Response to RC1

I find this paper to be an interesting contribution with a few minor issues. My main concern is with section 6.2 which I think needs some more care in how they infer things from the data. A common issue throughout the paper is that the meanings of things like color ratio and depolarization ratio are given much context (what does a value of X really mean).

Response:

The authors thank the reviewer for comments on the manuscript. As suggested, Section 6.2 has been revised and more care has been taken when interpreting the time evolution of the volcanic aerosols. 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 $\sim$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.

We have also more explicitly defined both the depolarization ratio and the color ratio in the revised manuscript. We note, though, that the color ratio is constructed based on only two measurements (532 and 1064 nm attenuated backscatter) and so it is difficult to infer, quantitatively, what the volcanic aerosol particle sizes are without assuming more about the complex refractive index and size distribution of the particles. It can, however, be used to infer relative size. This is explained in more detail in the revised manuscript and in the responses to comments that follow.

Page 7, line 26. Does mean there was effectively no change in the values during measurement period?

Response:

Indeed, there was little change in the optical properties during the measurement period for the Kasatochi case study. We refer the reviewer to the revised Fig. 8 (Figure 1 below), which shows how the optical properties changed over time during the measurement time period.

Page 7, line 28. You commonly refer to layers as either sulfate or ash. While sometimes these layers separate themselves, other times they can be mixed in a complex fashion. You may wish to define your layers as layers optically dominated by ash or sulfate aerosol rather than imply that they are distinctly one or the other. Is it possible that complex mixing is responsible for the rather large variations in the backscatter to extinction ratio? Alternatively, is it consistent with noise or variability in the sulfate and/or ash itself?

Response:

Thank you for suggesting this. In the revised manuscript, the authors are careful to refer to layers as 'ash-rich', 'sulfate-rich' and as being 'optically dominated' by either ash or sulfate. To the reviewer's second point: as we are measuring the aerosol layers over a number of days across the globe, it is possible that complex mixing of ambient aerosol will be occurring over time. Sedimentation, dehy-

