General comments:

The more frequent occurrence of pyro-Cb smoke plumes lofted into the stratosphere is a rich target for analysis into a kind of aerosol about which we know relatively little. I'm particularly interested to see apparently successful modeling of three-wavelength values of both linear particle depolarization ratio and lidar ratio from lidar measurements of one such plume. This is a new result and potentially quite useful, since previous particle modeling studies did not have access to such a complete lidar observation and also because they, in general, resorted to much more complicated models than what the current authors have found to be useful. So, for this reason, primarily, I would like to see this paper published. On the other hand, many aspects of the manuscript seem rather weak and unconvincing, so I believe major revisions are appropriate.

REPLY: We thank very much Reviewer #3 for his/her careful reading, comments and suggestions, which we address in the following. With his/her suggestions, we believe that the new version of the manuscript is significantly improved, and our findings are promoted in a better way. The author's answers along with the changes in the manuscript are listed below.

Remark: The figure numbers and the page numbers in the referee comments and in our replies correspond to the original manuscript.

First, the discussion and elimination of other models is not convincing. It may not be strictly necessary to show that other models perform worse if the near-spherical model is able to reproduce all the available measurements (because a simple model that fits all the observations has benefits over a more complicated model just by virtue of being simpler), but since an attempt is made to do so, it should be done thoroughly and correctly.

REPLY: We thank the reviewer for this comment. We added a more extensive discussion in the introduction on how our study differentiates for previous studies. Please see specific comments 3, 10 and 14 for the detailed changes according to this comment.

Second, an unsubstantiated claim is made that this could improve AERONET retrievals. The idea of the new model improving or complementing AERONET is potentially quite appealing, and if done right this could be a major focus of the paper, so again it should be addressed thoroughly, not haphazardly.

REPLY: We thank the reviewer for this very constructive comment. We improved the manuscript following the Reviewer's suggestions. Please refer to response for specific comment 13.

Finally, the speculation about the role of sulfuric acid for explaining the depolarization measurements seems a bit far-fetched and very difficult to validate while other potential explanations have not been adequately discussed. In my opinion, this is the weakest part of the manuscript and the best solution may be to simply not offer explanations for the shape at all, but rather to present this work as an advancement in the modeling of the optical properties alone. Otherwise, if the authors want to keep this, then a better, more thorough discussion of alternate theories and ways to distinguish between theories is needed.

REPLY: We thank the reviewer for this comment. At the current stage the discussion on the possible physical explanation for the smoke PLDR values is removed from the manuscript. More efforts will be made to investigate this issue and re-assess the possible coexistence of smoke particles and particles of sulfuring nature and whether this could affect the former in such a way to form near-spherical particles.

Specific comments:

1. *page 3, line 10:* *I think rather than "an explanation that could justify" the values, you're more fundamentally searching for "a model that can reproduce" the observations. This is a more precise statement of what this calculation is able to do and valuable enough at this stage.*

REPLY: Thank you for this comment, please find our answer below.

2. page 3, line 10. *"non-typical spectral dependence". What do you mean non-typical? Compared to what? It's my understanding that there are only a very small number of observations of three wavelengths of smoke particle linear depolarization ratio and not much discussion of two-wavelength observations. So, how do we know what spectral dependence is typical?*

REPLY: We thank the reviewer, we rephrased the following paragraph in order better position our research and to highlight the two comments above (**page 3, line 9**):

In contrast to prior studies, for our investigation for the stratospheric smoke originating from the Canadian wildfires, we do not adopt morphologically complex shapes of bare or coated smoke aggregates, which are associated with excessive computations. Instead, we propose a much simpler model of compact near-spherical particles. Our starting point and main assumption is that the particle near-spherical-shape can be highly depolarizing, as shown in the work of Mishchenko and Hovenier (1995) and Bi et al. (2018). Our analysis shows that for the Canadian stratospheric smoke observed above Europe in August 2017, the PLDR and LR measurements along with their spectral dependence, can be successfully reproduced with the proposed model of compact near-spherical particles. The size and refractive index of the particles are estimated as well, and seem to agree well with past observations for aged smoke. We further examine the capability of this model to be used on an operational level and in particular as an extension to the AERONET operational aerosol retrieval (Dubovik et al., 2006), since it provides a much simpler and faster solution with respect to more complicated shapes for stratospheric smoke particles (e.g. Mishchenko et al.,2016; Ishimoto et al., 2019)."

3. Besides just listing three papers at page 3, line 14, the introduction should discuss how this study is similar and different to the modeling studies in those and other papers, including those that use other modelled particle shapes other than near-spherical. Besides the 3 references listed, consider Kahnert (2017), Liu and Mishchenko (2018), Kanngießer and Kahnert (2018), Ceolato et al. (2018), and Luo et al. (2019) (some of these are mentioned later in the manuscript but not the introduction). Also Mishchenko et al. (2016) used multiple particle shapes, not just the near-spherical.

REPLY: Thank you for your comments, an updated discussion has been included in the introduction to point how our study is different from the previous work. We specifically added the following paragraph (**page 2, line 25**):

"In the past, many studies have used simpler or more complicated particle shape models in order to reproduce the lidar measurements of smoke and provide a physical insight on light interaction with these particles. In Kahrent (2017), the PLDR of black carbon aggregates covered by a cell of sulphates was simulated by two different models; a closed cell (i.e. each monomer in the aggregate is coated separately) and a coated aggregate model (i.e. the whole aggregate is coated). Their analysis showed that for thicker coating the coated cell model of volume equivalent radius of 0.3 to 0.4μm, can provide PLDR values of the order of 15% at 532nm. Mishchenko et al. (2016) and Liu and Mishchenko (2018) used rather complex morphologies for smoke particles, in order to reproduce the PLDR values measured by Burton et al. (2015). Amongst others, these morphologies included a) a fractal aggregate partially embedded in a spherical sulphate cell, b) two-externally-mixed spherical sulphate cells, each hosting an aggregate (models 6 and 11 in Fig. 1 in Liu and Mishchenko, 2018) and c) a high-density aspherical soot core, encapsulated in a circumscription spheroid cell (with axial ratio of 0.9 to 1.2; model 4 in Fig. 2 in Mishchenko et al., 2016). All these morphologies reproduced successfully the smoke optical properties measured by Burton et al. (2015). Moreover, Luo et al. (2019) used twenty different configurations of coated fractal aggregates and showed that for relatively small fractal dimension (i.e. relatively fresh aggregates), and for small black carbon fractions (i.e. densely coated aggregates; configuration C in Fig. 2 in Luo et al. (2019)), the PLDR values can reach up to 40, 15 and 6% at 355, 532 and 1064nm, respectively. Ishimoto et al. (2019) used fractal aggregates and artificial surface tension induced on the particles to mimic the effect of coating by water soluble materials forming around the particles. This particular study present results for both the PLDR and the lidar ratio (LR), which is indicative of the composition of the particles. In Liu and Mishchenko (2019), tar ball aggregates were used to model exceptionally strong PLDR as those measured by Burton et al. (2015). The aforementioned studies highlighted the fact that that in order to reproduce significant PLDR values (higher than 20% at 532nm), the fractals need to be coated (i.e. shapes of "Type-B, size 11, Vr = 20" shown in Fig. 4 of Ishimoto et al. 2019). We should point out though that most of them refer to monodispersed particles, and averaging over size could possibly supress some of the observed features.".

