Reply to Anonymous Reviewer 2's review of revised manuscript

Anonymous Reviewer 2 has reviewed our 1st revised version of the manuscript, finding "Good" for all of the 6 sections of the review, except for "Presentation quality", which they have graded "Fair".

The reviewer has then made recommendation of "reconsider still requiring major revisions" with:

"The authors should correct what in my view are inconsistencies and explain better how the aerosol transmittance correction is performed to retrieve the "true" scattering ratio."

In the remainder of this response (see below), we have replied to each of the Reviewer 2's comments, showing the reviewer's comments in black (Times font), and our replies to these in dark-red colour (Arial font). Where excerpts of text are shown from the original or revised manuscripts, these are shown in italics.

Review of manuscript ESSD-2020-246 (1st revised version)

The paper has been streamlined, taking some parts to a supplement, some errors have been corrected and, overall, it is more readable now. However, I still have some strong concerns, especially in the description of how the correction for the stratospheric aerosol transmittance (neglected in the dataset under rescue) is carried out (section 2.4), which, in my view, has not been clarified in the revision.

Major concerns

Lines 175-176: the authors state that *"The lidar backscattering ratio (SR(λ, z)) is commonly defined as the ratio between the total backscatter ($\beta_T(\lambda, z)$) and the molecular backscatter $\beta_m(\lambda, z)$, at the altitude z and wavelength λ"*, with which I agree.

However I find again the next sentences, namely *"$\beta_T(\lambda, z)$ is the sum of $\beta_m(\lambda, z)$ and the aerosol attenuated backscatter ($\beta_a{}^A(\lambda,z)$). That definition is related to the fact that in the retrieval of SRo(z) the two-way total transmittance(T^2) correction was neglected (Hostetler et al., 2006)"* most confusing.

That cannot be the definition of the backscattering ratio, where the aerosol backscatter coefficient should be the "true" one, not the aerosol attenuated backscatter coefficient. I could accept that Eq. (2) is what the author of **G-66**, obtained because of his neglecting of stratospheric transmittance, but not as the common definition.

The reviewer is right, and that was a typo in that 2nd sentence. Sorry for that.. We have corrected the typo (line 176 of the revised manuscript), replacing:

"…aerosol attenuated backscatter (β_a^A (λ,z))." with *"…the aerosol backscatter ($\beta_a(\lambda, z)$)."*

I think a difference should be clearly made between this scattering ratio with β_a non-corrected for atmospheric transmission and the corrected one (and transposed to another wavelength in addition), which, in my understanding, is what the authors aim to produce.

Yes, the reviewer is understanding correctly – our aim is to explain each step of the data processing methodology.

We have clarified the text to make it clearer where we are describing our specific recovery methodology, and where we are making a more general statement about a particular metric or measured quantity.

The authors seem implicitly recognize that in their reply to my first review when they say "The 2 two-way transmission terms are part or the exact definition of SR(λ,z) with no assumptions about the two-way transmission".

Yes, that's correct – we were trying to explain the method, but that was a typo, now corrected.

Furthermore I also find the reference to Hostetler el al., 2016 (dealing with CALIPSO inversion algorithm) misleading, as it does not support the neglect (at least I don't see how) of the two-way total transmittance in **G-66**.

Yes, we agree that follow-on sentence was confusing. Sorry. We have deleted it in the revised manuscript:

In summary, we agree with the reviewer's main point that the wording of this part of the revised manuscript required still some improvement. We have improved the text to now introduce the total and aerosol attenuated backscatter after equation 5 (lines 190 and 210 of the revised manuscript). As the reviewer hints, our aim is to be fully transparent in the method to keep the aerosol backscatter and aerosol extinction uncorrected by two-way aerosol transmittance in the initial derivation (at the measured wavelength of 694nm), correcting for aerosol transmittance only after conversion to the target wavelength of 532nm. The reason for postponing the aerosol transmittance correction is that the Blue Hill and Fairbanks sun photometer TAOD measurements were at 500nm wavelength (see supplementary material), much closer to the 532nm wavelength than 694nm. Note also that the order of the equations 7 and 8 has been changed in the revised manuscript in this improvement to the explanation of the methodology.

