Response to RC4

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

General

The paper is well written. Carefully analysed CALIOP observations are presented. The paper is appropriate for ACP.

The only negative and confusing point is that obviously the volume depolarization ratio and volume color ratio are used instead of the particle depolarization ratio and particle color ratio. But I am not sure what is shown. The authors have to clarify that when discussing equations 1 and 2, see details.

Minor revisions are at least required. However, major revisions (switch to particle depolarization ratio) would significantly improve the paper.

Response:
The authors thank the reviewer for their comments on the manuscript. As suggested, we have now included the particulate depolarization ratios. The 1064 nm lidar ratio ($S_{p,1064}$) and layer-effective particulate color ratio (χ_p) can be simultaneously retrieved using the two-color method of Vaughan (2004). We went to considerable effort to set up such an analysis scheme to perform the calculations. However, we found that the method was rather insensitive to variations in $S_{p,1064}$ because of the relatively weak signals and low optical depths of the volcanic aerosol layers. We therefore decided that these results added nothing to the value of the paper. We have added this comment in the revised manuscript as follows:

"We also note that the layer-effective particulate color ratio, χ_p, can be retrieved using the two-color method of Vaughan (2004). This approach seeks to minimise a non-linear function by simultaneously varying $S_{p,1064}$ and χ_p using the method of non-linear least squares. However, for the case studies considered here, we found that the method was rather insensitive to variations in the 1064 nm particulate lidar ratio; often resulting in non-physical solutions for $S_{p,1064}$. We expect that this was due to the relatively weak signals and low optical depths of the volcanic aerosol layers under examination. As these results were inconclusive, and require a more complete treatment of the sources of error, we decided this analysis was outside of the scope of the present analysis and therefore do not report the results here."

Upon implementing the $S_{p,1064}$ retrieval code we noticed an error in the $S_{p,532}$ retrieval. The error was due to the way the initial lidar ratio (defined by Eq. (7) in the original manuscript) was calculated. In the original code, η values of 0.6 were used in Eq. (7) and η values of 0.90 (for Puyehue) and 0.95 (for Kasatochi and Sarychev) were used in Eq. (3) when we should have been using the same η values in both Eq. (7) and Eq. (3). We have now corrected this error by using an η value of 0.95 for Kasatochi and Sarychev and 0.90 for Puyehue in both Eqs. (3) and (7). We have found that this error resulted in lidar ratios that were biased high by $\sim$4%. To illustrate this, we have plotted the original dataset against the η corrected dataset in Figure 1 of this document.

During this process we also found a bug in the lidar ratio retrieval code. The bug was due to the way the trapezoidal integration procedure (used to evaluate the integral term in the denominator of Eq. (3)) handled masked values. Specifically, if there was at least one masked value in an array then the integral of the array would be evaluated as being masked; leading to a masked lidar ratio retrieval, which was rejected from the analysis. We have revised the code now so that an

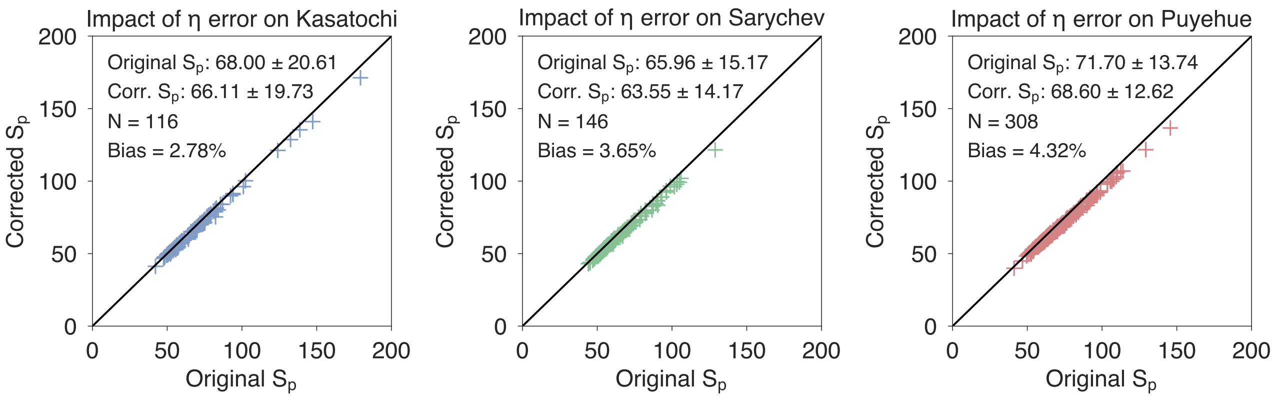

Figure 1: Comparison of S_p for the original dataset and the dataset corrected for the η error.