References *(that are not included in the initial version of the manuscript)*:

Kahnert, M.: Optical properties of black carbon aerosols encapsulated in a shell of sulfate: comparison of the closed cell model with a coated aggregate model, Optics Express, 25, 24579-24593, 10.1364/OE.25.024579, 2017.

Kanngießer, F., and Kahnert, M.: Calculation of optical properties of light-absorbing carbon with weakly absorbing coating: A model with tunable transition from film-coating to spherical-shell coating, Journal of Quantitative Spectroscopy and Radiative Transfer, 216, 17-36, https://doi.org/10.1016/j.jqsrt.2018.05.014, 2018.

Liu, L., and Mishchenko, M.: Scattering and Radiative Properties of Morphologically Complex Carbonaceous Aerosols: A Systematic Modeling Study, Remote Sensing, 10, 1634, 2018.

Luo, J., Zhang, Q., Luo, J., Liu, J., Huo, Y., and Zhang, Y.: Optical Modeling of Black Carbon with Different Coating Materials: The Effect of Coating Configurations, Journal of Geophysical Research: Atmospheres, 124, 13230-13253, doi:10.1029/2019JD031701, 2019.

4. How are the ranges arrived at that are shown in Table 1, and also specifically the fixed values for the distribution widths?

REPLY: The fixed width of the size distribution is a simplification we used in order to reduce the retrieval complexity, considering that this parameter does not greatly affect the lidar-derived optical properties (e.g. Burton et al., 2016). We specifically chose the fixed σ_g = 0.4 to represent a moderately wide size distribution. The fixed width of the shape distribution σ_s is also necessary for the reduction of the retrieval complexity. A small value of this width is used to avoid the wash-out of the characteristic optical properties of near-spherical particles which are shown for a relatively narrow aspect ratio range (e.g. Bi et al., 2018).

For the other microphysical properties used as inputs for the T-matrix calculations, the ranges were selected based on the ranges found in the literature for smoke particles. More specifically, in Muller et al. (2005) for aged Canadian and Siberian smoke, values of effective radius $reff$ from 0.16 (± 0.04) to 0.41 (± 0.14)μm, and real part of the refractive index m_r values of 1.37 (± 0.04) to 1.65 (± 0.03) were retrieved from 3b + 2a lidar data inversion. In a following study by Muller et al. (2007) the $reff$ of one-day-old smoke plumes was found to be 0.13 (± 0.04)μm, while for 18-day-old smoke plumes, the $reff$ was much larger, equal to 0.37 (± 0.06)μm. Nicolae et al. (2013), combined lidar measurements and mass spectroscopy for 3/4-day-old SW Romanian smoke, and derived an $reff$ of 0.19 (± 0.11)μm, while for 3-old Ukraine smoke the corresponding value was 0.40 (± 0.12)μm. For the same case studies, the complex refractive index values spanned from m = 1.41 (± 0.07) + i0.005 (± 0.003) to m = 1.66 (± 0.09) + i0.05 (± 0.01). In Giannakaki et al. (2015) for South Africa smoke, the $reff$ values derived from lidar data inversion at 355nm spanned from 0.11 to 0.28μm, and the complex refractive index values were derived to be m = 1.43 (± 0.07) + i0.016 (± 0.01). In Dubovik et al. (2002), the climatological mean value of the complex refractive index derived from AERONET measurements for tropospheric smoke in the United States and Canada is m = 1.5 (± 0.05) + i0.0094 (± 0.003).

To cover the range of the reported values in the studies listed above, in the initial version of the manuscript we used ranges shown in Table.1. For the revised version, we further extended the refractive index values as follows: m_r= 1.35 – 1.85 and m_i = 0.005 – 0.55.

We included the information above in the revised manuscript (page 3, lines 24 - 26 and page 18, Table 1).

References *(that are not included in the initial version of the manuscript)*:

Burton, S. P., Chemyakin, E., Liu, X., Knobelspiesse, K., Stamnes, S., Sawamura, P., Moore, R. H., Hostetler, C. A., and Ferrare, R. A.: Information content and sensitivity of the $3\beta + 2\alpha$ lidar measurement system for aerosol microphysical retrievals, Atmos. Meas. Tech., 9, 5555–5574, https://doi.org/10.5194/amt-9-5555-2016, 2016

5. Can you eliminate water or ice cloud as an explanation for the measurements in CALIOP?

REPLY: CALIOP measurements at the northeastern Canada on 15 August 2017 (https://www-calipso.larc.nasa.gov/products/lidar/browse_images/show_v4_detail.php?s=production&v=V4-10&browse_date=2017-08-15&orbit_time=17-55-33&page=3&granule_name=CAL_LID_L1-Standard-V4-10.2017-08-15T17-55-33ZD.hdf), show the stratospheric smoke layer at 11-14.5 km, where radiosonde measurements show temperatures below -40°C (Fig. 1 below). The radiosonde temperature profiles are from three stations close to the position of the smoke plume (Fig. 4b in manuscript): Churchill (Lat: 58.73, Lon: -94.08), Inukjuak (Lat: 58.45, Lon: -78.11) and Baker Lake (Lat: 64.31, Lon: -96.00). These low temperatures should exclude the presence of water clouds from CALIOP data, since even without the presence of aerosol particles, at these temperatures water can freeze homogeneously (Wallace and Hobbs, 2006; Fig. 6.29). Moreover, the ground-based lidar measurements on 23 August 2017 at Leipzig, show the stratospheric smoke layer at 14-16 km, where radiosonde measurements from the closest station (Lindenberg) provide temperatures below -50°C. Again the low temperatures indicate the absence of water clouds.

Regarding ice formation, the CALIOP PLDR values are below 20% both for the aforementioned overpass and for the closest overpass from Leipzig on 23 August 2017 (https://www-calipso.larc.nasa.gov/products/lidar/browse_images/show_v4_detail.php?s=production&v=V4-10&browse_date=2017-08-23&orbit_time=01-29-01&page=1&granule_name=CAL_LID_L1-Standard-V4-10.2017-08-23T01-29-01ZN.hdf (~90 km away and approximately 1 hour after the end of the ground based lidar measurements reported from Haarig et al., 2018), while the attenuated color ratio (i.e., the ratio of particle backscatter coefficient at 532nm to particle backscatter coefficient at 1064nm) is below 1. Further analysis of CALIOP data provides a mean (median) value of the backscatter related Angstrom exponent (BAE) at 532/1064nm, of 0.9 (0.9) with a standard deviation on 1.07. For PLDR, typical values for cirrus clouds are usually no less than 40% (Chen at al., 2002; Noel et al., 2002; Voudouri et al., 2020) and the color ratio is expected to be close to 1 due to the large size of ice crystals compared to the lidar wavelengths. For the BAE, values close to zero are expected, although the CALIPSO data are highly noisy at these altitudes as indicated by the standard deviation.

