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)

We would like to thank the referee for the valuable comments on our paper, we believe that the manuscript has been improved significantly due to their suggestions. To facilitate the review process, we have copied the comments in black text and renumbered them for easy cross-referencing. Our responses are in standard blue text. We have responded to all the comments made by the referee and have revised the manuscript accordingly.

Referee #1 comments:

1 General remarks:

The manuscript discusses an interesting approach and brings together in situ observations of microphysical aerosol properties and chemical composition at ground (at Cabauw in The Netherlands) and optical modeling and comparison of the modeling results with lidar profiles of measured aerosol optical properties. There are many examples of such so-called closure experiments in the literature (since about 25 years), however, such exercises are still needed and thus the manuscript is a good addition to the literature in this field of optical closure studies.

In contrast to the other reviewer, I do not think that the paper should be rejected. It shows the present state of the art when combining ACTRIS observations from super sites… equipped with (a) aerosol monitoring tools and (b) remote sensing instruments. I also do not agree that this manuscript is a measurement report. Closure studies as presented here are more than just observations.

My main point of criticism is the following one: A lot of essential information is given in the rather extended supplement. That means one has to be very ‘active’ as reader and switch from main text to supplementary material and back and so one. A fluent reading is not possible. In the detail section, I will provide a few suggestions how this can be overcome, at least a bit.

Thank you for acknowledging the significance of our study and providing valuable suggestions for improving our manuscript.

In response to your comments, we have streamlined the main text by integrating critical information from the supplementary material directly into the main manuscript, wherever feasible. In addition, in order to minimizes the amount of supplementary material while still providing an access to readers who interested in this information, we have transferred the FigureS21-42 in the original manuscript to a public repository (https://doi.org/10.5281/zenodo.11174465).

Furthermore, we have carefully considered your detailed suggestions and have made corresponding adjustments to further improve and clarify our study.

We hope that these revisions address your concerns and look forward to your further guidance and feedback.

2 Details:

2.1 The Abstract needs to be adjusted after finalization of the revision.

We have carefully revised the abstract to better reflect and summarize the contents of our manuscript.

2.2 p2, l53: Cooney et al. and Melfi references are not appropriate here, in the context of aerosol extinction retrieval… Cooney and Melfi are pioneers in the field of Raman lidar developments because they introduced the temperature and water vapor Raman lidar technique.

We have revised our manuscript to correct this oversight (line 53) and ensure that our references accurately reflect the context of cited works. Thank you for bringing this to our attention.

2.3 p3, l81: be more specific already here, mention time periods.

We have adjusted accordingly and the time period is stated at the beginning of the methodology description on line 81.

2.4 p3, l84: I would prefer to include Figure S1 in the main text, and even Figure S2.

We have combined Figure S1 and S2 into a single figure, which is now included in the main text as Figure 1.

2.5 p5, l144: Avoid confusion (with lidar backscatter at 180°) already in the beginning, mention the angle range directly after … backscatter coefficient (7° to 170°).

We now mentioned the angle range (7° to 170°) directly after introducing the backscatter coefficient, to avoid any potential confusion with lidar backscatter at 180° on line 146. Thank you for highlighting this point.

2.6 p6, l174: When having a near range telescope you should be able to show extinction and lidar ratio values even down to 500 m height (after overlap corrections). And for heights above about 1000 m, you should be able to use the far range observations (after overlap correction) and then we would have much better, less noisy lidar ratios between 1000 and 2500 m height. So, why are these data not included? ….. should be stated…. I would recommend to use your own Raman analysis method instead of using the automated SSA software, and in this way, to optimize the lidar products in these optical closure studies.

We thank the referee for these observations and suggestions. Encouraged by this comment, we investigated the application of an overlap correction to reduce the minimum valid altitude for the extinction and lidar ratios. For that, the overlap function has been determined using the method proposed by Wandinger and Ansman (2002) from measurements with relatively low aerosol loads (ext. coef < 100 Mm-1, clean/no residual layer), and the results were used to reprocess the data. The average overlap correction found agrees with our expectations based on the telecover tests, being < 3% for ranges above ~700m. We found that the overlap correction improved the retrievals, allowing us to reduce the minimum valid altitude for all cases down to 810 m, and also demonstrating a better match with the model calculations. Examples are provided below (Figure R1-R3), showing the comparison of profiles before and after overlap correction with model results. For heights below this altitude, the overlap vertical gradient increases rapidly and the uncertainty of the a-priori lidar ratio becomes more important, even for clean days. To avoid these issues, we keep the minimum valid altitude as 810 m and we have incorporated the reprocessed data to the revised manuscript.