In summary, we have improved this part of the revised manuscript to address the reviewer’s valid concerns, and have copied and pasted this revised text for lines 190-210 below for maximum clarity:

“The single-wavelength elastic lidar systems provide profiles of attenuated total backscatter. The “true” total backscatter is calculated from $\beta_T(\lambda,z) = \beta_T^A(\lambda,z)\ T_j(\lambda,z)$*, where* $\beta_T^A(\lambda,z) = [\beta_m^A(\lambda,z) + \beta_a^A(\lambda,z)]$ *. Substituting into Eq.(2), taking into account that* $\beta_m(\lambda,z) = \beta_m^A(\lambda,z)\ T_j(\lambda,z)$ *and re-arranging specifically for this case of the 694nm backscattering ratio, gives:*

$$\beta_a^A(\lambda,z) = \frac{[\ SR(694,z) - 1]\ \beta_m(\lambda,z)}{T_j(\lambda,z)} = \frac{[\ SR(694,z) - 1]\ \beta_m(\lambda,z)}{T_m(\lambda,z)\ T_{O3}(\lambda,z)} \qquad (6)$$

Whereas the term $T_j(\lambda,z)$ *in equation 6 usually specifies the full two-way transmittance correction, at this stage of the processing methodology, we correct only for the attenuation due to molecular backscatter and ozone absorption:*

$$T_j(\lambda,z) = e^{-2\int_{sup}^{z} \alpha_j(\lambda,z)dz} \qquad (7).$$

The $\alpha_j(\lambda,z)$ *term is thus the vertical profile of 694nm extinction due only to molecular scattering (*$\alpha_m(\lambda,z)$*) and ozone absorption (*$\alpha_{O3}(\lambda,z)$*), with* z_0 *being the altitude of the lidar. This postponement of the aerosol attenuation correction is due to the 500nm wavelength of the contemporaneous AOD measurements being much closer to the target wavelength of 532nm (see Supplementary Material), the method preserving the two-way molecular and ozone transmittance corrections to be applied at the measurement wavelength of 694 nm.*

The conversion to aerosol extinction was carried out after first translating the aerosol backscatter from 694nm to 532nm, applying the corresponding wavelength exponent kb(z,t), calculated from in-situ size distribution measurements of the Northern Hemisphere mid-latitude Pinatubo aerosol cloud (Jaeger and Deshler, 2002; 2003):

$$\beta_a(532,z) = \left[\frac{532}{694}\right]^{kb(z,t)} \beta_a\ (694,z)\ . \qquad (8)$$

Note again that the derived 532nm aerosol backscatter ($\beta_a(532,z)$*) has at this point only been corrected for two-way molecular scattering and ozone absorption transmittance effects.*

The aerosol extinction, $\alpha_a(532,z)$ *, at this point still uncorrected for two-way aerosol transmittance, is then calculated by the expression:*

$$\alpha_a(532,z) = EB_c(z,t)\ \beta_a(532,z) \qquad (9)$$

where $EB_c(z, t)$ *are altitude- and time-dependent coefficients to convert from aerosol backscatter to aerosol extinction (at λ = 532 nm), derived from the same Pinatubo aerosol size distribution measurements (Jäger and Deshler, 2003).*

Both the EBc and kb factors are derived from log-normal size distribution fits to balloon-borne optical particle counter measurements of the Pinatubo aerosol cloud from Laramie, Wyoming, USA. Each of the conversion factors in Jäger and Deshler (2002; 2003) represent averages over 4 height ranges: tropopause–15, 15–20, 20–25, and 25–30 km, and are provided for the 4-month periods November–February, March–June, and July–October of each year after the eruption. We used EBc(z,t) and kb(z, t) for the equivalent 4-month periods after the March 1963 Agung eruption, based on matching the same time-offset after the June 1991 Pinatubo eruption.

2. In addition, I still cannot understand how the sunphotometer-derived AOD is used to correct for the stratospheric AOD. It would seem that an AOD-constrained inversion is attempted but it is not clear how it is achieved:

The method we applied uses monthly means of the total AOD (TAOD) from the sunphotometer, converted from 500 to 532 nm (described above) assuming this to be tropospheric (tAOD) in the first step, to produce for each site a $\mathrm{T_a}(532,z)_*$

profile, so as to account for the tropospheric aerosol transmittance from the lidar altitude across the troposphere up to 11 km and the stratospheric aerosol transmittance in the lower stratosphere from 12 to 24 km. However, when the stratospheric AOD (sAOD_*) is calculated in the next step (from the first guess $\alpha_a^{Ta}(532,z)_*$), the resulting first guess total AOD (tAOD + sAOD_*) will be higher than the observed TAOD. The second step is aimed to estimate the magnitude of a consistent value for tAOD for each measurement to constrain $\mathrm{tAOD} + \mathrm{sAOD} \approx \mathrm{TAOD}$.