Moreover, for the overflight close to Leipzig on 23 August 2017, the lidar ratio (LR) measured from the ground based system is (66 ± 12) sr at 532nm. This is similar to the LR observed in the past for aged smoke particles (i.e. Fiebig et al., 2002; Veselovskii et al., 2015; Burton et al., 2012) but quite high for cirrus clouds (Gouveia et al., 2017).

Based on the above, although we cannot exclude the possibility of small ice crystals formed inside the stratospheric plume, we believe that the aforementioned characteristics indicate that this is not an ice cloud but rather a large smoke plume. Similar questions were raised also from Anonymous Reviewer 1, and Anonymous Reviewer 2. Hence we added the following paragraph to the manuscript to address the reviewers comment (**page 6, line 24**):

"Owning to the altitude of the smoke plume, one could attribute such PLDR values to the beginning of ice formation. Indeed, radiosonde temperature profiles from three stations located underneath the smoke plume (green stars on Fig.3b), reveal that the temperature above 11 km drops below -40C, at which point homogeneous ice formation can occur (Wallace and Hobbs, 2006). However, the PLDR values of cirrus clouds are usually no less than 40% (Chen at al., 2002; Noel et al., 2002; Voudouri et al., 2020). Further analysis of CALIOP data provides a mean (median) value of the backscatter related Angstrom exponent (BAE) at 532/1064nm of 0.9 (0.9) with a standard deviation on 1.07. For the BAE values close to zero are expected for cirrus clouds, although, as indicated by the large standard deviation, CALIPSO data are highly noisy at these altitudes. A recent study by Yu et al. (2019) also showed that the largest fraction of stratospheric smoke particles consisted of organic carbon (98% compared to 2% for black carbon). Particles of such high organic carbon content serve poorly as ice nuclei (Kanji et al., 2017; Phillips et al., 2013). Although the possibility of small ice crystals formed inside the smoke layers cannot be excluded, (largely due to the absence of in situ measurements) the aforementioned characteristics indicate that this plume consists of smoke particles rather than ice crystals.”

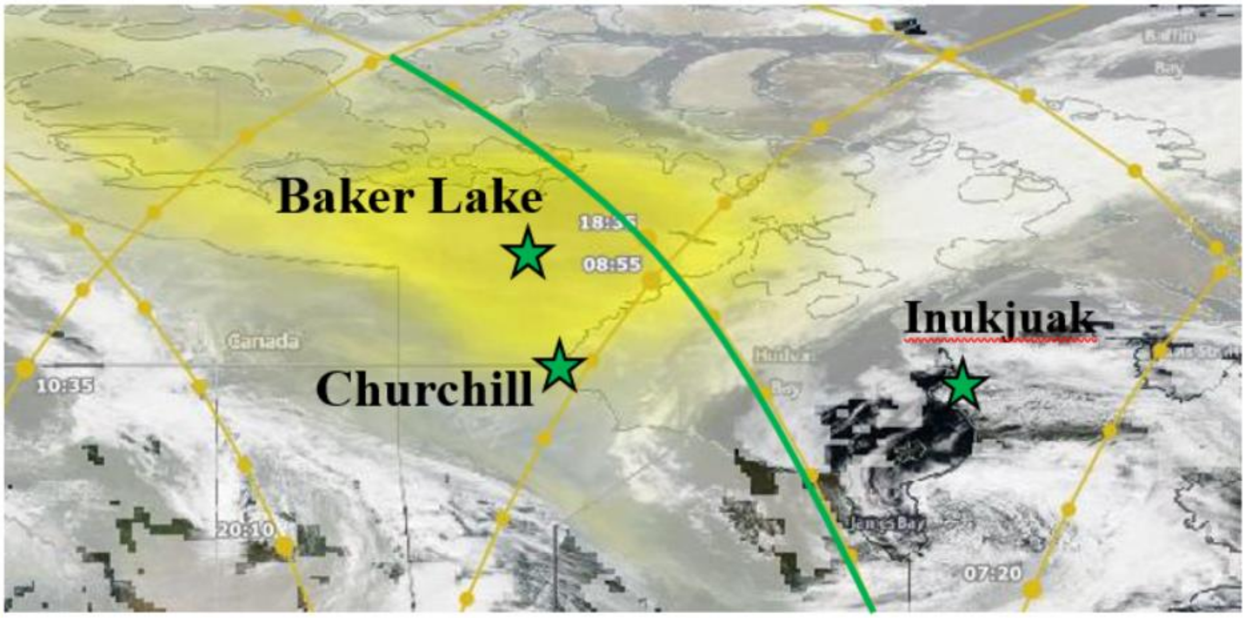


Figure 1. Corrected surface reflectance from MODIS on 15 August 2017, over-plotted with the PyroCb aerosol index product from Suomi NPP/OMPS (in yellow). Green stars indicate the position of the radiosonde stations used, while green line marks the CALIPSO overflight during 18:22 – 18:35 UTC. Maps are generated from the NASA Worldview Snapshots.

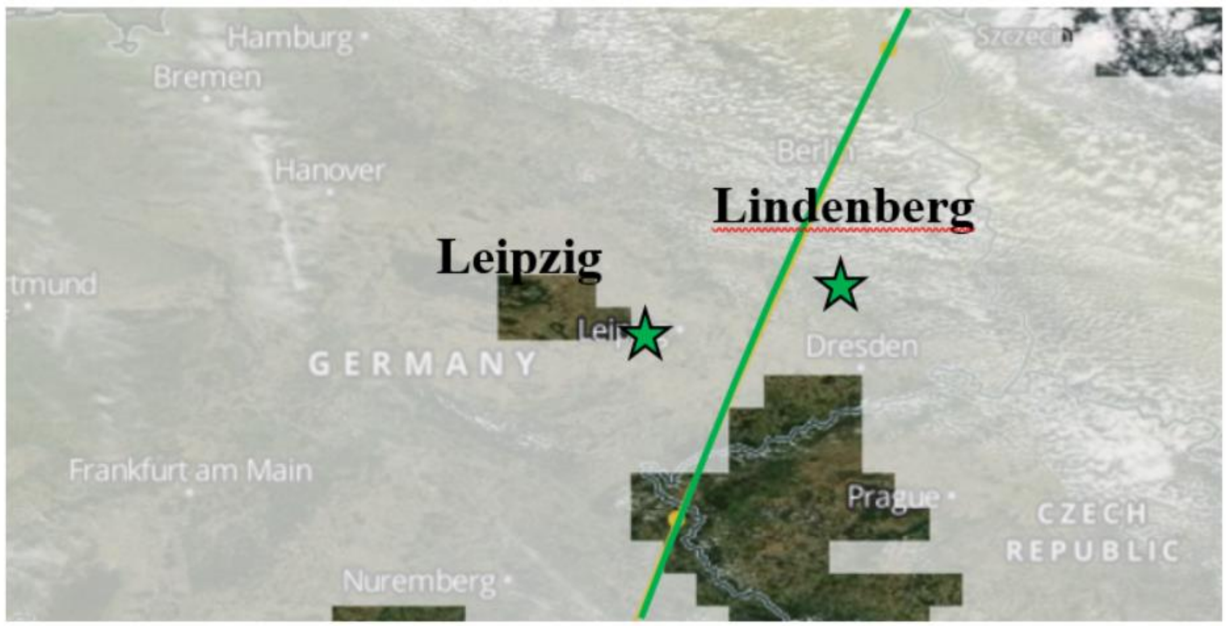


Figure 2. Same as Fig. 1 but for 23 August 2017. The CALIPSO overflight is approximately 90 km from Leipzig station, at 01:23 – 01:48 UTC, 1 hour after the end of the ground based lidar measurements.

