

## Response to reviewer 2:

### General comments:

The manuscript describes the measurements of a volcanic aerosol cloud from the Cumbre Vieja volcano eruption in Sept. 2021 that was transported 1500 km over the Atlantic Ocean from the island La Palma to the measurement site in Mindelo on the Cape Verde Islands. Measurements were performed with a multi-wavelength Raman and depolarisation lidar and a sun- and moon-photometer. Additionally, trajectory calculations are used to verify the origin of the measured air masses in the boundary layer and in the free troposphere. The comparison of the measured values of the optical properties with the ones from a clean reference period a few days earlier and from lidar measurements of other campaigns indicate that the measured aerosol in the boundary layer is sulfate aerosol from the volcanic eruption.

As there are only few high quality multi-wavelength lidar measurements of aerosol stemming from volcanic eruption - especially spanning the wavelength range from 355 nm to 1064 nm, the retrieved aerosol optical properties add valuable data to the global database that can be used for aerosol characterization. The manuscript is very well organized and written.

*We greatly appreciate the review and detailed comments provided. As many comments were made with respect to the uncertainties of the lidar-derived optical products, we now have included a dedicated paragraph on this issue in the manuscript. You can find it in lines 141–150. Our responses to the specific comments are as follows.*

### Specific comments:

Las Palmas is a municipality and city on Gran Canaria. The Cumbre Vieja volcano is located on the island called La Palma.

*Thank you for the correction! This point was also raised by RC1. We corrected the spelling of the island throughout the whole manuscript.*

The dual-field-of-view channels are mentioned in chapter 2.1 but unfortunately not used. Why? And if not used, why mentioning it?

*Thank you for your question. The dual-field-of-view (FOV) channels are mentioned in the manuscript as they are now part of the standard PollyXT setup. Since the dual-FOV polarization lidar technique was implemented only recently (Jimenez et al., 2020a, 2020b) it is not included in the instrument's overview paper (Engelmann et al., 2016). The technique is powerful, allowing the determination of microphysical liquid-water properties, which combined with the lidar-derived aerosol properties can be used for studying aerosol-cloud interactions. Since the scope of the paper is not aerosol-cloud-interactions we did not use the dual-FOV channels, nevertheless, their availability is worth mentioning. We have also included a relevant statement in lines 118–121 of the revised manuscript.*

The absolute scale of the "attenuated backscatter coefficient" shown in the fig. 2 need reference values at reference heights for each plot. This is even more necessary as measurements of two different days are compared (figs. 2c and 2d).

*It is worth to mention that here the calibrated attenuated backscatter coefficient is shown. Thus, it is a quantitative property which can be compared to each other. However, it is clear that different attenuation in lower altitudes leads to different values at higher altitudes even though aerosols properties there are similar. Thus, values are not comparable in terms of backscatter intensity at a given altitude. The aim of providing the plot is to show the vertical structure on the days of interest. We made this now clearer in lines 187–189 of the text.*

Figs. 3e and 4e: the x scales should be the same for an easier comparison of the values in the two plots.

*Thanks for the hint! We changed the x scales of Fig. 4e accordingly to the one of Fig. 3e. For the particle backscatter and extinction coefficient, we explicitly chose different x scales and prefer to leave it like that to provide as much detail as possible for the very different atmospheric conditions on these two days. Instead, we inserted the profiles from the Fig. 3 as reference in grey lines.*

Fig. 5b and 5c: the red 1064 nm (RR) line does not match the description in the figure caption. If an increased smoothing of 742.5m is used for the particle extinction coefficient and the lidar ratio at 1064nm, then there can be only one value in the considered height range between 0.25 km and 1.0 km, but there is a line of values between 0.75 km and 1.0 km.

*The vertical smoothing is performed by using a moving average filter, i.e., a vertical distance of 7.5 m between the data points is preserved. However, for an arbitrary smoothing length “s”, each data point contains information of the height range from 0.5\*s below this point to 0.5\*s above it. Thus, with increasing s, the height, where the profile starts, increases, namely it starts always at 0.5\*s, but then it continues in steps of 7.5 m. For the 1064 RR products, with considering a smoothing length of 742.5 m, the data points of the extinction coefficient and lidar ratio in Figs. 5b and 5c (reaching now from 650m to 800m) contain information of the height range 278.5–1171.5 m. This limitation to this height range is done to exclude effects of the incomplete overlap (lower end) and noise (upper end). Please note the new height range of the 1064 extinctions products (650–800 m instead of 750–1000 m). We found a mistake in our plotting and had to adapt the range accordingly. This correction also led to minor changes with regard to the layer mean value, which we updated as well. Furthermore, we added an explanation concerning the smoothing to Sect. 2.1 (lines 141–150).*