array containing masked values will still be evaluated. This is achieved using the cumulative trapezoidal integration module from the Scipy library (`https://docs.scipy.org/doc/scipy/reference/generated/scipy.integrate.cumtrapz.html`). The result of this revision on the analysis is that more data points (more valid lidar ratio retrievals) are now analysed. The results presented in the revised manuscript do not, however, differ significantly from the results presented in the original manuscript and so the main conclusions drawn from the original manuscript have been retained. The impact of this correction on the analysis is shown for a specific example of an observation of an ash layer for the Puyehue case study (Figure 2 of this document). Here the added data points are in red and lidar ratios that have been corrected for the η error are in blue (Figure 2d of this document). Figure 3 (of this document) shows how the correction impacts the overall lidar ratio PDFs. In the revised manuscript, Figs. 2–10 of the original manuscript have been corrected for the η error and the integration bug (corresponding to η + integration values that are annotated on the subplots of Figure 3 of this document). The values in Tables 1–3 have also been corrected in the revised manuscript.

Details:

Abstract:

P1, L9: Please state the wavelength (532 nm) again in the case of the volume depolarization ratio.

Response:
Accepted.

P1, L10-12: A volume depolarization ratio of 0.08, 0.05, 0.25 tells us almost nothing as long as we do not know the backscatter ratio (total-to-Rayleigh backscatter). So again, why not trying to determine the particle depolarization ratio? At least for a few examples.

Response:
The authors disagree that the volume depolarization ratios tell us “almost nothing” without the scattering ratio. The volume depolarization ratios presented do show distinctions between the layers identified as sulfates and the layers identified as volcanic ash (Fig. 9 of the original manuscript). One could argue that, for CALIOP, the volume depolarization ratios are more useful than the particulate depolarization ratio as the volume depolarization ratios are direct measurements (i.e. do not require