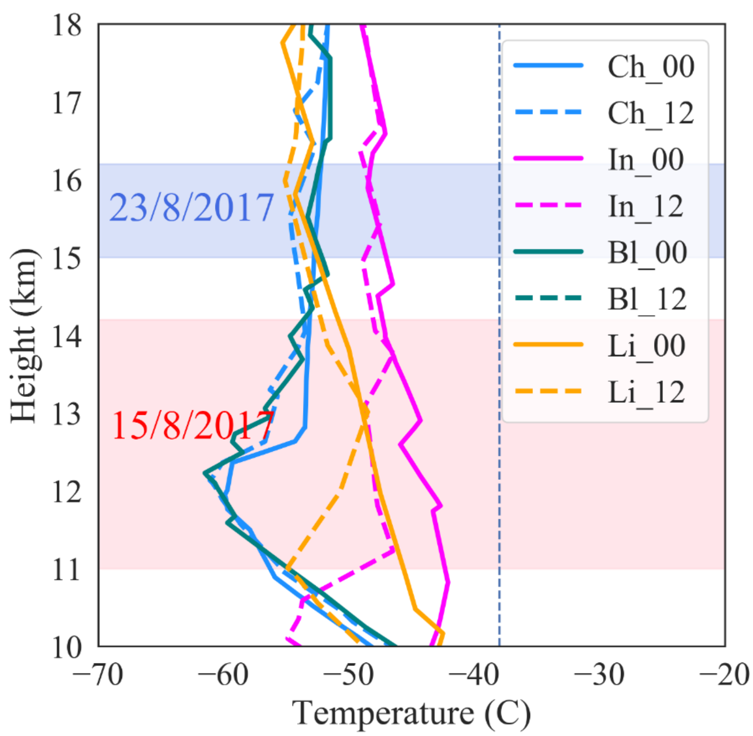


Figure 3. Radiosonde temperature (T) profiles from Churchill (Ch), Inukjuak (In), Baker Lake (Bl) and Lindenberg stations. Solid lines represent denote the measurements at 00:00 UTC, while dashed lines the measurements at 12:00 UTC. For the first three stations (Ch, In, Bl) measurements from 15 August 2017 are used, while for Li station from 23 August 2017. The pink box indicates the height of the smoke plume above northeastern Canada (11 -14 km) and the blue box the height of the plume after 8 days above Leipzig station (15 - 16 km).

References *(that are not included in the initial version of the manuscript)*:

Burton, S. P., Ferrare, R. A., Hostetler, C. A., Hair, J. W., Rogers, R. R., Obland, M. D., Butler, C. F., Cook, A. L., Harper, D. B., and Froyd, K. D.: Aerosol classification using airborne High Spectral Resolution Lidar measurements – methodology and examples, Atmos. Meas. Tech., 5, 73–98, https://doi.org/10.5194/amt-5-73- 2012, 2012.

Chen WN, Chiang CW, Nee JB. Lidar ratio and depolarization ratio for cirrus clouds. *Appl Opt*. 2002;41(30):6470-6476. doi:10.1364/ao.41.006470

Fiebig, M., Petzold, A., Wandinger, U., Wendisch, M., Kiemle, C., Stifter, A., Ebert, M., Rother, T., and Leiterer, U.: Optical closure for an aerosol column: Method, accuracy, and inferable properties applied to a biomass-burning aerosol and its radiative forcing, J. Geophys. Res., 107, 8130, https://doi.org/10.1029/2000JD000192, 2002.

Gouveia, D. A., Barja, B., Barbosa, H. M. J., Seifert, P., Baars, H., Pauliquevis, T., and Artaxo, P.: Optical and geometrical properties of cirrus clouds in Amazonia derived from 1 year of ground-based lidar measurements, Atmos. Chem. Phys., 17, 3619–3636, https://doi.org/10.5194/acp-17-3619-2017, 2017.

Noel, V., Chepfer, H., Ledanois, G., Delaval, A., and Flamant, P.: Classification of Particle Effective Shape Ratios in Cirrus Clouds Based on the Lidar Depolarization Ratio, Appl. Optics, 41, 4245–4257, https://doi.org/10.1364/AO.41.004245, 2002.

Veselovskii, I., Whiteman, D. N., Korenskiy, M., Suvorina, A., Kolgotin, A., Lyapustin, A., Wang, Y., Chin, M., Bian, H., Kucsera, T. L., Pérez-Ramírez, D., and Holben, B.: Characterization of forest fire smoke event near Washington, DC in summer 2013 with multi-wavelength lidar, Atmos. Chem. Phys., 15, 1647– 1660, https://doi.org/10.5194/acp-15-1647-2015, 2015.

Voudouri, K. A., Giannakaki, E., Komppula, M., and Balis, D.: Variability in cirrus cloud properties using a Polly^{XT} Raman lidar over high and tropical latitudes, Atmos. Chem. Phys., 20, 4427–4444, https://doi.org/10.5194/acp-20-4427-2020, 2020.

Wallace, J.M., Hobbs, P.V. Atmospheric Science: An Introductory Survey: Second Edition (2006), DOI: 10.1016/C2009-0-00034-8

***6. Figure 7** had a panel of water vapor mixing ratio but no explanation of this measurement or description of what impact this figure has on the analysis of this case.*

REPLY: Thank you for noticing this. Figure 7 (b) was actually used to indicate the prevailing stratospheric conditions for the months following the smoke injection. Since the physical process explanation is removed from the revised version of the manuscript (please refer to comments 20 to 24 below), Fig. 7 (b) is removed also.

7. For Figure 8 and Table 4 and others, please define the meaning of the error bars. Is this a true calculated uncertainty including both random and systematic error, or is this the standard deviation of available measurements, or something else? If it's standard deviation, how well do you think this captures the actual uncertainty of the lidar measurements at each wavelength? I ask because you mentioned that the 1064 nm extinction measurement is more challenging to make and we also know from literature that particle depolarization ratio in particular can be subject to significant systematic error in some circumstances (e.g. Burton et al. 2015, Freudenthaler 2016, Belegante et al. 2018).