The error bars in figs. 3 to 5 are not explained:

1. What are the error bars showing? Should be mentioned in each figure caption.

*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 in a dedicated section. We also clarified it now in the caption of Fig. 3 and refer to the new paragraph concerning the errors. As we avoided to repeat redundant information, which are the same in all three figures, we prefer to add the explanation of the error bars only to the caption of Fig. 3.*

2. Why does the error not change over large height ranges for e.g. in figs. 3c, 3d, and 3e, etc?

*This point should be clearer now with the better explanation of the errors in the dedicated section in lines 141–150. As the errors provided are relative ones, and the quantities itself in Fig. c,d,e are only slightly changing with height, it appears that the error bars are nearly constant.*

3. Figs. 3e and 4e should have the same x-scale for a better comparison.

*Done.*

4. The error bars in fig. 5b of the 355 nm and 532 nm are unrealistically small.

*We have added now a dedicated section (see lines 141–150) concerning the error calculation in the methodology part. Thus, the way we calculated the uncertainties should be clear. For the extinction, we obtain the uncertainty from the error of the linear fit made to get the deviation in the extinction formula. The resulting error is in the order of  $10 \text{ Mm}^{-1}$  for the plotted height ranges and, thus, seems to be small compared to the large values we measured.*

5. In fig. 5c the error bar of the 1064 nm RR curve is much larger than the layer variability. What does it show?

*The error was calculated using the error propagation and includes the errors of the particle backscatter and extinction coefficient. With the added explanation of the errors in Sect. 2.1 (lines 141–150) it should be clearer now.*

The header of table 1 is a bit confusing:

*We rephrased the header now.*

1. In fig. 5b the values at 1 km height are already less than half of the mean values below. What does then the "edge effect" mean?

*"Edge effect" meant that we did not want to include the transition from the PBL to the lofted layer into the mean values for the PBL (and vice versa). Thus, we reflected that choosing 1 km as the upper boundary for the PBL was not appropriate and we changed it to 800 m. But we concluded, that "edge effect" is a misleading term and rephrased it accordingly.*

2. It is unclear what "standard deviation" means and how it is derived. Is it the uncertainty due to signal noise or the parameter variability over height?

*In this context, "standard deviation" means the statistical error due to the averaging over the layer, i.e., it is the parameter variability over height. As this quantity strongly depends on the vertical smoothing, we now decided to replace these values for the intensive optical properties with the layer mean of the given errors, which were used for the error bars in Figs. 3–5. We clarified this now also in the header of table 1.*

3. Is it possible that table 1 does not show any uncertainties?

*In the table of the preprint, the +- values did not include the uncertainties, which were used for the error bars in Figs. 3–5, but only the standard deviation (see answer above). We changed it now for the intensive optical properties, which do not show a significant vertical variability within a homogenous aerosol layer. For these parameters, the table now shows the layer mean error based on the uncertainties of the error bars. We clarified it in the table and, together with the new paragraph in Sect. 2.1 concerning errors/uncertainties, it should be clear, what is meant.*

4. Why is there no 1064 nm AOD?

*Thanks for this hint. We now added it.*

5. What do the +- values of the AOD mean? Standard deviation?

*The uncertainty values of the AOD describe the parameter variability. The AOD was calculated from the layer mean extinction coefficient while its uncertainty values were derived using the Gaussian error propagation with the standard deviation of the mean extinction as input. With the improved presentation of table 1, it now is clearer.*

The error bars / standard deviation values in the plots and table need a better discrimination and clearer description. This is especially necessary if these values should be used in other studies for aerosol typing.

*Thank you very much for the detailed feedback concerning the uncertainties! Of course, they are of particular importance! Thus, we added a new paragraph to Sect. 2.1 of the revised version of the manuscript, where all errors are explained in detail. Furthermore, we included a description of the used errors to the captions of the respective figures and the table.*

Furthermore, are systematic uncertainties considered?

*We calibrate our system according to ACTRIS/EARLINET standards. Thus, if we aware of any systematic error, we correct for it (e.g., depolarization calibration, polarization effects in the receiver unit). Remaining systematic errors are considered as described in the new paragraph concerning the uncertainties.*

Considering the uncertainties and variability, is the increase of the lidar ratio mentioned in lines 237ff really significant?

*You are right, it is not significant. We rephrased it accordingly.*

In line 293f it is stated about the AE values:

During the measurement period, the values decreased to  $0.7 \pm 0.1$  and  $1.7 \pm 0.3$ , respectively, due to hygroscopic growth of the sulfate particles.