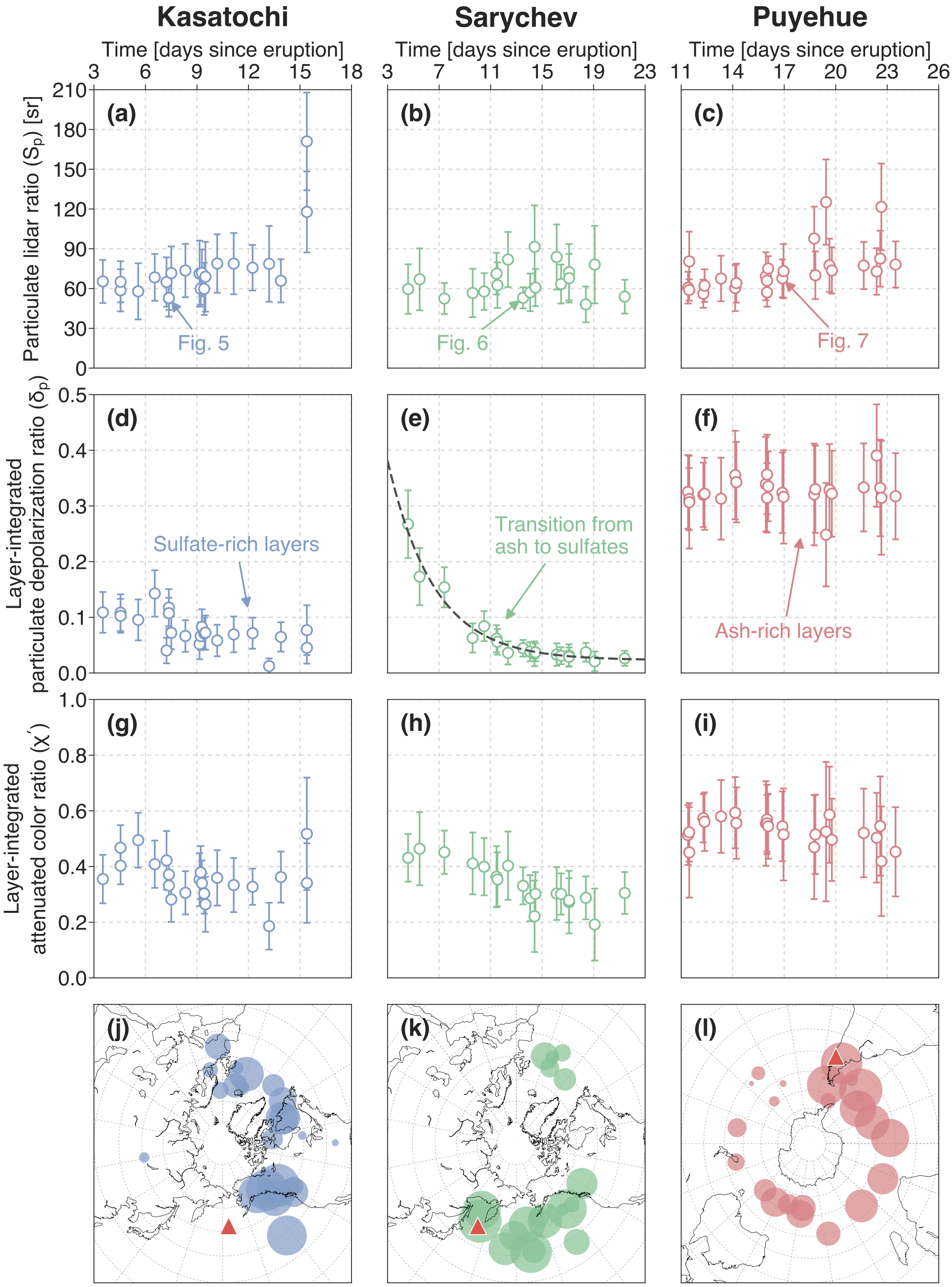

Figure 1: Revised version of Fig. 8 of the original manuscript.

dration and coagulation processes are also likely to be occurring. Therefore some variability in the lidar ratio should be expected. We note, however, that the lidar ratio retrieval becomes more sensitive (and uncertain) to changes in the return backscatter signal as the two-way transmittance approaches 1 (see Fig. 4 of original manuscript). As the majority of the aerosol layers were optically thin ($\tau_e < 1$), the large variability (high standard deviations) in the lidar ratio PDFs (Fig. 2 of original manuscript) is probably also, in part, due to the high sensitivity to noise in the backscatter return signal. However, based on the observational evidence provided by the color and depolarization ratios, we believe that CALIOP has captured compositional changes in the volcanic aerosols under examination; particularly for the Sarychev case (see Figure 1e of this document).

Page 8, Some of these figures are much too small to see much detail in. I know I can blow them up to see them but my experience is that ACP makes them into JPGs for the final figures and they are always ‘infinite’ resolution like some bad TV show.

Response:

To improve readability, we have increased the size of all figures on page 8. We have also increased the font size in all figures. The majority of our figures are in pdf (vectorised) format and so no resolution will be lost in the ACP typesetting stage for these figures. All other figures are in png (non-lossy) format with a dpi of 600. We have been careful to follow the ACP guidelines on producing high quality figures as described here: http://www.atmospheric-chemistry-and-physics.net/for_authors/manuscript_preparation.html

Page 9, Here I will be a curmudgeon, I hate VEI. People use it like it is a quantitative assessment of volcanic explosivity and I think it is disappointingly far short of that and often is not relevant to stratospheric impact. Check this out (a commercial site but the definition is correct) http://geology.com/stories/13/volcanic-explosivity-index/ . The definition is a mess.

Response:

Reference to the VEI has been removed in the revised manuscript as the authors agree that its use here is not relevant to the study.

Page 9, How do you avoid ice-rich layers? (line 20) Also, since there is a composition change from sulfate to ash, how sure are you that the changes in the color ratio are due solely to size rather than simply that they are a different color?

Response:

Ice-rich layers are avoided based on the Ash Index (AI) criterion. If a stratospheric layer were ice-rich then we would expect the AI to be strongly negative (Prata et al., 2015). Since our criterion is set so that we only accept AI $\geq$ 1 we assume that ice-rich layers have been removed from the analysis. The explicit definition of the AI has been included in the revised manuscript.