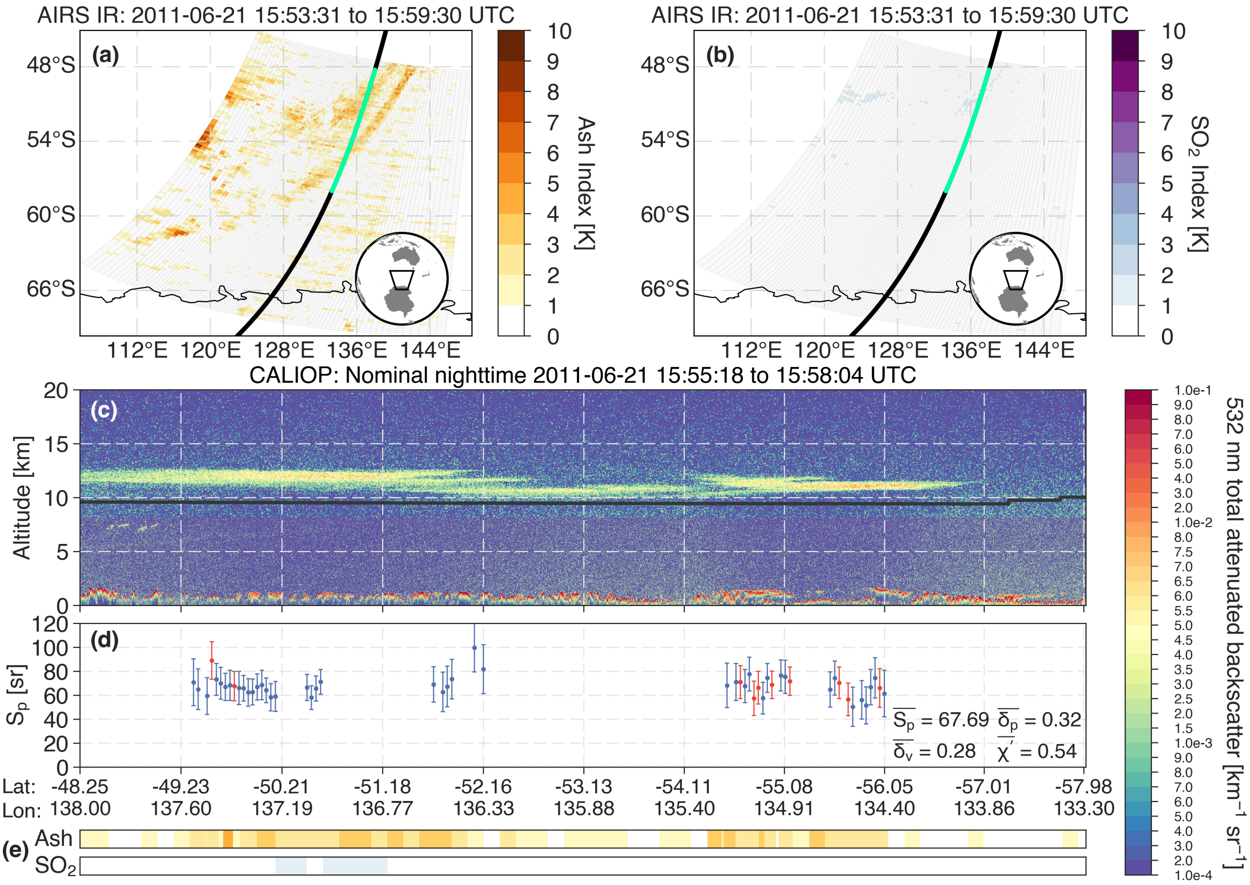

Figure 2: Revised version of Fig. 7 of original manuscript. Red data points on panel (d) indicate retrievals that were added after the integration bug was corrected.

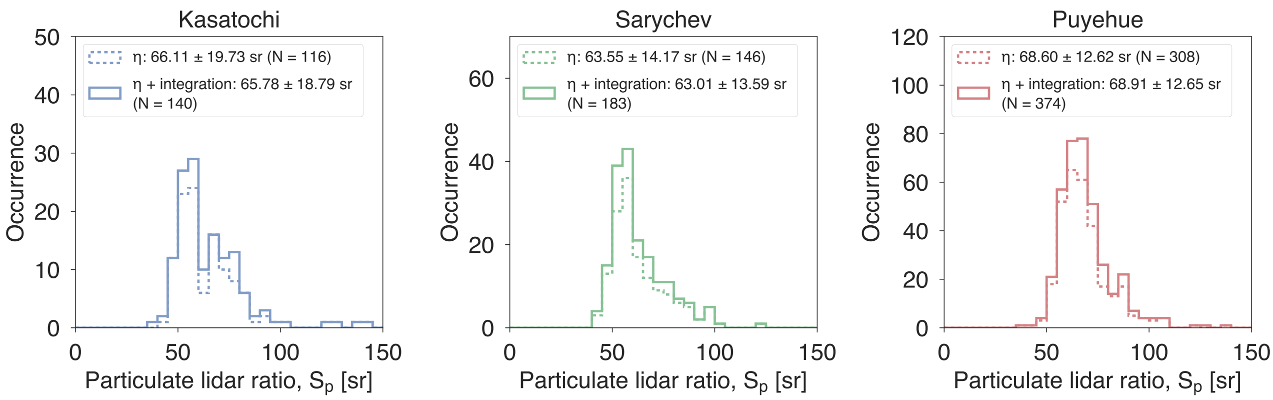

Figure 3: Impact of integration bug on S_p for each case study. The mean and standard deviation of S_p for the η correction and the η + integration correction are annotated on each plot.

a lidar ratio retrieval). This is in fact the reason why we focussed on the volume depolarization ratios in the initial submission. However, we agree that the particulate depolarization ratios provide useful intrinsic information on volcanic aerosols. We have calculated the particulate depolarization ratio by adapting the method of Tesche et al. (2009) to layer-integrated properties:

$$\delta_p = \frac{\gamma_m(\delta_v - \delta_m) + \gamma_p \delta_v (1 + \delta_m)}{\gamma_m(\delta_m - \delta_v) + \gamma_p(1 + \delta_m)} \quad (1)$$

where

$$\gamma_m = \int_{r_t}^{r_b} \beta_m(r) dr, \quad (2)$$

$$\gamma_p = \int_{r_t}^{r_b} \beta_p(r) dr \quad (3)$$

and

$$\delta_m = \int_{r_t}^{r_b} \delta_m^C dr = \delta_m^C (r_t - r_b). \quad (4)$$

Here the particulate backscatter profile, $\beta_p(r)$, is calculated using the retrieved 532 nm particulate lidar ratio and the numerical integration procedure of Fernald (1984). We also define δ_m as the layer-integrated molecular depolarization ratio. Due to CALIOP's narrow band optical filter, the integral of δ_m can be evaluated as above using the depolarization ratio at the central Cabannes line, which can be assumed to be a constant; $\delta_m^C \approx 0.003656$.