REPLY: The values in Tab. 4 and Fig. 8 are the systematic uncertainty of the PLDR in the given layer boundaries. It is good that the reviewer points out the standard deviation of points within the layer as an additional source of uncertainty. The standard deviation for the PLDR in the stratospheric smoke layer is 0.010 at 355 nm, 0.006 at 532 nm and 0.016 at 1064 nm and is now included in the revised manuscript (see Table 4 and Fig. 8 in the revised manuscript). PLDR at 1064 nm was measured during a different in a separate time window (23:45 – 00:30 UTC, compared to 20:45 – 23:15 for the rest of the measurements), because it is not possible to measure the extinction coefficient and the depolarization ratio at that wavelength at the same time for the current configuration of BERTHA lidar system. The increased standard deviation at 1064 nm is caused by the shorter averaging period, 40 minutes instead of 2h30min, and the geometrically thinner aerosol layer, around 0.5 km instead of 1 km.

We included the standard deviation of PLDR measurements to the revised manuscript and we added the following (**page 7, line 22**): "The layer-integrated PLDR value at 355 nm is 22.4 ± 2.5 %, decreasing to 18.4 ± 1.2 % at 532 nm and 4 ± 2.3 % at 1064 nm. The, uncertainties in PLDR values include both the systematic error and the standard deviation of the measurements."

8. page 7, line 20-21. *Lidar ratio increase from UV-visible suggests that it also increases from visible to near-IR. This should be deleted. There's nothing in Muller et al. (2007) that addresses the lidar ratio values in the near-IR one way or the other.*

REPLY: Thank you for this comment. We rephrased the following paragraph (**page 7, lines: 19 - 21**): "The increasing tendency of the LR from the UV to the visible part of the spectrum has been also reported before for aged Canadian smoke (Müller et al., 2007). Measurements reported in Haarig et al. (2018) suggest that there is an increase also at the Near-IR, although currently there are no other available measurements of the LR of smoke particles at this wavelength."

9. page 7, line 22 "far from typical". I urge you to reword and avoid "typical". Muller et al. 2007 was a quite valuable paper, but the cases in it are somewhat limited, and it is now more than a decade old. Something that does not conform to Muller et al. 2007 is not necessarily "non-typical". We are still seeing new and different observations, by now including many of depolarizing smoke. I would say that this manuscript and other recent papers make a more convincing case that there is no "typical" for smoke or else that we do not have sufficient observations yet to know what is "typical", rather than that the depolarization ratios in this case are non-typical.

REPLY: We agree with the reviewer; this terminology is now changed throughout the manuscript. To address the comment specifically, we rephrased the following sentence (**page 8, line 22**): "On the other hand, the PLDR values of stratospheric smoke are much larger than those usually reported for tropospheric smoke (e.g. Muller et al., 2007)."

and (**page 8, line 24**): “So far, the only study presenting comparable results for tropospheric smoke is Burton et al. (2015).”

10. The notation in Equation 8 is confusing and doesn’t really make sense. Please define what are i and n? Is this a summation over the three wavelengths? In that case, you have two subscripts (i and lambda) that mean the same thing? Or maybe i is binary and means lidar ratio and depolarization ratio, but in that case, you do not show how the different wavelength measurements are combined. Equation 8 furthermore should arguably have the measurement uncertainty rather than the measurement itself in the denominator. This would be a more meaningful cost function considering you intend to compare the result to the measurement uncertainty in Eq. 9. Doing this could have a significant impact on your results, specifically the result for the Chebyshev model that is shown in Figure 12. The only simulated point that doesn’t fit the measurements is the 532 nm depolarization which has a very small reported uncertainty and therefore not much tolerance. But if the cost function reflected the error bars as well, you might find there is a solution that fits that point at the expense of slightly larger discrepancy in another quantity where the uncertainty tolerance is much larger (e.g. lidar ratios). I’m also curious how many solutions fit the criteria in Eq. 9 (or revised criteria) besides the minimum. Looking at this would give some insight into the uncertainty of your modeled results and the degree to which the set of measurements is sensitive to the complete set of free parameters in your model.

REPLY: In the original version of the manuscript Eq. 8 and 9 are written as following:

$$\sum_{i=1}^{n}\left(\frac{M_{\lambda}^{i}-S_{\lambda}^{i}}{M_{\lambda}^{i}}\right)=min \qquad \textbf{(Eq. 8)}$$

$$S_{\lambda}^{i}\leq e\left(M_{\lambda}^{i}\right) \qquad \textbf{(Eq. 9)}$$

For both equations subscript “i” is indeed binary, and denotes to depolarization (δ) and lidar ratio (LR) values at the three lidar wavelengths λ (6 values in total), so that n = 6. “M” denotes to measurements and “S” denotes to simulations. If we expand Eq. 8 we get:

$$\frac{(\delta_{355}^{M} - \delta_{355}^{S})^2 + (\delta_{532}^{M} - \delta_{532}^{S})^2 + (\delta_{1064}^{M} - \delta_{1064}^{S})^2 + (LR_{355}^{M} - S_{355}^{S})^2 + (LR_{532}^{M} - S_{532}^{S})^2 + (LR_{1064}^{M} - S_{1064}^{S})^2}{(\delta_{355}^{M} + \delta_{532}^{M} + \delta_{1064}^{M} + LR_{355}^{M} + LR_{532}^{M} + LR_{1064}^{M})} = \mathbf{min}$$

While for Eq. 9 we get:

$\delta_{355}^{S} \leq e(\delta_{355}^{M})$, $\delta_{532}^{S} \leq e(\delta_{532}^{M})$, $\delta_{1064}^{S} \leq e(\delta_{1064}^{M})$, $LR_{355}^{S} \leq e(LR_{355}^{M})$, $LR_{532}^{S} \leq e(LR_{532}^{M})$, $LR_{1064}^{S} \leq e(LR_{1064}^{M})$

We also realized that we erroneously reversed the order of these two equations in the manuscript. Equation 9 actually precedes Eq. 8.

We revised our methodology according to the reviewer's suggestions: First, we search through the pre-calculated T-matrix look-up-tables for all the results that satisfy Eq. 9 (i.e. for each measured δ and LR, at each wavelength λ, the simulated value must be within the corresponding measurement error e). These are then the "possible solutions". Then, the best solution is selected based on the minimization criteria of Eq. 8.

Based on the reviewer comments we see now how the notation in Eq. 8 and 9 can be confusing. We updated it in the revised version, and we also included the measurement error "e" in the denominator of Eq. 9:

$$\left|\delta_{\lambda}^{M} - \delta_{\lambda}^{S}\right| \leq e(\delta_{\lambda}^{M}) \text{ and } \left|LR_{\lambda}^{M} - LR_{\lambda}^{S}\right| \leq e(LR_{\lambda}^{M}) \qquad \textbf{(Eq. 8)}$$

$$\sum_{\lambda = 355,\, 532,\, 1064} \left(\left(\frac{\delta_{\lambda}^{M} - \delta_{\lambda}^{S}}{e\,(\delta_{\lambda}^{M})} \right)^2 + \left(\frac{LR_{\lambda}^{M} - LR_{\lambda}^{S}}{e\,(LR_{\lambda}^{M})} \right)^2 \right) = \mathbf{min} \qquad \textbf{(Eq. 9)}$$

Again "M" denotes to measurement, "S" to simulation and λ to wavelength.