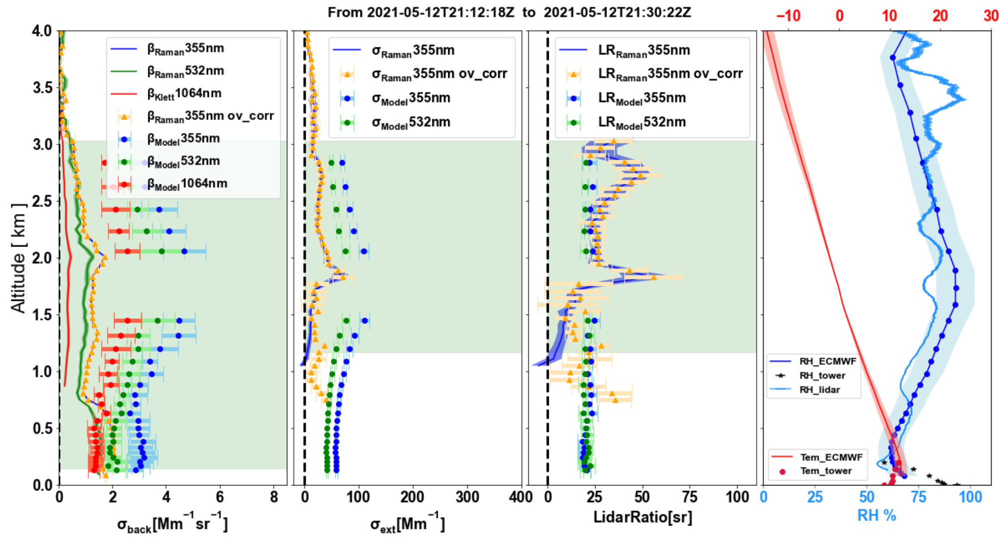

Figure R1: Comparison of profiles before (blue line) and after (orange triangle) overlap correction with model results (blue dot) from 21:12 to 21:30 at UTC time on 2021-05-12.

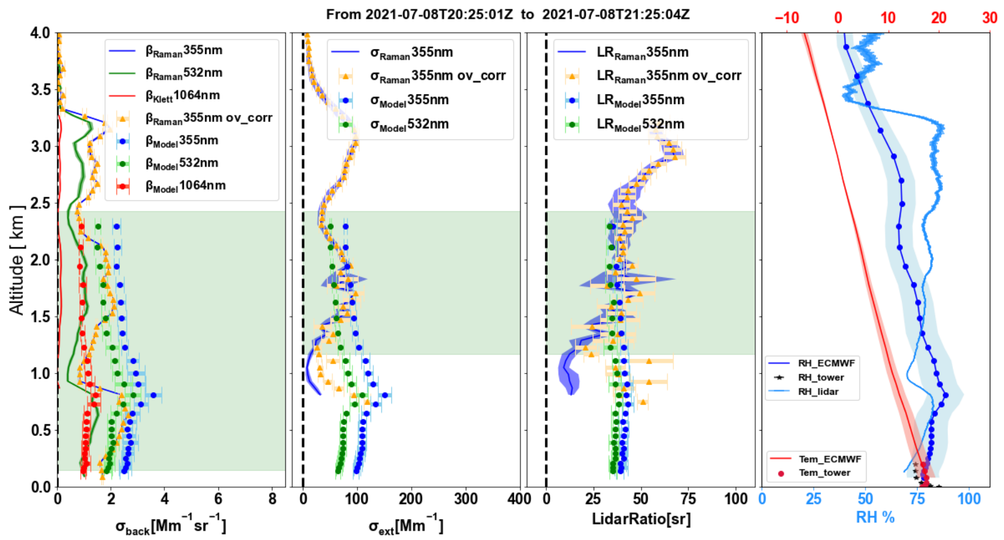

Figure R2: Similar to Figure R1 but showing profiles from 20:25 to 21:25 at UTC time on 2021-07-08.