Introduction:

P2, L22: Later on, in this paper, you mention the Mattis paper which also deals with the same volcanic eruptions in the high northern latitudes in 2008 and 2009. I checked that paper and found lidar ratios and depolarization ratios for 355 and 532 nm for high- northern-latitude volcanic aerosol in the upper troposphere and stratosphere.

So, I was surprized that you did not give any reference to this paper in the introduction. Is there a specific reason, or did you simply forget? Mattis found lidar ratios of 30-40sr for 532nm and 60-80 sr for 355nm in August 2008 (upper troposphere, clearly related to volcanic aerosol), and 30-50sr for both wavelength between 14-18 km height one year later. And, by the way, Mattis found volume depolarization ratios of 0.015. Such low numbers really indicate sphercial particles, in contrast to your high numbers of 0.05 to 0.08 for the volume depolarization ratio, so that I started to think about the particle depolarization ratio.

So, please give proper reference to that Mattis paper in the introduction!

Response:

Thank you for bringing our attention to this. Reference to the Mattis paper has now been included in the introduction. We note Mattis et al. (2010) were using Raman observations of the volcanic layers which have a much higher signal-to-noise ratio than the native CALIOP measurements. And so we question whether it would be possible, in practice, to measure volume depolarization ratios as low as 0.015 using CALIOP.

Instead, you mention papers that deal with volcanic layers in the lower troposphere. Please give the heights of these volcanic layers so that the reader can make his/her own conclusion how useful such information is in a paper dealing with stratospheric volcanic layers.

Response:

The tropospheric layers mentioned in the introduction were describing previously reported lidar ratios for ash-rich volcanic layers. To our knowledge, there are no reported lidar ratio retrievals for ash-rich layers residing in the stratosphere. When discussing sulfate-rich aerosol layers, we gave reference to Sawamura et al. (2012) who report lidar ratios for stratospheric aerosols produced by the Nabro eruption, O'Neill et al. (2012) who report stratospheric lidar ratios for Sarychev and Hoffmann et al. (2010) who report stratospheric lidar ratios for Kasatochi. However, we agree that the heights of the layers should be reported and have included them in the revised manuscript.

P4, L19-20: Again, I am not very happy that you do not make any attempt to provide particle depolarization ratios.

Response:

Particle depolarization ratios have now been included.

Section3: I appreciate the careful consideration of potential multiple scattering effects!

Response:

Thank you.

Now, I got confused! Equation 1 leads, to my opinion, to the particle depolarization ratio. Right? Please clarify that! Are these cross and co-polarized backscatter coefficients for particles???? or for the total (Rayleigh plus particle) backscattering. Please make that very very clear!

If that is for the total backscatter then please put an index p to the ones in equation 2!... or are these total (Rayleigh plus particle) backscatter coefficients as well???

Response:

Equation 1 does not lead to the particulate depolarization ratio. $\beta'_\perp$ and $\beta'_\parallel$ are used to indicate the cross and co-polarised channels of the **total** (molecular + particulate) attenuated backscatter and therefore Eq. 1 leads to the layer-integrated **volume** depolarization ratio. The volume depolarization ratio is taken from the level 2 layer products, and is actually defined as the ratio of the summation of the co and cross-polarised channels (Vaughan et al., 2005):

$$\delta_v = \sum_{k=top}^{base} [\beta'_{m,532,\perp}(r_k) + \beta'_{p,532,\perp}(r_k)] \Big/ \sum_{k=top}^{base} [\beta'_{m,532,\parallel}(r_k) + \beta'_{p,532,\parallel}(r_k)], \tag{5}$$

where $\beta'_{m,532,\perp}(r)$ and $\beta'_{p,532,\perp}(r)$ are the molecular and particulate components of perpendicular attenuated backscatter at 532 nm, $\beta'_{532,\perp}(r)$, and $\beta'_{m,532,\parallel}(r)$ and $\beta'_{p,532,\parallel}(r)$ are the molecular and particulate components of parallel attenuated backscatter, $\beta'_{532,\parallel}(r)$. The perpendicular and parallel components of attenuated backscatter make up the total attenuated backscatter at 532 nm (i.e. $\beta'_{532}(r) = \beta'_{532,\perp}(r) + \beta'_{532,\parallel}(r)$). This definition has been included in the revised manuscript.

