual layer.

The maximum height up to which the vertical extension of ground-based in-situ measurements (in particular the lidar ratio) can yield meaningful results was determined based on ceilometer data, as described in the original text (line 194-196). In the revised version of our manuscript, we have emphasized this point (line 193-195) to clarify the basis our methodology.

6. The authors overstate their findings. They state "a representative lidar ratio can be estimated based on ground-based in-situ measurements". However, the presented results give the impression that they are by no way superior to an analysis by an average lidar operator. The suitability furthermore hinges on the assumption of vertical homogeneity which - though not unreasonable - should still be supported by some form of measurement. They continue "This allows to extend extinction profiles to lower altitudes, where they cannot be retrieved, or for use with simple elastic backscatter lidar to derive extinction profiles." Again, lidar operators have been doing pretty well with assuming lidar ratios based on experience. The authors would need to be more specific to support their statement. And conclude "The proposed method could be further applied to predict aerosol optical depth and also might be beneficial for large-scale or global radiation simulations." It is quite customary to simply assume constant lidar extinction coefficients from the lowermost trustworthy measurement height to the surface. This approach generally shows good agreement to Sun-photometer observations of aerosol optical depth. This approach also assumes vertically homogeneous aerosol conditions but is much more straightforward than the authors' work.

    In addition to the points highlighted in response 1, we wish to reiterate the significant contribution of our research to the atmospheric science community.

    Our study effectively bridges the gap between in situ measurements and remote sensing lidar observations, addressing a notable gap in the literature where few studies have integrated these two independent methodologies for vertical aerosol profiling.

    Our approach extends beyond the conventional column-integrated optical depth (Sun-photometer observations) closure to provide a broad exploration of vertical aerosol profiles. This allows for a deeper understanding of aerosol optical properties across different atmospheric layers.

    Additionally, we believe that science should embrace multiple perspectives rather than be confined to or satisfied with a single methodology. By integrating different methodologies and independently verifying the results, we can improve reliability of our measurements.

    In conclusion, we believe that the integration of in situ and remote sensing measurements is definitely worthwhile, offering a more accurate assessment of aerosol optical properties and their spatial distribution. This methodological innovation aligns with ACP's scope of enhancing our understanding of the atmosphere's composition and its broader impacts.

7. The choice of references regarding the lidar technique in general and lidar ratios in particular is quite unusual. I suggest to consult with the corresponding co-authors to find more suitable references.

    We have consulted with our co-authors and have revised our reference list (line 45-59) to include more appropriate and widely recognized sources in the field.

References:

Mensah, A. A., Holzinger, R., Otjes, R., Trimborn, A., Mentel, T. F., Ten Brink, H., Henzing, B., and Kiendler-Scharr, A.: Aerosol chemical composition at Cabauw, the Netherlands as observed in two intensive periods in May 2008 and March 2009, Atmos. Chem. Phys., 12, 4723–4742, https://doi.org/10.5194/acp-12-4723-2012, 2012.