2.2.1 What does "sup" (the lower limit of the integral in the exponential in Eq. (8)) denote?

In the original and 1st-revised manuscripts, we used the term "sup" to denote the altitude of the lidar site. On reflection, perhaps that was not so clear, and in this 2nd-revised manuscript we have changed all instances of "sup" instead to "z_o". We also added in the manuscript, at the end of the sentence on line 197:

"… and z_o being the altitude of the lidar."

2.2.2 I have several problems with the first guess $T_a(532)$ mentioned in line 210:

a) How is it chosen?

We produced a first guess $T_a(532,z)_*$ for each measurement day at each site, in the range from 11 to 24 km. The $T_a(532,11\mathrm{km})_*$, a unique value from the lidar altitude to 11 km, was calculated using Eq.(8) and the TAOD(532) value for the month the measurement was conducted. $T_a(532,z)_*$ values from 12 to 24 km were calculated for each level using Eq.(8) and the uncorrected $\alpha_a(532,z)$.

b) It is stated it is "a unique value for all altitudes" (line 210). What does "unique" mean in this context?

By unique, we mean the same value is used for all the altitudes from the lidar up to 11 km.

c) The z-dependence seems to have disappeared. That would imply that $\alpha_a^{Ta}(532,z) = 0$

There is no z dependence below 11 km, but we do the calculation for each level at 12km and above, so there is a z-dependence in the main region of interest, from 12 to 24 km.

2.3 Likewise, how is the first-guess $\alpha_a^{Ta}(532,z)$

The first guess ($\alpha_a^{Ta}(532,z)_*$), was derived applying the correction for the first guess two-way aerosol transmittance $T_a(532,z)_*$ in Eq.(10).

2.4. What does mean that the profile of $T_a(532,z)$ has a constant value of $T_a(532)$ from the surface to 11 km? Again, if T_a is constant it means that the extinction coefficient is 0 (Eq. (8)). I suspect that the authors mean that they don't care about the profile of T_a between the surface and the 11 km height and that they consider only the *value* of T_a between the surface and that height; but in any case, the way they express it is, in my understanding, formally wrong.

Apologies, we may have used the term "constant" incorrectly there, perhaps we should have said "representative value" or similar. As we explained above we used the same unique/representative value from the lidar altitude to 11 km.

2.5. For the same reason, I suspect the Ta(532, z) profile between 12 and 24 km mentioned in line 215 might not correspond to the two-way transmittance between the surface and height z, but between 12 km and the height z. The authors might have a problem with their notation, failing to indicate the limits between which the atmospheric transmittance is calculated. Note that the definition of the transmittance (Eq. (8)) involves a definite integral, so the limits should be indicated, unless one of them is always conventionally the same, which I think might not the case throughout the paper.

No, the method is different between 12km and 24km. For that main region of interest, Ta(532,z) was calculated at each level z, to account for the corresponding two-way transmittance between the surface and height z.

I would tend to agree with the other reviewer's remark in his/her first review that an iterative procedure would be in order. From the authors reply to that remark (not very clear anyway to me) it seems they justify that only one (or two?) iterations would suffice, because there are other sources of uncertainty overshadowing that one. But this should be reflected in the paper text and the way of performing the aerosol correction should be clearly stated, in a way that could be reproduced by a reader. In the present form of the explanation, I would be unable to do it.

We agree with the reviewer. We added the sentence at the end of section 2.4:

“Although more iteration of those final steps would be possible, with the high magnitude of the estimated $\alpha_a^{Ta}(532,z)$ mean error, around 60%, compared to an estimated 15-20% maximum improvement achieved by the iteration procedure, we do not believe those additional calculations would be worthwhile in this case.”

We modified the text between the lines 206 and 231. We moved the 2 paragraphs from lines 217 to 230, inserting them in line 206. And several parts of the text where rewritten taking in consideration the reviewer’s comments in this point 2. The revised text now reads:

“The algorithms for the solution of the single wavelength lidar equations apply the two-way transmittance correction to the raw lidar returned signal, together with squared distance correction, before the backscattering ratio is calculated. In our case the available information we have the backscattering ratios which have been derived without conducting the two-way transmittance correction (G-66) for any species. That is the reason that this correction was included in the retrieval of $\beta_a(694,z)$ in equation (8). However only the molecular and ozone two way transmittance corrections (T_m $(694,z)$, $T_{O3}(694,z)$) were included in this step.