Following the reviewer's suggestion, we present our results in the updated manuscript, including all possible solutions (Tables 3,4 and Figures 7-10).

For the near-spherical particles we found ten possible solutions, with the mean geometric radius r_g to range from 0.25 to 0.45μm ($reff$ = 0.4 to 0.7μm), the refractive index to range at m_r = 1.35-1.55 and m_i = 0.005-0.03, while for the shape distribution the mean axial ratio ε_s ranges at 1.1-1.4. We should note that although significantly larger values of m_i were included in the updated simulations (originally was m_i (max) = 0.05, while now m_i (max) = 0.5), no possible solutions were found for m_i higher than 0.03.

The solution selected, that minimizes, the new cost function in Eq. 9 in updated manuscript, is r_g = 0.25μm ($reff$ = 0.38μm), m_r = 1.55, m_i = 0.03 and ε_s = 1.3, which is the same with the solution in the original version of the manuscript. By adding the error of the measurements to the denominator of Eq. (9), solutions were provided also using Chebyshev particles of second (T2) and fourth (T4) degree. For T2 the deformation parameter ranges from u = –0.25 to 0.15, while for T4 particles only one solution was found with u = -0.1. These u values suggest small deviations from sphericity, meaning that these morphologies also resemble near-spherical shapes. For Chebyshev particles, slightly larger particles were retrieved (maximum r_g found is 0.55μm ($reff$ = 0.8μm) while for the refractive index, both the imaginary and the real part were found to exceed the values found for near-spherical particles. Specifically, m_i ranges from i0.005 to i0.055 and m_r ranges from 1.35 to 1.8.

We included the following in the manuscriot to highlight this comment (***page 5, line 3***):

"Following this methodology, for the near-spherical particles ten possible solutions were found, listed in Table 3 along with the resulting cost functions calculated by Eq. (8). For these solutions, the mean axial ratio ε_s of the particles covers the range 1.1 to 1.4 while the mean geometric radius is always higher than 0.25μm ($reff$ = 0.4μm) and up to 0.45μm ($reff$ = 0.7μm). For the complex refractive index m, the imaginary part m_i does not exceed i0.03, while the real part m_r takes values of 1.35 to 1.55. The minimization of the cost function (Eq. 8) is achieved for near-spherical particles with ε_s = 1.4 (Fig.9), m = 1.55 + i0.025 and r_g= 0.25μm (Fig. 10), suggesting a strong accumulation mode for the size distribution of the particles, with sufficiently small m_i so as the characteristic enhancement in PLDR does not wash out due to the strong absorption (Bi et al., 2018). All possible solutions as well as the solution that minimizes the cost function are presented in Fig. 8 and 9."

And (***page 5, line 4***):

"For Chebyshev particles of second (T_2) and fourth degree (T_4) used herein, the search in the constructed look-up-tables provided the solutions listed in Table 4. For all the solutions, deformation parameter for Chebyshev particles of the second degree ranges from u = –0.25 to 0.15, while for particles of the fourth degree only one solution was found with u = -0.1. These u values suggest small deviations from sphericity, meaning that these morphologies also resemble near-spherical shapes. Only for two cases the size of the particles was found to be larger than for the near-spherical shaped particles. In particular r_g ranges from 0.15μm ($reff$ = 0.2μm) to 0.55μm ($reff$ = 0.8μm). For the complex refractive index, values in some cases exceed the corresponding values for near-spherical particles. The imaginary part m_i ranges from i0.005 to i0.055,and the real part m_r ranges from 1.35 to 1.8. The minimization of the cost function (Eq. 8) is achieved for Chebyshev particles of the second degree with u = -0.25 (resembling an oblate near-spherical particle), complex refractive index m = 1.65 + i(0.03) and mean geometric radius r_g = 0.2μm. For Chebyshev particles of the fourth degree, the sole solution presented values u = -0.1, m = 1.35 + i(0.01) and r_g = 0.55μm. All possible solutions as well as those that minimize the cost function are presented in Fig. 10 and 11."

Simulations were also performed for other reported spectrally dependent PLDR and LR values found in the literature for smoke exhibiting large PLDR values (Burton et al., 2015; Hu et al., 2017; Ohneiser et al., 2020). Results for these fittings are presented in the manuscript Supplement (Tables 4-9 and Figures 4-9). Results are in line with the results presented for the case study of Haarig et al. (2017).

11. In the figure 10 comparison with AERONET, the use of a generic biomass burning solution instead of a solution for the same smoke plume seems like a needless shortcut that undercuts your ability to draw conclusions from it. I realize there were no precisely coincident AERONET measurements, but a previous paper studying the same event that you already reference by a coauthor (Haarig et al. 2019) shows an AERONET retrieval that is at least of the same smoke plume, and also apparently better agreement, so clearly it's possible to get better fidelity than the unrelated generic case given by Dubovik et al. (2002). Furthermore, comparing a fit to a mono-modal size distribution to the fit from a bimodal size distribution and then noting that the modes don't line up is not particularly useful, and it's not obviously tied to the presence or absence of near-spherical particles per se. If you must compare a mono-modal fit to a bimodal fit, then at least calculate the effective radius and variance (quantities that are more comparable from different distribution types) from each of them and compare that instead.

REPLY: We agree with the reviewer. Unfortunately, in AERONET Version 3 Inversion products, the quality-assured Level 2 data have excluded the size distribution presented in Haarig et al. (2018). The same holds for Level 2 data of Version 2 Inversion products (the previous AERONET version):

https://aeronet.gsfc.nasa.gov/cgi-bin/type_one_station_opera_v2_inv2?site=MetObs_Lindenberg&nachal=0&year=25&month=7&day=22&aero_water=0&level=2&if_day=0&if_err=0&place_code=10&year_or_month=0

https://aeronet.gsfc.nasa.gov/cgi-bin/data_display_inv_v3?site=MetObs_Lindenberg&nachal=0&year=2017&month=8&day=23&aero_water=0&level=2&if_day=0&if_err=0&place_code=10&DATA_TYPE=76&year_or_month=0

We can speculate that this particular retrieval was rejected from Level 2 by failing to fulfill some criteria for the quality control or for the success of the fit (i.e. the sky residual error is as high as 6.5%), thus we cannot include it in the paper. However, in order to reply to the reviewer's question, since we agree that the comparison with the climatological-mean size distribution for biomass burning aerosols does not provide a solid conclusion, we performed the comparison with the AERONET retrievals discussed in Haarig et al. (2018). We agree with the reviewer that the comparison using the effective radius $reff$ and effective variance $veff$ is more meaningful and we present the comparison with the AERONET data in Table 1 below. We can see that for AERONET retrievals $reff$ is slightly higher (by 10%) while $veff$ is much higher (by 33%).

The calculation of r_g and $veff$ from $reff$ and σ_g provided by AERONET, is done using Eq. 1 and 2 (Hansen and Travis, 1974):

$$v_{eff} = \exp\left(\ln(\sigma_g^2)\right) - 1 \qquad \textbf{(Eq. 1)}$$

$$r_{eff} = r_g(1 + v_{eff})^{5/2} \qquad \textbf{(Eq. 2)}$$

Table 1. Comparison of $reff$ and $veff$ for the size distributions retrieved by AERONET on 23 August, 2017 at Lindenberg site, and the size distribution retrieved from the measurements at Leipzig on the same date, using near-spherical particles.