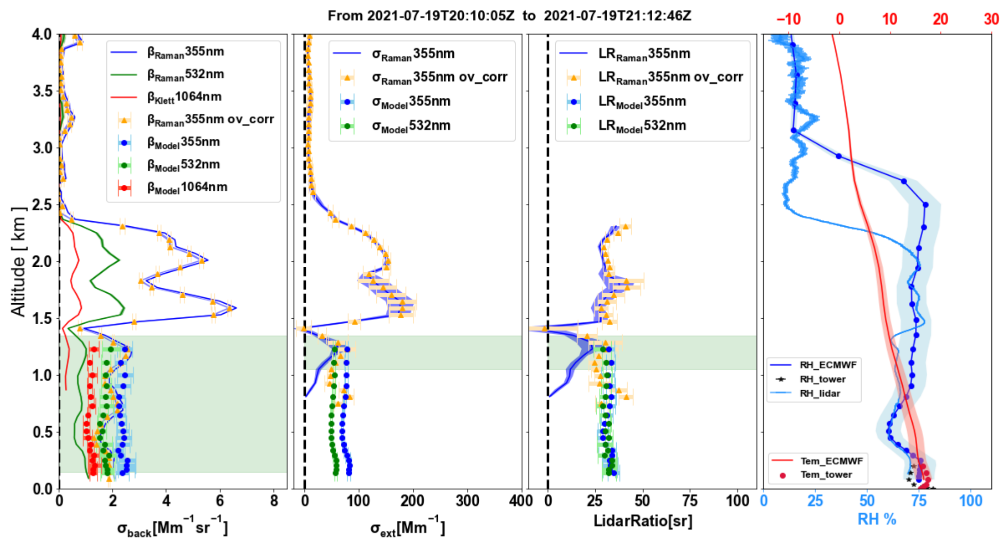

Figure R3: Similar to Figure R1 but showing profiles 21:12 to 22:00 at UTC time on 2021-07-19.

Regarding the far field range telescope (FFR) not being used for the retrievals: The Cabauw station has been prioritizing lidar processing using the Single Calculus Chain (SCC) in a networked effort for centralized, harmonized and quality assured data processing in the framework of ACTRIS/EARLINET. Unfortunately, the SCC cannot yet combine the near and far field telescopes in its retrievals. That is a more practical reason why only the near field range telescope (NFR) was used in the retrievals. Although it is true that the combination of the near and far field telescopes would yield optical products with better signal-to-noise ratios, the retrieved extinction profiles below ~1500 m wouldn't be greatly different at 355 nm and the SNR improvement on the 532 nm extinction would still be insufficient to compensate for the low pulse energy we had for the visible wavelength during the campaign. This reduces the added value of including the FFR signal for this work. For those reasons, we will remain using the Single Calculus Chain for the backscatter and extinction retrievals in the revised paper.

The paragraph describing the lidar processing (section 2.3.1) was changed accordingly in line 178-184 of the revised manuscript

2.7 Another question: What about the Raman lidar observations of the water vapor mixing ratio. In combination with ECMWF temperatures (usually very accurate) one could present them in the panels with ECMWF T and RH profiles. Even during daylight conditions, I could imagine that signals are good enough to show water vapor data up to 1000 m height.

The ECMWF RH profiles are rather uncertain (as usual for modelled water vapor profiles), so one needs more information about the 'real world' RH conditions. I would appreciate, if one shows radiosonde profiles in the respective panels in Figures 5,6,7,8, and if possible the Raman lidar RH profiles. The humidity has such a large and critical impact on the modelled optical properties, one needs to show better RH values, even if ECMWF RH values are considered in all the modelling, the reader should know about the quality (uncertainty) of these ECMWF RH profiles.

Thanks for your suggestions. We recognize the importance of accurately representing real-world RH conditions and the critical impact of humidity on modelled optical properties, thus, we have included radiosonde profiles and Raman lidar water vapor mixing ratio derived RH profiles in Figure 6-8 in the revised manuscripts. This addition is aimed at providing readers with a clearer understanding of the quality and uncertainty associated with ECMWF RH profiles.

2.8 Figure 1 is certainly confusing for non-lidar scientists, especially regarding all the vertical white lines up to 5 km height. Is that just noise or is that strong backscatter from clouds…? Furthermore, what do you mean: an overview is given in Figure 1…., when nothing is explained? What is then the message to the reader? The Raman lidar observations need to be better indicated by thicker lines and brighter color, maybe yellow or orange.

We apologize for any confusion caused by the initial demonstration of Figure 1. In response to your feedback, we have revised the description of Figure 1 (line 195-198) to clarify its content and purpose. Additionally, we have improved the visualization of Raman lidar observations within the figure by employing thicker lines and brighter colors.

2.9 p9, l232: I would include Table S3 in the main manuscript.

Response: Thank you for your advice. We acknowledge the significance of the data in Table S3. Accordingly, we have incorporated Table S3 into the main text as Table 1 in the revised manuscript.

2.10 p9, l243: One should better highlight and explain, how the vertical profile is obtained…. Maybe, one should have a subsection (on vertical aerosol profile) , and show a sketch…, showing T and RH profiles, a well-mixed PBL, and maybe even T and RH profiles for a well-mixed layer, i.e. pot temp = const, RH increasing according to mix ratio = const. In addition, the optical properties as modelled at the surface (indicated by a big symbol) should be shown and finally the aerosol extinction profile, that is in agreement with the RH height profile structures.

Such a sketch would support the reader to understand the closure results…. in Figs. 5-8.

