Response to RC3

In the following response, reviewer comments are in black and author responses are in blue.

The paper by Prata et al. presents lidar ratio of stratospheric volcanic ash and sulfate aerosols retrieved from CALIOP measurements; an important quantity for deriving aerosol properties from a backscatter lidar like CALIOP. The paper is well suited for publication in ACP after consideration of the following comments:

General comments:

The description of the used method is hardly to follow for a reader less experienced with this method. The manuscript often refers to former papers for important equations. The chapter should be reworked in a way that all important points are included in this manuscript. It should also be clearly worked out why the a priori lidar ratio which was used for the calculation of the particle backscatter coefficient and the effective two-way transmittance does not affect the retrieved lidar ratio.

Response:

The authors thank the reviewer for this comment. In the revised manuscript, the methods section has been reworked to include key equations and describes the relevant steps needed to retrieve the lidar ratio using the Fernald method. Explicit definitions of the AI and SI have now also been included.

In regard to the *a priori* lidar ratio, the reviewer has misunderstood the retrieval method here. The *a priori* lidar ratio is not used to calculate the particulate backscatter coefficient and effective two-way transmittance. This is because the particulate backscatter coefficient, $\beta_p(r)$, does not appear in the two-component lidar ratio solution (see Eq. (3) of original manuscript). Also, the effective two-way transmittance is measured based on the mean attenuated scattering ratio - and so no *a priori* assumptions of the lidar ratio are required to estimate the transmittance. For the top layer, it is measured as the mean attenuated scattering ratio in a clear air region immediately below the layer.

The iterative lidar ratio solution in Eq. (3) (original manuscript, now Eq. (7) in revised manuscript), however, does require an initial estimate of the lidar ratio to begin the iteration. The choice of the initial lidar ratio will affect the number of iterations required for consecutive solutions to converge. As noted in Fernald et al. (1972), in general, Eq. (3) will converge rapidly but will converge more slowly for very clean atmospheres. In practice we have found that solutions converge rapidly when initialising Eq. (3) with the result of Eq. (7) (original manuscript, now Eq. (15) of revised manuscript).

Minor comments: The mean depolarization ratio for Puyehue in the abstract (0.28) differs from the mean value given in Table 2 (0.29).

Response:

This error has now been corrected in the revised manuscript.

You report about an exponential decay in the mean depolarization ratio for the Sarychev layer with time. Do you see changes also in one or more of the other properties?

Response:

Indeed we do see changes in both the lidar ratio and the layer-integrated attenuated color ratio with time for the Sarychev case study. The attenuated color ratio also decreases with time; similar to the depolarization ratio. The lidar ratio is quite variable showing no significant increasing or decreasing trend with time. We have now made mention of the change in color ratio with time for the Sarychev case in the abstract of the revised manuscript.

Upon revisiting the time series analysis we noticed an error in the code that was used to construct the time series from the lidar data. In the original code, the **cumulative** mean was being calculated for each optical property in the time series. The time of each observation was also, incorrectly, calculated as a cumulative mean, which resulted in the incorrect residence time for each data point presented in Fig. 8 of the original manuscript. In the revised manuscript, this error has been corrected so that the curtain means and root mean square errors are calculated for each CALIOP/AIRS observation and are plotted together with the curtain mean of the time of each observation. Note that we define the 'curtain mean' as the mean of all CALIOP layer optical properties (i.e. S_p, δ_v, δ_p and χ') within a collocated AIRS granule, which equates to a ~6 minute subset of a CALIOP granule. This revision only affects the original data plotted in Fig. 8 of the original manuscript. It also impacts the calculation of the e-folding time of the Sarychev depolarization ratios. We have therefore attached the revised version of Fig. 8 (Figure 1 of this document) below.

Page 4, line 31: At this point 'η' is not defined. Please make sure that all variables are defined when using them the first time.

Response:

The multiple scattering factor is defined on page 4 line 18 of the original manuscript (before the line that the reviewer is referring to). However, in Sect. 2 of the revised manuscript we have been more explicit in defining η:

"We note that the effective two-way transmittance profile, $T^2_{e,\lambda}(0,r)$, is related to the particulate two-way transmittance profile via $T^2_{e,\lambda}(0,r) = T^{2\eta}_{p,\lambda}(0,r)$, where η is defined here as the multiple scattering factor (Platt, 1973)."