		AERONET	
Date	**Time**	$reff$	$veff$

23.08.2017	05.42.40	0.42 μm	0.24
Near spherical particles			
		0.38 μm	0.18

(*) Provided by AERONET
(**) Calculated using Eq. (1) and Eq. (2)

References *(that are not included in the initial version of the manuscript)*:

Hansen, J.E., Travis, L.D. Light scattering in planetary atmospheres. Space Sci Rev 16, 527–610 (1974). https://doi.org/10.1007/BF00168069

12. On a related note, could there be a coarse mode that your model is ignoring that might explain some of the features of your observations? Have you tried to eliminate the possibility of an optically significant coarse mode?

REPLY: The features observed by the ground-based lidar measurements (Haarig et al., 2018) are adequately reproduced by the assumed size distribution. Similar simulations presented by Bi et al., (2018; Fig 2), suggest that for near-spherical particles the measured spectral dependence of PLDR (i.e. steeply decreasing from UV to Near-IR) could not be reproduced by coarse mode particles. Thus, an optically significant coarse mode would have to be investigated with a different shape model.

The following has been added to highlight the reviewer's comment (**page 8, line 22**): "A common feature observed in our results is that the ground based lidar measurements are adequately reproduced by the assumed monomodal size distributions for which the mean particle geometric radius r_g did not exceed 0.55μm. This fits well with in-situ measurements of aged smoke particles (i.e. Dahlkoetter et al., 2014). The presence of a pronounced accumulation mode is also suggested by the extinction related Angstrom exponent (EAE) values measured in Leipzig. According to Eck et al. (1999), a strong spectral slope in EAE can be associated with a prominent accumulation mode of the size distribution for smoke particles. Similar simulations presented by Bi et al., (2018; Fig 2), suggest that for near-spherical particles the measured spectral dependence of PLDR (steeply decreasing from the UV to the Near-IR) could not be reproduced by coarse mode particles. Thus, the possibility of an optically significant coarse mode would have to be investigated with a different shape model. In any case though, the retrieved fine mode is in good agreement with in-situ measurements of aged smoke particles (i.e. Dahlkoetter et al., 2014). The presence of a pronounced accumulation mode is also suggested by the extinction related Angstrom exponent (EAE) measured in Leipzig (-0.3±0.4 at 355/532nm and 0.85 0.3 at 532/1064nm). According to Eck et al. (1999), a strong spectral slope in EAE can be associated with a prominent accumulation mode of the size distribution for smoke particles."

13. Page 7, line 10. This brief sentence about extending AERONET to include near spherical particles is an interesting idea, but unsupported. To address this properly, please consider at least these 3 points. First, as stated above, there would have to be a fair comparison between AERONET results and your results for similar cases. As part of this, there would have to be an assessment of not just the size distribution, but also a reconstruction of the lidar measurements using the AERONET solution.

- *Can you show that the AERONET retrieval fails to reproduce the lidar ratio and linear particle depolarization ratio adequately?*
- *Conversely, how does your near-spherical model do in reproducing AERONET radiances at all AERONET wavelengths?*

Whatever the answer to each of these questions, there's something to be learned. If the near-spherical model does a better job of reproducing lidar measurements and is also better at modeling AERONET measurements, then it could genuinely be an improvement for AERONET. If it doesn't improve the AERONET fits but improves the lidar fits, it might be less useful for AERONET alone (at least it would suggest that it might be hard for AERONET to operationally use the model if there is not sufficient measurement information content to distinguish near-spherical from spherical particle shapes), but might still be potentially of significant value for combined AERONET-lidar retrievals (e.g. constrained backscatter lidar retrievals). Even if the near-spherical model does a worse job at modeling AERONET measurements but a better job at modeling lidar measurements, it at least points us to the need for further modeling studies to find a single model that can unify both types of measurements.

We thank the reviewer for the constructive comment, which gives us the opportunity to provide a more thorough analysis in the updated manuscript, that better positions our research.

Considering the first question, unfortunately there are no available AERONET retrievals that we can use (please see reply for Comment 11).

We should note though that the recent study by Hu et al. (2019) for the same stratospheric smoke used the forward model of AERONET and failed to reproduce the measured depolarization ratios, indicating the limitations of this model for non-spherical smoke particles. Specifically, Hu et al. (2019) used the GARRLiC algorithm (Lopatin et al., 2013), which combines lidar with sunphotometer measurements in order to derive the optical and miscrophysical properties of particles in the atmosphere. GARRLiC utilizes the forward model of AERONET (Dubovik et al., 2006).

As mentioned above, the inversion retrievals for Lindenberg site on 23 August 2017 at 05:42 UTC were rejected both from Level 1.5 and Level 2 quality-assured data of AERONET Version 3 algorithm. Products from Version 2 (the AERONET version used for the products presented in Haarig et al. 2018) does not include lidar ratio and depolarization ratio values. Thus, there are no available, coincident AERONET lidar ratio and depolarization ratio products to compare.

Considering the second question, instead of reproducing the sun-photometer measurements (which entails taking into account the multiple scattering in the atmospheric column), we reproduce the phase function of the stratospheric smoke particles, as a first-level approximation (single scattering) of the full solution. Since there are no available retrievals of phase functions from AERONET for this case to compare (please see reply for Comment 11), we follow an invert procedure to reply to reviewer's comment. We specifically try to reproduce the calculated phase function of near-spherical particles, using the non-spherical model of AERONET. As shown below this is not possible, at least within a residual error of <5% (Holben et al., 2006), indicating the limitations of the AERONET non-spherical model in reproducing the scattered light from near-spherical particles in the atmosphere.

Specifically, in Fig. 4-7 we present the phase function at 440nm calculated for the near-spherical stratospheric smoke particles (purple line in the plots), and the comparison with the phase functions at 440nm calculated using the AERONET non-spherical model, using a large suite of size distributions and refractive indices (blue lines). In Fig. 4-7 we show the results only for r_g of 0.1, 0.25, 0.5 and 1μm, respectively, for all refractive indices. The complete set of results (more r_g and AERONET wavelengths of 670, 870 and 1020nm) are provided in the Supplement.

In Fig. 4-7, the phase function is presented in the left plots, whereas we also include the degree of linear polarization (-P12/P11) in the middle plots and the values of P22/P11 in the right plots, for the sake of completeness. On a relative note, the large differences seen for -P12/P11 and P22/P11 highlight the higher information content for near-spherical particles in polarized measurements (as the PLDR measurements).