=> It is not clear, why the AE decreases. What is the difference between the aerosol parcels at the start and at the end of the "measurement period" - with respect to particle growth? Why do the latter grow more than the former?

*You are right, it is not clear, why the AE decreases. It was stated like this in the article of Navas-Guzman et al., 2013, but not further explained, how the hygroscopic growth may have*

*changed during the night. To answer your question, we can only speculate. One reason could be that the relative humidity increased during the night or that particles arriving at the end of the measurement period followed a different or longer trajectory so that they were exposed longer to high relative humidity. Also, a changing portion of sulfate particles and sulfuric acid droplets is possible. Either, more sulfate particles, which are the larger ones, are present at the end of the night, or there are more sulfuric acid droplets, which have a stronger hygroscopic change [Navas-Guzman et al., 2013]. Of course, coating and mixing with locally-produced aerosol cannot be excluded, leading to a change in the effective radius, too. As the aim of mentioning these findings was just to give reference values for aerosol optical properties of volcanic sulfate it is not meaningful to extend this discussion in the text. Thus, we clarified in the text the origin of this statement.*

**Some correction proposals:**

Line 28: diameter lower than => diameter smaller than

*Done.*

Line 63: ...which is in the range of  $30 \pm 5\%$  for pure dust ... => at which wavelength?

*Done.*

Line 65: ...lower particle linear depolarization ratio ... => typical values?

*Done.*

Line 66: ; John et al., 2011). => (John et al., 2011).

*Done.*

Line 145: In addition, the add(horizontal) distribution of the volcanic plume was monitored.

*Done.*

Line 150: A time series of the AOD => A time series of the CIMEL AOD at Mindelo

*We clarified that the columnar AOD from the sun photometer is meant and not the lidar-derived one.*

Line 151: the hourly mean AOD was about 0.4. => at which wavelength?

*Done.*

Line 154: AODs, with values close to 1.0, => at which wavelength?

*Done.*

Line 193: than the once measured => ones

*Done.*

Line 195: The lidar ratio at 1064nm is in line with dust observations at Leipzig, Germany => are there no changes due to long range transport to be expected?

*The lidar ratio can be linked to the mineralogical composition of mineral dust, which depends mainly on the source region and does not change much during the transport. At least, previous observations of the lidar ratio at 355 and 532 nm pointed to this dependence (Veselovskii et al, 2020). The dust source region (Mauritania, Algeria or in more general terms Western Sahara) was similar for the observations at Leipzig in 2021 and the ones presented in the current manuscript. Therefore, we do expect a similar behavior for the dust lidar ratio. A comment about the similar source regions was added to the manuscript in lines 223–225.*

Line 207: In addition, vertical smoothing was reduced, which improves the accuracy of the near-field profiles => in which sense does the accuracy improve? The resolution is improved. But for which purpose?

*The resolution of the profiles in Fig. 5 was increased to reduce the overlap effect and to display the profiles down to lower altitudes than in Fig. 3 and 4, where a larger vertical smoothing was required to avoid too much noise in altitudes above 1 km. Furthermore, an improved vertical resolution allows us to better illustrate the vertical variability within the PBL. We clarified this now in the text. But you are right, the words accuracy, precision etc. should be used with more caution.*

Line 278: the measured quantities => Do you mean aerosol "quantities" or aerosol "optical properties"?

*You are right. "Aerosol optical properties" seems to be the more appropriate phrasing. We changed it now, also at equivalent positions in the text.*

Line 321: The intense pollution caused an unusually high AOD of more than 1.0 at different spectral bands. => change "different spectral bands" to "at the smaller wavelengths" - or so.

*Done.*

Line 351ff: " Having measurements at all three wavelengths allows us to get new insights in lidar-based aerosol typing and to enlarge our data sets. The findings of this study provide useful insights on the lidar-derived optical properties of volcanic aerosol."

=> This are diffuse statements. What are the new and useful insights?

*Thank you for this comment. You are right, these sentences were not clearly formulated and are redundant in the current state. We rephrased the paragraph to better point out the benefit of our study. The new and useful aspects of this study are that we provide aerosol optical properties measured with a multiwavelength-Raman-polarization lidar even at 1064 nm, which is a novel feature and rarely done so far. It was the first time that volcanic sulfate could be studied applying this technique. Our findings enlarge the existing data sets and can, thus, help to improve the lidar-based aerosol typing. Besides this aspect, they will also help to improve the quantification and modeling of the radiative effects of tropospheric volcanic aerosol.*