As explained above, the aerosol two-way transmittance correction, $T_a(532,z)$, was deliberately postponed until the final step to derive $\alpha_a^{Ta}(532,z)$, due to the available contemporaneous measurement information for AOD being at λ = 500nm (see Supplementary Material). We converted the measured AOD at 500nm to 532nm, using Ångstrom exponents covering the cited wavelength range from 1995 to 2019 from the nearest Aerosol Robotic Network (AERONET, 2020) stations. Although the tropospheric aerosol layer in the eastern US will have had different physical and chemical properties in the 1960s (e.g. Went, 1960; Husar et al., 1991), this is only a small change in wavelength, with the method then introducing much less error than had we converted from 500nm to 694nm to apply the aerosol attenuation correction. We note that monthly mean TAOD at 532 nm from Blue Hill Observatory, MA, from 1961 to 1966, were measured to be in the range from 0.1 to 0.4, consistent with the elevated background TAOD reported for the Eastern US during the sixties of the 20th century (Husar et al., 1981; Supplement 1).

We produced a first guess $T_a(532,z)_*$ for each measurement day at each site, in the range from 11 to 24 km. The $T_a(532,11km)_*$, a unique value from the lidar altitude to 11 km, was calculated using Eq.(7) and the TAOD value for the month the measurement was conducted. $T_a(532,z)_*$ values from 12 to 24 km were calculated using Eq.(7) and the uncorrected $\alpha_a(532,z)$.

The first guess aerosol scattering corrected by the total two-way transmittance ($\alpha_a^{Ta}(532,z)_*$), was derived applying the correction for the two-way aerosol transmittance $T_a(532,z)$

$$\alpha_a^{Ta}(532,z) = \frac{\alpha_a(532,z)}{T_a(532,z)} \qquad (10)$$

Since we are using the measured TAOD, which includes the stratospheric AOD, to calculate the first guess $T_a(532,z)_*$ we applied a second step after producing a first guess $\alpha_a^{Ta}(532,z)_*$ profile, where we then calculate the stratospheric AOD ($sAOD_*$), integrating $\alpha_a^{Ta}(532,z)_*$ between 12 and 24 km Then $sAOD_*$ is used in Eq.(11) to estimate the tropospheric AOD (tAOD) for each measurement:

$$tAOD = TAOD - sAOD_* \qquad (11)$$

Then the former $T_a(532,11km)_*$ values for each measurement will be replaced by new ones derived using the calculated tAOD corresponding to each measurement. Then the final profile of $T_a(532,z)$ for each measurement will consists of the the new $T_a(532,11km)$ calculated using tAOD and the already derived $T_a(532,z)$ values from 12 to 24 km that were calculated using the uncorrected $\alpha_a(532,z)$ in Eq.(7). Those profiles of $T_a(532,z)$ are applied in equation (10) producing the definitive values of $\alpha_a^{Ta}(532,z)$.

The method we applied uses monthly means of the total AOD (TAOD) from the sunphotometer, converted from 500 to 532 nm (described above), assuming this to be tropospheric (tAOD) in the first step, to produce for each site a $T_a(532,z)_*$ profile so as to account for the tropospheric aerosol transmittance from the lidar altitude across the troposphere up to 11 km and the stratospheric aerosol transmittance in the lower stratosphere from 12 to 24 km. However, when the stratospheric AOD ($sAOD_*$) is calculated in the next step (from the first guess $\alpha_a^{Ta}(532,z)_*$), the resulting first guess total AOD (tAOD + $sAOD_*$) will be higher than the observed TAOD. The second step is aimed to estimate the magnitude of a consistent value of tAOD for each measurement, to comply with the constraint $tAOD + sAOD \leq TAOD$.

Although more iteration of those final steps would be possible, with the high magnitude of the estimated $\alpha_a^{Ta}(532,z)$ mean error, around 60%, compared to an estimated 15-20% maximum improvement achieved by the iteration procedure, we do not believe those additional calculations would be worthwhile in this case.“

3. Lines 203-204: the authors say, referring to Eq. (9) that “$EB_c(z, t)$ are the altitude and time dependent backscattering to extinction conversion coefficients from λ = 694 nm to λ = 532 nm” but only “532” appears on both sides of Eq. (2), and the reference given (Jäger and Deshler, 2003) shows that they actually convert

from backscatter to extinction at 532 nm. The conversion from 694 to 532 nm has been made in Eq. (7) through the kb(z,t) exponent.

The reviewer is right. We have re-worded the text in that section to more clearly explain the methodology (see the excerpts of text from the improved manuscript at the end of our reply to the reviewer's point 1).

