Response to reviewer 1:

General Comments:

In this manuscript, the contamination of long-range transported volcanic aerosol particles was captured in the local Planetary Boundary Layer (PBL) above the Ocean Science Center at Mindelo, Cabo Verde using ground-based lidar observations. More specifically, the authors use two case studies, one before (typical local PBL) and one during the 2021 eruption of the Cumbre Vieja volcano at La Palma island which is located at a distance of about 1500 km from the measurement site to showcase changes in the lidar optical properties and therefore demonstrate the importance of volcanic aerosol detection for health related purposes even further away from the volcanic activity. More specifically, the authors support on a single case study and they report the particle extinction coefficient, the particle extinction-to-backscatter ratio and the particle linear particle depolarization ratio at three wavelengths (355, 532 and 1064nm). The authors use auxiliary information to support that the changes inside the PBL are due to the volcanic activity alone using, for example, AERONET data, backward trajectories, fire presence using FIRMS (although not shown) and FLEXPART simulations. Given the need for accurate detection of volcanic particles in the atmosphere and the scarce lidar observations during volcanic eruptions the study has potential but in the current version it lacks scientific interest and clarity. Therefore, the manuscript may be considered for publication after major revisions.

Thanks for the critical comment. We have now changed the focus of the manuscript a bit, so that the focus is more on the novel and unique optical properties rather than on the pollution event itself while preserving all main information. We also have changed the title so that it becomes clear that we focus on case studies. Furthermore, we completely revised the abstract so that the content of the manuscript should be much more obvious. With all these changes, we believe that the manuscript has significantly improved and also its scientific interest and clarity has become much clearer.

Specific comments:

The title is misleading. In the manuscript, two case studies using a PollyXT lidar are shown. One before the eruption at La Palma island and a second one during the volcanic activity. Then, the authors use the two case studies to discuss the changes in the lidar optical properties over the measurement site. Therefore, I suggest choosing a more suitable title including the word case study. Please note and correct throughout the manuscript: The name of the island is La Palma and should not be confused with Las Palmas which is the capital of Gan Canaria.

Thank you for making us realize that the title was misleading. We have now changed our title to "Tropospheric sulfate from Cumbre Vieja (La Palma) observed over Cabo Verde contrasted to background conditions – lidar case study of aerosol extinction, backscatter, depolarization and lidar ratio profiles at 355, 532 and 1064 nm", which reflects better the contents of the manuscript. Additionally, we have also corrected the spelling of "La Palma" throughout the manuscript. Many thanks for this hint!

The abstract should be short, clear, and summarize the findings of the study. As written, the abstract is misleading and lacks scientific importance. It is misleading because, the authors mention the full duration of the volcanic activity, but they do not mention that the findings rise from one individual case study. It also lacks scientific importance because although it is mentioned that a special version of a PollyXT system was used which allowed the calculation of the particle extinction coefficient, the particle extinction-to-backscatter ratio and the particle linear particle depolarization ratio at three wavelengths (355, 532 and 1064nm), there is no mentioning of the 1064nm wavelength which is the added value compared to the standard high-power lidars which operate at 355 and/or 532nm. In fact, there is no comprehensive summary of the findings per wavelength per aerosol optical property. More specifically, the lidar ratio and linear particle depolarization ratio which are of great importance in aerosol classification are absent from the abstract.

Thank you for your critical but constructive comment. We have restructured and shortened the abstract to point out that we performed a case study and not a long-term analysis and highlighted the availability of the measurements at 1064 nm. Furthermore, we added the results, especially for the lidar ratio and the depolarization ratio, at the particular wavelengths.

The authors support the changes in the PBL optical properties during the volcanic eruption to be caused by the presence of sulphate aerosols and they exclude the presence of other aerosol sources using FLEXPART and backward trajectories together with fire location from FIRMS. It would be beneficial to include a FIRMS figure and I was also wondering whether there are in situ observations at Mindelo site to check the presence of black/organic carbon. This will solidify your conclusions regarding the higher lidar ratio observed during the volcanic activity and its origin.

Thank you for your recommendations! We added a FIRMS figure in combination with backward trajectories to the appendix B of the manuscript. We also agree that in-situ observations would be helpful for the argumentation and checked the availability of in-situ data (e.g., in the Global Atmosphere Watch World data center https://ebas-data.nilu.no/*) but, unfortunately, no measurement data of black or organic carbon at Mindelo is publicly available for that time period.*

Then, I really missed a long-term report of the lidar optical properties including the full period of the volcanic activity. The authors advertise in quite a few points in the manuscript that the lidar has captured the full volcanic activity. So, why did you choose to focus on one case study only and not include the full dataset? Furthermore, it will be of great importance to go a step further and estimate the mass concentration of the long-range transported sulphate aerosols.