Section 3.1: How is the BTD algorithm defined? Please give more information about this.

Response:

The BTD algorithms for the AI and SI are defined as

$$\text{SI} = \text{BT}(1407.2\ \text{cm}^{-1}) - \text{BT}(1371.5\ \text{cm}^{-1}). \tag{1}$$

and

$$\text{AI} = \text{BT}_1 - \text{BT}_2 + \text{BT}_3 - \text{BT}_4 \tag{2}$$

where

$$\begin{aligned}\text{BT}_1 =& \frac{1}{4}[\text{BT}(856.44\ \text{cm}^{-1}) + \text{BT}(856.75\ \text{cm}^{-1}) \\ &+ \text{BT}(857.06\ \text{cm}^{-1}) + \text{BT}(857.37\ \text{cm}^{-1})],\end{aligned}$$

$$\begin{aligned}\text{BT}_2 =& \frac{1}{4}[\text{BT}(964.25\ \text{cm}^{-1}) + \text{BT}(965.04\ \text{cm}^{-1}) \\ &+ \text{BT}(965.44\ \text{cm}^{-1}) + \text{BT}(966.24\ \text{cm}^{-1})],\end{aligned}$$

$$\text{BT}_3 = \frac{1}{2}[\text{BT}(1131.79\ \text{cm}^{-1}) + \text{BT}(1133.96\ \text{cm}^{-1})]$$