4. If $EB_c(z,t)$ (and kb(z,t) by the way), are time dependent, how and where is the time dependence taken into account? I suspect it is used to calculate the β_a and α_a uncertainties (Eqs. (20) and (21)), but this should be stated the first time the coefficients appear. Otherwise it is misleading, because the left-hand sides of Eqs. (7) and (9) should have a time dependence. Which are the "nominal" values used for kb(z,t) and $EB_c(z,t)$, around which their uncertainties have been used to calculate the β_a and α_a uncertainties in Eqs. (20) and (21)?

We have re-worded the text in that section to more clearly explain the methodology (see the excerpts of text from the improved manuscript at the end of our reply to the reviewer's point 1).

Other concerns

1. Line 143: it would be helpful to explain that SR(694,z) means the backscattering ratio at 694 nm and range z.

Added

2. Line after Eq. (1): while $\frac{dn(z)}{dt}$ is the photoelectron flux (electrons/s) resulting from scattering at range z, defining n(z) as "the number of photons at the altitude z" does not make much sense. In the explanation of Eq. (1), leave $\frac{dn(z)}{dt}$ as the photoelectron flux (electrons/s) resulting from the photons scattered at range z.

Corrected.

Erased: "*n(z) is the number of photons at the altitude z*".

Added: *"(electrons/s)" after "The average photoelectron flux…"*

3. The authors say that they have eliminated the squared superscript to indicate the two-way transmittance, but this has not been done consistently. There are examples of the superscript still remaining (for example, but not only, in lines 149,150 and 177).

The reviewer is right. Corrected. The squared superscript was eliminated in lines 149, 150, 152, 178, 195 and 206.

In figure 5 the labels of both panels were corrected, replacing respectively:

"*Nantucket sounding (No T-2 Corr.)*" by " $\boldsymbol{\alpha_a}$ "Nantucket sounding"

"*Nantucket sounding (T-2 Corr.)*" by " $\boldsymbol{\alpha_a^{Ta}}$ "Nantucket sounding"

4. There is still one instance (line 177) where SRo(z) is used.

The reviewer is right. Corrected. In lines 177, 470, 489, 490, 492, 493, 494, 497 and 498.

5. Line 215: "This profile of Ta(532,z) is applied in equation (11)". Equation (10) is probably meant.

The reviewer is right. Corrected.

Other Changes-Corrections made by the authors in the current version:

1) The reference:

Antuña-Marrero, J. C., Mann, G. W. , Keckhut, P., S. Avdyushin, B. Nardi and and L. W. Thomason, Ship-borne lidar measurements showing the progression of the tropical reservoir of volcanic aerosol after the June 1991 Pinatubo eruption. , *Earth Syst. Sci. Data,* (*Under Discussion*), https://doi.org/10.5194/essd-2020-81, 2020b.

Was updated to the one from the published paper:

Antuña-Marrero, J.-C., Mann, G. W., Keckhut, P., Avdyushin, S., Nardi, B., and Thomason, L. W.: Shipborne lidar measurements showing the progression of the tropical reservoir of volcanic aerosol after the June 1991 Pinatubo eruption, Earth Syst. Sci. Data, 12, 2843–2851, https://doi.org/10.5194/essd-12-2843-2020, 2020a.

2) We have added Dhomse et al. (2021) as a 2nd citation for the UM-UKCA Agung aerosol dataset, in addition to the Dhomse et al. (2020) ACP paper, since the 6Tg Agung simulation is now published as a "complete" volcanic forcing dataset, in the sense that these simulations are now provided in a form for use in climate model simulations. That is, they're provided as SW & LW waveband-mapped aerosol optical properties (extinction, absorption and asymmetry parameter). The 3 lines we have added the Dhomse et al. (2021) reference are listed below:

Line 536 -- changed "(Dhomse et al., 2020)" to "(Dhomse et al., 2020; Dhomse et al., 2021)".

Line 571 -- there was a missing cite on line 590 -- added "(Dhomse et al., 2020; Dhomse et al., 2021)" after "UM-UKCA Agung aerosol simulations".

Line 579 (caption to Figure 12) -- changed "(Dhomse et al., 2020)" to "(Dhomse et al., 2020; Dhomse et al., 2021)".

The added reference is:

Dhomse, S. S., W. Feng, A. Rap, K. S. Carslaw, N. Bellouin and G. W. Mann, 2021, "SMURPHS/ACSIS Agung volcanic forcing dataset (mapped to UM wavebands) -- from HErSEA ensemble of interactive strat-aerosol GA4 UM-UKCA runs (Dhomse et al., 2020, ACP)" (Version v1) [Data set]. Zenodo. http://doi.org/10.5281/zenodo.4744687