To the reviewer’s second point, we never suggest that there is a transition from sulfate to ash, rather, we suggest that there is a composition change from ash to sulfate (based on the depolarization and color ratio changes for the Sarychev case). Sulfate aerosols are generally in the 0.1–1 μm radius size range and ash particles that have resided in the stratosphere for more than 2 weeks would likely be sub-micron to micron size. Indeed, O’Neill et al. (2012) report effective radii of 0.25 μm for the Sarychev aerosols over the Arctic. This means that the size of the particles under examination is less than/comparable to the sampling wavelengths (532 nm and 1064 nm). In this sense, we are talking about scattering and absorption, rather than reflection, which means changes in the ‘color’ of the particles (in the usual sense of the word) could not be inferred using CALIOP measurements.

The color ratio can change due to changes in the size, complex refractive index and shape of the aerosols being measured. We speculate that ash particles were present in the initial observations of the CALIOP measurements for Sarychev case and so a combination of the sedimentation (contributing to a reduction in particle size) and sulfate formation (contributing to a change in the imaginary part of the refractive index) led to changes in the color ratio. This discussion will be included in the revised manuscript.

Page 13, line 11. I think it would be more proper to say 'unambiguously identifying this layer as containing non-spherical particles. It is not necessarily an either/or situation...

Response:

Accepted. The sentence in the revised manuscript has been amended to read:
"The Puyehue layers (Fig. 7) are quite similar to the sulfate-dominated layers in terms of the geometric thickness; however, the layer-integrated optical properties, along with the AIRS ash signal, unambiguously identify this layer as containing non-spherical ash particles."

Section 6.2. I find much of this discussion to be speculative and perhaps the authors are over analyzing their results. Certainly, changes over time that are small compared to the measurement uncertainty is not terribly convincing. They authors are seem to forget that they never measure the same aerosol and that for an inhomogeneous cloud they cannot really be sure that some of the differences are not just variability in the cloud. The authors also do not mention that the aerosol is mixing with ambient aerosol throughout this period and so some changes are may be a result to that process. I would not bother with the humidity explanation and the comparisons with the Icelandic eruption are not likely to be particularly relevant. It is extremely common for sulfate aerosol to contain volcanic material (and meteoritic, etc.) while optically suggesting spherical particles. Given the high number densities after an eruption some coagulation between ash and sulfate is bound to occur in mixed layers. Perhaps some of these arguments would hold together if we had any idea of how big the ash particles are (i.e., what does the color ratio mean?). (For that matter how good do the authors believe the color ratios are? My impression of the 1064 nm channel on CALIOP is that it is not very robust though differences are real even if not correct).

Response:

Thank you for this comment. Section 6.2 has been revised to accomodate the reviewers suggestions. We have removed the discussion of the changes in layer-integrated attenuated backscatter and the coagulation and condensation processes as we agree that this part of Sect. 6.2 is speculative based on the evidence (see also revised Figure 1 of this document). We instead comment on the fact that changes in the lidar ratio may be due to variability and inhomogeneity in the aerosol layers. We have also now incorporated discussion on the possibility of volcanic aerosol mixing with ambient aerosol during the measurement period. The humidity explanation has been removed; however, as there are few ground-based observations of the volcanic ash lidar ratio together with depolarization ratio (at 532 nm), we believe that comparison of the Eyjafjallajökull observations with the Puyehue observations is justified.

As discussed above, while we are not able to retrieve particle size, the color ratio can indicate relative changes in particle size. For these reasons we can infer that the Puyehue particles were larger than the Kasatochi and Sarychev particles. Reference to O'Neill et al. (2012) has been included in the revised discussion as they report on particle sizes for the Sarychev case.

The quality of the layer-integrated attenuated color ratios depends on the correct identification of the layer-top and base, the reliability of the 532 and 1064 nm calibration constants and the SNR. The 1064 nm channel calibration depends on the assumption that the color ratio for high cirrus clouds is

1. The calibration procedure is described in Section 7.1.2.2 of the level 1 ATBD (Winker et al., 2006) and the assumption of the cirrus cloud color ratio was determined to be justified based on a validation study using the Cloud-Physics Lidar (Vaughan et al., 2010). We therefore believe that, while there may be some variability in the calibration of the 1064 nm channel, the color ratios used here are robust enough to infer relative changes in particle size.

References

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