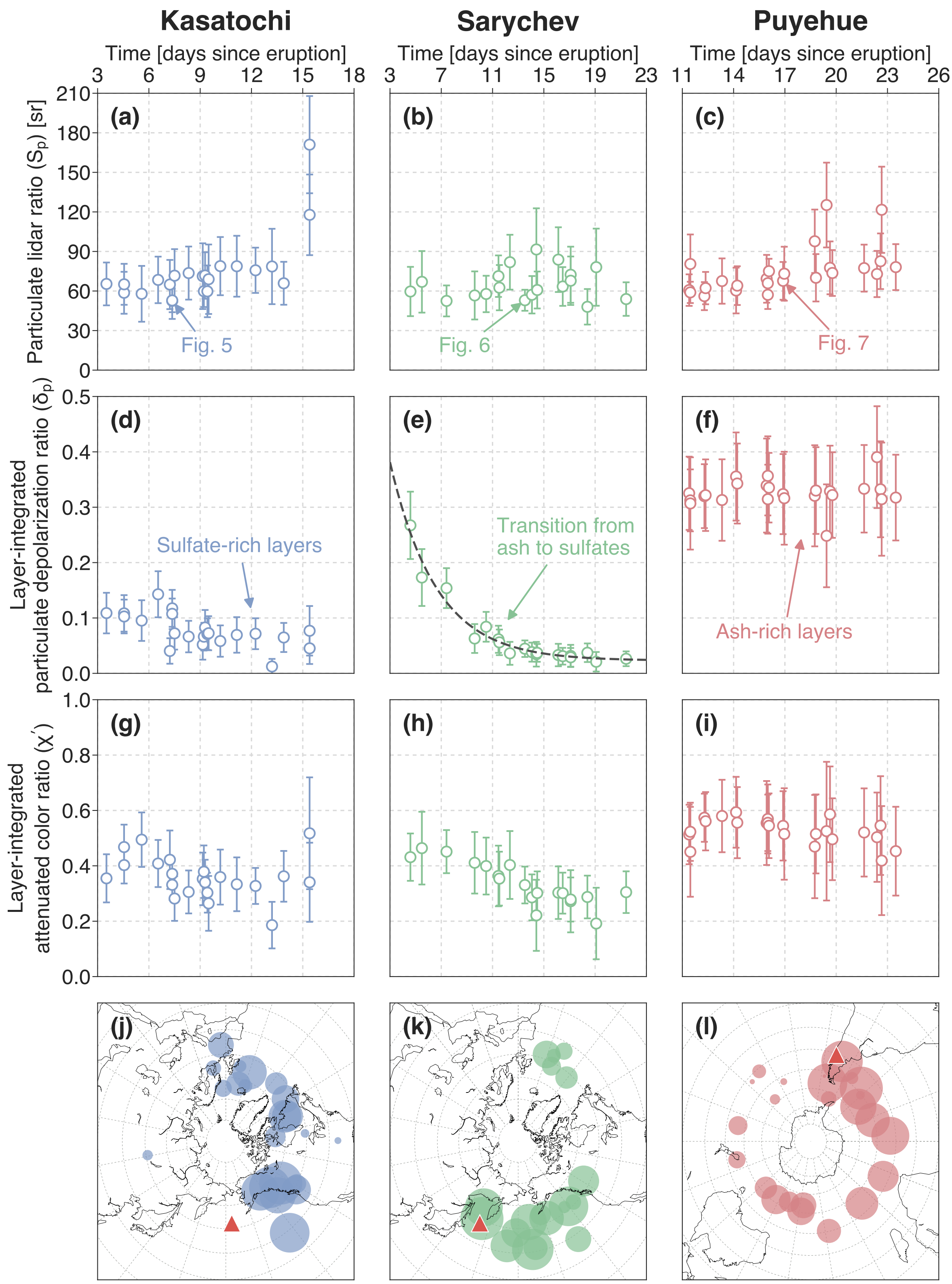

Figure 1: Revised version of Fig. 8 of the original manuscript.

and

$$\text{BT}_4 = \frac{1}{2}[\text{BT}(1080.92\ \text{cm}^{-1}) + \text{BT}(1082.41\ \text{cm}^{-1})].$$

Here BT(ν) is the brightness temperature measured at wavenumber, ν. These equations have been added to Sect. 3.1 of the revised manuscript.

Section 3.1: Why do the conditions differ for the different volcanic layers?

Response:

We assume that the reviewer is referring to the SI and AI threshold conditions. The reason the conditions differ is that we are looking for a volcanic ash signal for the Puyehue case study and an SO_2 signal for the Kasatochi and Sarychev case studies. In order to detect volcanic ash we require that the AI be greater than or equal to 1 K and the SI be less than 1 K to ensure that we are measuring a layer with an ash signal but, importantly, not an SO_2 signal. Similarly, we require that the Kasatochi and Sarychev layers only exhibit an SO_2 signal (SI $\geq$ 1 K) and do not exhibit an ash signal (AI $<$ 1 K). To make this point clear we have revised the relevant part of Section 3.1 as follows:

"For the Puyehue case study, this set of collocated AIRS pixels is scanned for an AI greater than or equal to 1 K and SI below 1 K. These conditions were set to ensure that the volcanic aerosol layers analysed for the Puyehue case study were dominated by an ash signal and, importantly, did not exhibit an SO_2 signal. Similarly, to ensure that observations of volcanic layers for the Kasatochi and Sarychev case studies were dominated by sulfates (and not an ash), the algorithm required an SI greater than or equal to 1 K and an AI below 1 K."

Section 3.2: What is meant by the 'mean scattering ratio'?

Response:

Thank you for this comment. The authors meant to refer to the mean of the **attenuated** scattering ratio profile, $R'(r)$. The attenuated scattering ratio profile is defined as the ratio of the total attenuated backscatter profile, $\beta'(r)$, to the attenuated molecular backscatter profile, $\beta'_m(r)$ (Vaughan et al., 2009):

$$R'(r) = \frac{\beta'(r)}{\beta'_m(r)} = \frac{\beta'_m(r) + \beta'_p(r)}{\beta'_m(r)} \tag{3}$$

For the top layer in a given CALIOP profile, the two way transmittance constraint is calculated by taking the mean of $R'(r)$ in the clear air region immediately below the detected aerosol layer i.e. $T_e^2(r_t, r_b) = \langle R'_{below}(r) \rangle$, where the particulate backscatter is assumed to be zero. The clear air region is defined by the 'clear air analysis depth', which is determined via an iterative process in the SIBYL algorithm (see Sect. 4.3 of Vaughan et al., 2005). This description is included in Sect. 2.2 of the revised manuscript.

Section 4.2: Mean color ratio for Sarychev layer does not agree with value given in Table 2.

Response:

This error has been corrected in the revised manuscript.

Section 4.2: In the Abstract it was reported that the depolarization ratio exponentially decreased with time. This is not reported in Section 4.2. As the change in the optical properties is an important point and thus reported in the Abstract it should also be referred to in the description of the Sarychev layer. How do the other optical properties behave? What are the properties in the beginning of the observations (, mid) and end? This is reported quite lately in the manuscript. To which periods does the mean value correspond to? If the mean values are calculated from the whole period what is the significance of this mean value?