I got confused because equation 3 deals with the Fernald 1972 approach! So, you have the potential to compute particle backscatter coefficients and particle depolarization ratios when using the later Fernald method (Appl. Opt.l, 1984). So, why not presenting particle related quantities: lidar ratio, depolarization ratio, color ratio?

Response:

Indeed, we had originally calculated the particulate depolarization ratio. We wanted to focus on the volume, rather than particulate, properties as they are direct measurements from the CALIOP instrument and are operationally used in the level 2 aerosol classification scheme (Omar et al., 2009). It is not trivial to calculate the particulate color ratio. To retrieve it you need the particulate backscatter profile at 1064 nm. This requires knowledge of the particulate lidar ratio at 1064 nm, which cannot be retrieved using the two-way transmittance method as the 1064 channel is ∼16 times less sensitive to molecular backscatter than the 532 nm channel. The 1064 nm lidar ratio and particulate color ratio can be retrieved using the two color method of Vaughan (2004); however, as discussed above, we found that the method was rather insensitive to changes in $S_{p,1064}$.

Figure 2 is very nice, but I am missing the particle depolarization ratio, and obviously the color ratio is also for Rayleigh plus particle backscatter coefficients, and thus not very helpful. . .. But, at the moment, I am not sure what is shown.

Response:

Thank you. We have now included the particle depolarization ratio, for all three case studies, in a revised Fig. 2. The particulate color ratios are not shown for reasons discussed above. The authors disagree with the notion that the layer-integrated volume color ratio is “not very helpful”. The value of χ' does show some distinction between the sulfate-rich (Kasatochi and Sarychev) layers and the volcanic ash-rich layers (Puyehue). The χ' parameter is also used in the aerosol classification scheme for CALIOP and thus it is a valuable piece of information when attempting to classify volcanic aerosols in CALIOP observations. Indeed, Vernier et al. (2013) use χ' measurements to separate ash from ice.

All the results in the figures are nice (figures 5,6,7,8 ,9), but I am still confused to see PARTICLE lidar ratios together with information on VOLUME depolarization ratios and VOLUME color ratio.

Correlations (Fig.9)1of PARTICLE lidar ratio versus VOLUME depolarization ratio are poor!!! Apples and oranges are correlated, to my opinion.

Response:

Figure 9b has been revised to compare the particulate lidar ratio against the particulate depolarization ratio. However, Fig. 9a that shows the volume color and volume depolarization ratios has been retained as the relationship between these parameters, we argue, is an important source of information for the classification of volcanic aerosols in CALIOP observations.

May be it is simply not easy to compute particle depolarization ratios and particle color ratios. But at least a figure showing both, the volume and particle depolarization ratio and maybe the same for the color ratio is required to convince the reader that such correlations as in Figure 9 are useful.

Response:

As previously discussed, the color ratio requires knowledge of the particulate lidar ratio at 1064 nm. While methods do exist to retrieve the 1064 nm lidar ratio (e.g. Vaughan, 2004) we believe that to include this retrieval method would be outside the scope of the present study. However, we are able to calculate the layer-integrated particulate depolarization ratio and have included a new figure comparing the particulate lidar ratio to the particulate depolarization ratio for each case study.

As the reviewer is aware, CALIOP is not a Raman lidar and cannot measure extinction and lidar ratios directly and must retrieve particulate quantities using lidar ratios that are either retrieved using transmittance constraints or default values for each aerosol subtype (Omar et al., 2009). Even if it were a Raman system, travelling at ∼7.5 km / second would not permit the long averaging times that improve the SNR in ground-based Raman systems, and spatial inhomogeneity and limited horizontal extent of features do not always permit CALIOP to increase its SNR by averaging over long along-track paths. So deriving particulate properties is dependent on SNR, as we have seen in this paper. Until we have operational HSRLs in space, we are limited then to using the elastic backscattering signals from CALIOP and analytical techniques using this form of lidar data.

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