Thank you for your suggestion. In response, we have improved the depiction of the calculation flowchart and made a sketch for the vertical profile calculations, now presented as Figure 3 in the revised manuscript. We added a much more detailed explanation of how the vertical profile is obtained, with all the relevant equations along with the reasoning behind it in section 2.4 of the manuscript.

3 Results and discussion:

3.1 I would prefer to start with Figure S1 and S2 in the main text! Four case studies are then discussed. To provide all necessary details (to the field site, trajectories etc…) in the main text, one probably has to reduce the number of case studies. In the case of Figure 5, I would prefer to see in addition Fig. S9 (showing the full advantage of a lidar, clearly indicating many different aerosol layers, rather than any well-mixed layer), Fig. S10, providing information about the chemical composition, and Fig. S11, showing the origin of the pollution. However, we need at least different trajectories at 250 m (representative for surface aerosol conditions), 900 m, and also one for the 1200-2500m aerosol layer.

In this way (Fig. 5, S9, S10, S11), we would have a complete story and could much better discuss the results of the closure study, and why there is disagreement, especially for heights above 1200m.

I also believe that a full set of observed information (including a much better description of the humidity conditions and air mass transport at different heights) will allow a critical and much deeper debate on the applicability of the closure approach presented here and the especially concerning the limits of the approach.

And as mentioned, I would include a nighttime radiosonde RH profile (19 May, 23:30 UTC, Figure S6 shows it), and if Raman lidar mixing ratio data are available even Raman lidar based RH profiles.

Fig. 5: I do not see (a), (b), (c), (d), where did you put/place these letters? If there are only 355 nm extinction and lidar ratio profiles, then one should not show 532nm in the boxes (with line and symbol explanations), and these white boxes should not hide values. This holds for all other figures and panels as well.

Thank you for your comprehensive feedback, which has guided us to make thorough adjustments to present our story more coherently.

Firstly, we have integrated Figures S1 and S2 into the main text within the methods section 2.1 to provide readers with a clearer understanding of the experimental setup from the outset.

Following your suggestion, we have combined the content of Figures 5, S9, S10, and S11 into a single figure (Figure 6 in the revised manuscript). This allows readers to grasp the entirety of a case study without the need to navigate between the main text and supplementary material.

As per your recommendations, we have incorporated the lidar derived RH profile, radiosonde RH profile, ECMWF profiles into a single plot as shown in Figure 6(d) in the revised manuscript. We also added backward trajectories at 3 different heights (Figure 6(f)) for providing a more comprehensive information.

Finally, we have improved the visualization of the original image, adding necessary numbers and preventing valid information from being hidden. Subsequent pictures have also been adjusted accordingly.

3.2 The same for Figure 6, we need in addition, Fig S12, S13, and 14 (with three trajectories) in the main text. And on 9 Sep, it was probably dark over Cabauw at 21 UTC…. so please show Raman lidar RH profiles plus radiosonde RH profiles (9 Sep 23:30 UTC).Now, we can discuss this closure study in very large detail, including the uncertainty in the model results caused by the ECMWF RH profile.

Similar to the previous response 3.1, we have made the corresponding changes in the revised manuscript.

3.3 I would skip the Fig. 7 closure study. There is already the 19 May case, and the lidar ratio shows marine conditions. Figure 8 is nice, could be presented with the figures S18-S20 here in the main manuscript, and again more trajectories for more heights (250 m, 800m, 1600 m) should be shown. Furthermore, Raman lidar and radiosonde water vapor profiles, if available. Alternatively, one could try to combine Figs. 7 and 8 in ONE figure and show only the optical properties, and briefly discuss the results of these closure study.

In the revised manuscript, we have removed original Figure 7 and provided a detailed discussion on only one clean case (Figure 8). The corresponding modifications are in the revised version.

3.4 Figure 9 shows just ONE 532 nm lidar ratio. I would remove this 532 nm value, so that only measured 355 nm lidar ratios are considered in Fig 9a and 9b.

In response to your suggestion regarding Figure 9, we have removed the retrieved lidar ratio at 532 nm to avoid any potential confusion.

3.5 The supplementary material is too much, no reader (except the reviewers) will study all details so one should reduce the amount of figures and plots to an absolute minimum.

Thanks for the suggestions. We have taken steps to streamline the content, reducing the number of figures and plots to an essential minimum. However, considering the potential value of these materials to interested readers, we have relocated the additional content to a publicly accessible repository (https://doi.org/10.5281/zenodo.11174465).

Ref.: Wandinger, U. and Ansmann, A.: Experimental determination of the lidar overlap profile with Raman lidar, Appl. Opt., 41, 511, https://doi.org/10.1364/ao.41.000511, 2002.