Thank you for this comment! In this manuscript, we focus on the case study analysis and the respective optical properties. Given the revised title and abstract it should become clearer now. But we are aware that a long-term study of the full period of volcanic activity would be of large interest. Continuous lidar measurements were performed during that time, but a complete quality control data set including successful and reliable cloud screening as a prerequisite for a high-quality aerosol optical data set is not yet available. That is why we, for now, analyzed single days within this period. However, a long-term study based on automatically retrieved lidar optical properties may be subject of a future publication as we added in the outlook. For now, we reworked the misleading paragraphs in the manuscript. Furthermore, we now added a profile of the sulfate mass concentration to Fig. 5 and a corresponding paragraph in lines 347–353 of the revised manuscript to highlight the potential of such lidar measurements for studies of air quality.

Overall, in its current form the manuscript is a report of a single case study in which the optical properties are the result of marine and sulphate aerosol mixture of unknown contribution with limited added value to the scientific community and at places highly speculative. The presentation and usage of English should be also substantially improved.

With the new title and the updated abstract, the focus of the manuscript should be clearer. Several new aspects are considered with the case study: It is the first time, optical properties at 1064 nm for sulfate aerosol are measured. We can clearly disentangle the effect of the sulfate from marine by comparing to background conditions and, thus, show the impact of the volcanic eruption at Mindelo on this specific day. We could show that no ash was transported from the volcano towards Mindelo. Furthermore, we reworked the manuscript linguistically. With all these changes, we believe that the manuscript is of high value for the scientific community.

L122-124: Can the authors comment on the stability of the calibration for this wavelength? What is the expected error in the optical products? The references in this sentence point to another lidar system and not PollyXT. What is the error estimation for this system? Furthermore, the 1064nm depolarization capability is also a new feature. What is the uncertainty of the particle depolarization ratio for this system?

The calculation of the extinction coefficient at 1064 nm via the rotational Raman method follows the methodology described in Haarig et al., 2016. The spectral cross-talk calibration methodology uses a liquid cloud as it was introduced in Haarig et al., 2022 and is referenced in the manuscript but briefly described here for your convenience: the strong backscatter signal at the cloud base is used to iteratively determine the spectral cross-talk correction factor. The elastic signal (1064 nm) multiplied with the spectral cross-talk correction factor is subtracted from the rotational Raman signal (1058 nm) so that the particle induced strong backscatter signal at cloud base (elastic backscattering process) is not contaminating the rotational Raman signal any more. The spectral cross-talk correction factor changes only when the neutral density filters are changed in one of the two channels (either 1064 or 1058 nm). As there was no change in neutral density filters between 24 September and 4 October 2021, the spectral cross-talk correction factor of 6.7e-4 +/- 0.3e-4 is valid for the whole period.

The calibration of the depolarization ratio at 1064 nm and the estimation of its uncertainties followed the same approach as the calibration at 355 and 532 nm (Engelmann et al., 2016). The Delta 90° calibration (Freudenthaler et al., 2009) with a linear polarizer after the pinhole was applied.

We also included a dedicated section describing the uncertainty estimation for all optical products now in the manuscript – see lines 141–150 of the revised manuscript. Following this section, the error for the particle depolarization ratio is about 0.01 at 1064 nm.

L178-179: Is there a reference to support the statement that the measurement on the 16th of September represent the typical situation over the location?

The lidar-derived optical properties have been already studied, quite intensively, in the framework of ASKOS and L2A+ (ESA funded projects). A comprehensive overview of the PollyXT lidar measurements conducted during the ASKOS intensive measurement periods is shown in Fig. S1. The attenuated backscatter coefficient at 1064 nm (Fig. 1a, 1c, 1e) in combination with *the volume depolarization ratio at 532 nm (Fig. 1b, 1d, 1f) reveal the typical aerosol conditions above Mindelo, which are a clean marine boundary layer (MBL; non-depolarizing spherical particles), with a dust aerosol layer (depolarizing non-spherical particles) on top of that.*