Response:

The authors agree that the decrease in the depolarization ratio with time for the Sarychev case study is an important finding. This finding has now been reported in Sect. 4.2. We have now reported on the change with time (beginning, middle, end) for each of the optical properties in Sects. 4.1, 4.2 and 4.3 in the revised manuscript. The mean values reported in Sect. 4 correspond to the whole time period for each case study. Given the positively skewed distributions shown in Fig. 2, we suggest that the median value is more significant than the mean values given for the case studies considered. This is now stated in the revised manuscript.

Section 4.3: How is a valid lidar ratio profile defined (time/length)? The number of cases/profiles resulting into the mean values should also be given for the other cases. Information about the CALIOP measurements (number, time, days, and location) should be given for the different cases should.

Response:

The lidar ratio retrievals are of a single value for any profile of attenuated backscatter and are constrained by the measurement of effective transmittance. It is not possible to retrieve a "lidar ratio profile" with that single constraint. Valid lidar ratio retrievals are those which satisfy constrained conditions i.e. that are constrained by an estimate of the effective two-way transmittance. We now explicitly define what we mean by 'valid' lidar ratio retrievals in Section 2.2:

"To ensure constrained conditions for the lidar ratio retrieval (i.e. clear air above and below a lofted layer with acceptable SNR), only stratospheric volcanic aerosol layers that had an extinction quality control flag equal to 1, a valid two-way transmittance measurement (i.e. $0 < T_e^2 < 1$) and a horizontal averaging value of 5 km were included in the analysis. We refer to 'valid' lidar ratio retrievals hereafter as having satisfied these criteria."

The number of (valid) lidar ratio retrievals resulting into the means are reported for each case study in the revised manuscript (Sections 4.1, 4.2 and 4.3). The time period for each case study and geographic region/locations analysed are also stated in these sections. The number of layers contributing to the mean geometric and optical properties are reported in Tables 1 and 2 of the revised manuscript and the specific measurement time periods for each case study are discussed Section 6.2 of the revised manuscript.

Section 4.3: Standard deviation of color ratio does not agree with the value given in Table 2.

Response:

This error has been corrected in the revised manuscript.

Section 5.1: What is meant by the aerosol scattering ratio?

Response:

The terms "aerosol scattering ratio" , "particulate scattering ratio", "backscatter ratio" and "scattering ratio" all appear in the literature and usually have the same definition. Here, the aerosol scattering ratio, $R_p(r)$, reported in Vernier et al. (2009), is defined in the present notation as

$$R_p(r) = \frac{\beta_m(r) + \beta_p(r)}{\beta_m(r)}. \tag{4}$$

This is distinct from the attenuated scattering ratio, $R'(r)$, which has not been corrected for molecular, particulate and ozone attenuation. We refer to it as the particulate (aerosol) scattering ratio in the revised manuscript for consistency:

"Vernier et al. (2009) highlighted how this issue would impact the CALIOP calibration region, concluding that undetected aerosols up to 35 km lead to an underestimation of the particulate (aerosol) scattering ratio (an average relative error of 6%), with the effects most pronounced in the tropics (20°N–20°S)."

Section 5.3: Why did you use the error calculation according to Equation 9? This formula is used for the calculation of random (statistical) errors. To my understanding, the errors considered in this manuscript are not random errors and thus the total error should be calculated from the absolute error values of the considered parameters.

Response:
Equation 9 is used in the standard procedure for calculating perturbation errors (see, for example, Chapter 4 of Hughes and Hase, 2010). We consider the errors discussed in Sect. 5 as being systematic i.e. they are errors that are constant through a given profile and cannot be reduced from averaging. This is the same definition used (and explained in detail) in Young et al. (2013). Specifically, we investigate how the errors in different key variables propagate into the lidar ratio ratio retrieval when they are perturbed. If it is assumed that the error in each perturbation variable is uncorrelated then the total error is calculated from the absolute errors by summing them together in quadrature (i.e. the square root of the sum of the squares of the errors). This is because we assume that the total error makes up an error surface composed of the independent component errors. Thus we use Pythagorus' theorem in N dimensions to construct the total error from the component errors (Hughes and Hase, 2010).