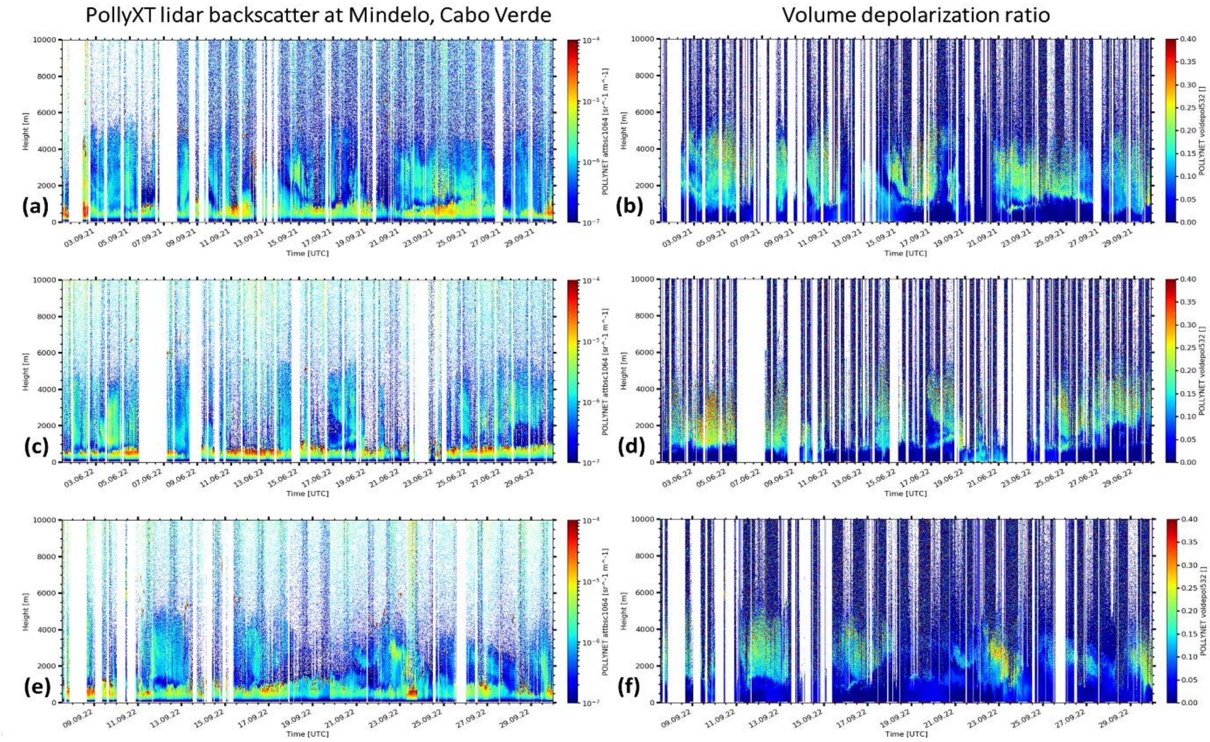

Figure S1: Overview of the lidar attenuated backscatter coefficient at 1064 nm (left column) and volume depolarization ratio at 532 nm (right column) as retrieved from the PollyXT lidar during the ASKOS operations in September 2021 (a, b), June 2022 (c, d), and September 2022 (e, f).

These results have been published (for now) in project deliverables (e.g., https://l2a.space.noa.gr/backend/assets/e7a1b125-e2b2-4489-a205-720bd4f8077a?download, last access: 11 January 2024). Additionally, the data are available in native format via the ESA Validation Data Center (EVDC) under DOI: 10.60621/jatac.campaign.2021.2022.caboverde (2023). We have now included the relevant citations in lines 208–211 of the revised manuscript.

Figures 3-5: What the error bars refer to? Do they include the systematic errors or just the variability caused by the time averaging?

The error bars show the statistical error in case of the particle extinction coefficient and a relative error (minimized systematic error + statistical error) of 15 % for the particle backscatter coefficient. The errors of the lidar ratio and the Ångström exponent were calculated using the error propagation. For the particle linear depolarization ratio, constant absolute errors of 0.02 at 355 nm and 0.01 at 532 and 1064 nm are considered as described now in the manuscript. Thus, minimized systematic errors are included as well as statistical errors. We added a detailed explanation of the uncertainties in Sect. 2.1 (lines 141–150) and clarified the meaning of the error bars in the figure caption.

Technical corrections:

Lines 67-69: Please give a reference.

Done, please see line 70 in the revised manuscript.

Lines 75-86: Note this publication about Cumbre Vieja volcanic eruption:

Bedoya-Velásquez, A.E.; Hoyos-Restrepo, M.; Barreto, A.; García, R.D.; Romero-Campos, P.M.; García, O.; Ramos, R.; Roininen, R.; Toledano, C.; Sicard, M.; et al. Estimation of the Mass Concentration of Volcanic Ash Using Ceilometers: Study of Fresh and Transported Plumes from La Palma Volcano. Remote Sens. 2022, 14, 5680. https://doi.org/10.3390/rs14225680

Thank you for pointing to this interesting publication that was missing in our review! We included it now!

Line 104: Please check that the coordinates of the measurement site are correct. I think it should be W, not E.

We corrected the wrong coordinate of the measurement site as well as of the volcano. Thank you for notifying!

Figures 2-5: Use the same thickness for all lines.

The different line thickness was chosen to satisfy the requirements for the color blindness tests but thanks to your comment we noticed that this solution may be confusing. Thus, we standardized the line thicknesses now.

Figure 3: Be consistent on the heights at which the error bars appear. For example, Figs. 5a, b and c all have the error bar in different locations.

We chose the different heights on purpose to ensure a better visibility of the error bars but we also acknowledge your comment as, of course, choosing consistent heights ensures a better comparability. Thus, we changed it according to your recommendation.

Figure 4d: ref. lines are missing from the legend. For example, the dotted orange line which most probably refers to the bae532/1064 (RR) on the 16th of September.

We added the missing symbols and labels in the legend.

Figure C1: Include the 16th of September 2021 as it is one of the case studies. The figure could also be a bit more zoomed to the region of interest.

Thanks for this suggestion. We have included it now accordingly.