Figures 5-7: Please indicate the aerosol free regions below and above the volcanic layer.

Response:
Thank you for this comment. The regions above the layers are assumed to be aerosol free. We account for and discuss errors that may be introduced by this assumption in Sect. 5.2. We have now indicated the clear air regions below each layer on Figs. 5–7 of the revised manuscript.

Section 6.1: The mean values of the lidar ratio for the Kasatochi and Sarychev layers shown in this case studies are smaller than the mean values reported for the whole measurements for these layers. Can you give more information about the changes over the time? Maybe give a time series of the lidar ratio for the different volcanic layers to illustrate the changes and / or variability over time. Otherwise the mean values of the case studies or the mean values over all suffer the loss of significance.

Response:
The purpose of Sect. 6.1 is to give the reader an idea for the spatial variation of lidar ratio across well-defined volcanic ash and sulfate layers. It also illustrates (Figs. 5–7 of original manuscript) the conditions under which the lidar ratio retrievals are successful and how the volcanic layers correlate

with the AI and SI. In the revised manuscript, we have added text to emphasise this point and have also discussed the time of the observations relative to the start of each eruption. We have also annotated where each of the selected observations correspond to on the time series plot given in the revised Fig. 8 (Figure 1 of this document). This figure also shows the time series of the color and depolarization ratios for each the three case studies.

Section 6.2: It is right that the measurements of the different volcanic layers correspond to different stages / ages of the volcanic layer. However the way it is described here could lead to misinterpretation of this information, as one could think that the measurements of the three volcanic layers can be related to each other and show an alteration of volcanic aerosol layers during time.

Response:

The authors did not intend to give this impression. Section 6.2 has been revised to make it clear that the aerosol layers should not be related to each other directly in terms of aerosol evolution. The revised description is:

"As volcanic aerosol layers evolve and disperse into the atmosphere their microphysical properties are expected to change with time. The Kasatochi and Puyehue layers were observable for a duration of $\sim$12 days, while the Sarychev observations covered a time period of $\sim$17 days. Figures 8a–c show that all observations were made more than three days after eruption onset. The Kasatochi and Puyehue volcanic aerosols were observed for a similar time period ($\sim$12 days); however, for the Puyehue case study, the aerosol layers had resided in the stratosphere for more than 11 days before the measurement period began. The Sarychev case study covered the longest observational time period, providing observations of sulfate-rich aerosols for over two weeks. All volcanic aerosol layers were subject to long-range transport across the globe as shown by the spatial distribution of observations plotted in Figs. 8j–l."

Section 6.2: The increase of the Puyehue ash layer with time is small compared to the uncertainties of the retrieved property, thus the statement derived from this changes is very speculative.

Response:

The authors agree. Indeed, the revised version of Fig. 8 (Figure 1 of this document) shows that this statement is even more speculative than first thought. We have therefore removed it from the revised manuscript. The revised statement is

"The particulate lidar ratios for all three case studies were quite variable with time (Figs. 8a–c). Over these time scales (1–2 weeks) it is likely that the volcanic aerosol layers are mixing with ambient aerosol, resulting in fluctuations in the lidar ratio with time. Changes in the lidar ratio may also be a result of sampling different parts of an inhomogeneous aerosol cloud."

Section 6.3, discussion about high lidar ratios for Puyehue: Should not the loss of the large particles also be reflected in the depolarization ratio? No changes are obvious there.

Response:

For the Puyehue case study, the ash layers had already resided in the atmosphere for $\sim$11 days before the CALIOP measurements were available. This means that the larger particles would have already sedimented out before the measurement period began (see Rose and Durant, 2009, for discussion on atmospheric residence times of volcanic ash). We therefore do not capture the fall out of larger particles in the depolarization ratio, but instead observe layers composed of small, irregular (depolarising) ash particles.

Page 8, lines 7-8: This statement about the volume and the particle depolarization ratio is misleading.

Response:
We assume the reviewer is referring to the statement on page 18 lines 7–8:

"Note that δ_v is not strictly a particle property, but for layers dominated by aerosols it can be used as a first approximation to the particulate depolarization ratio, δ_p (Wiegner et al., 2012)."

We agree and have removed it from the revised manuscript.

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