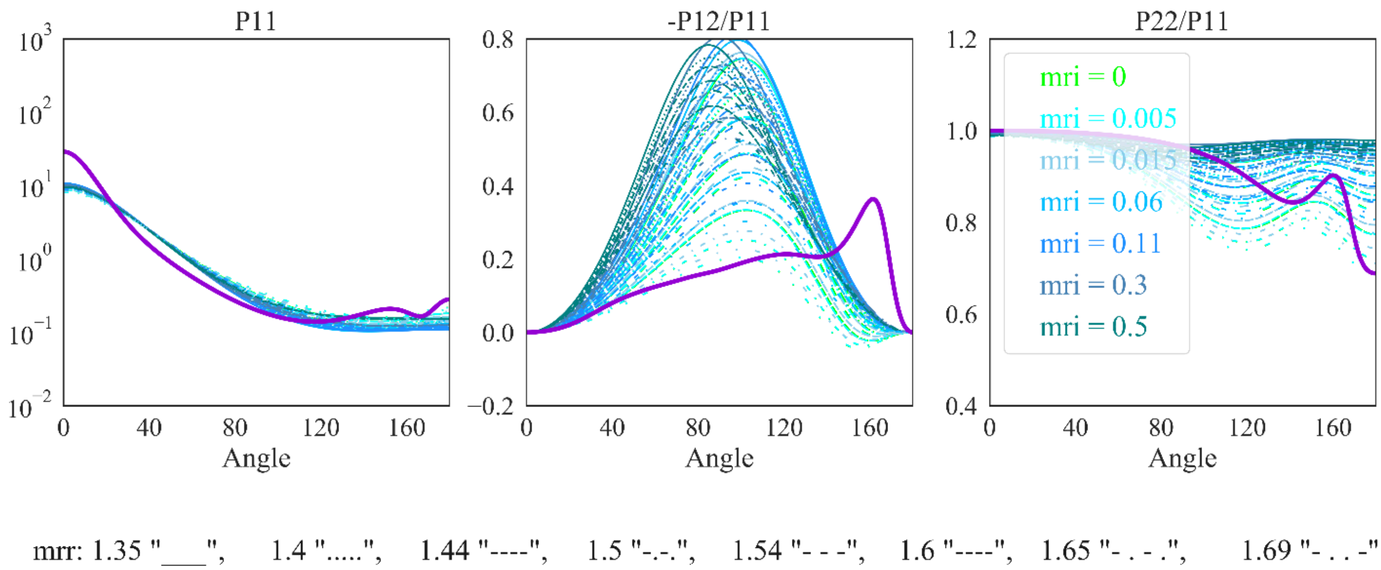
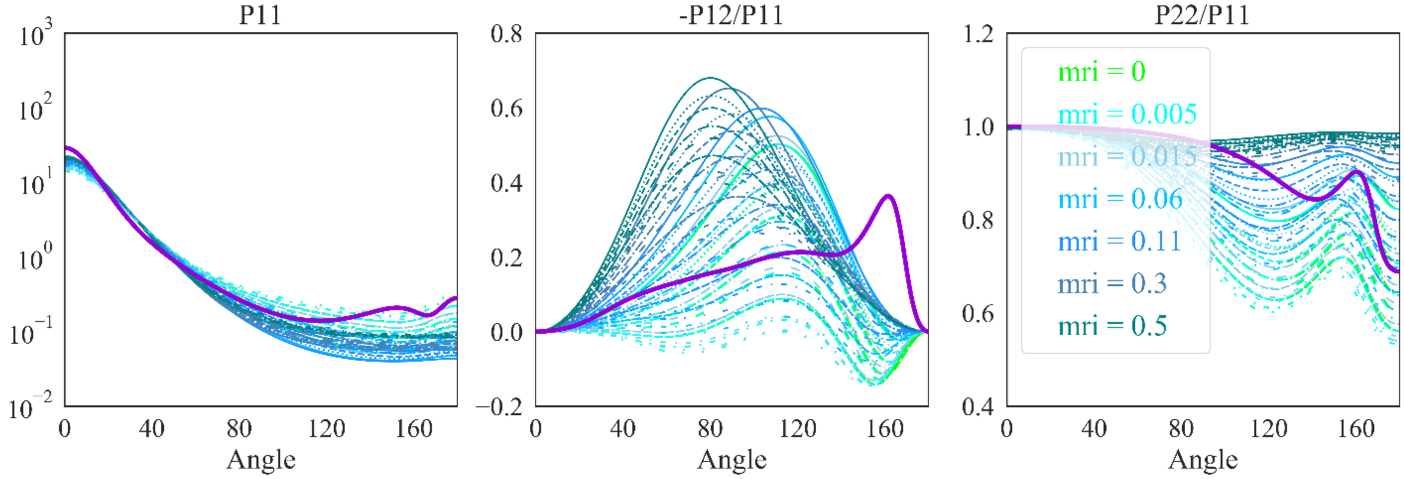
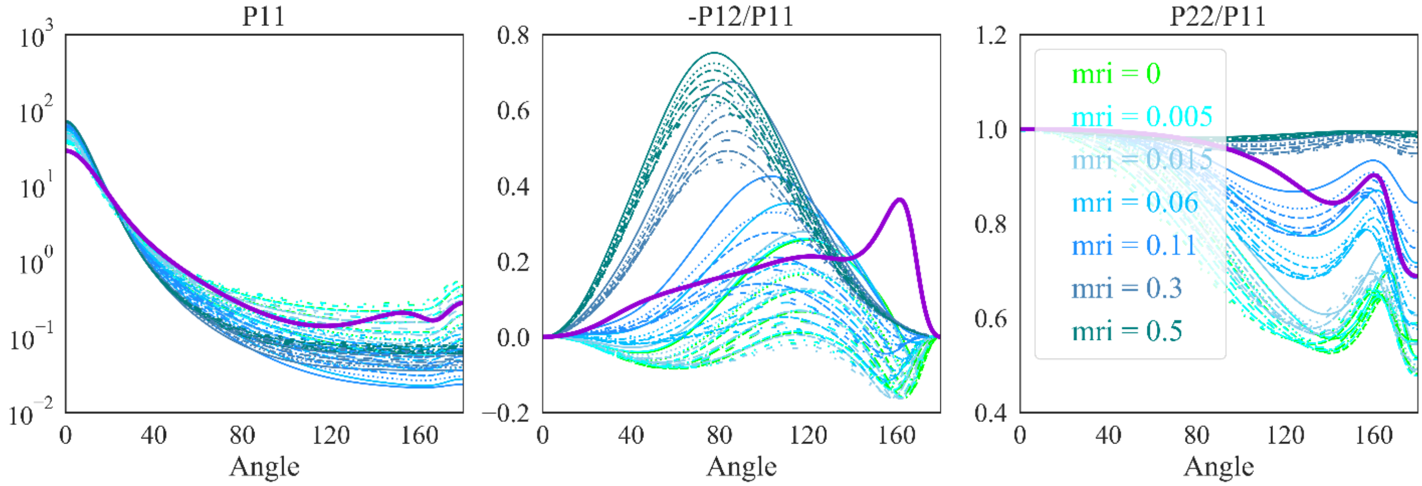


Figure 4. The elements of the scattering matrix at λ = 440nm. Left: P11 (phase function), middle: -P12/P11 (degree of linear polarization), right: P22/P11. Purple lines in the plots: calculations considering the near-spherical particle properties derived for the stratospheric smoke particles from the Canadian fires, with mean axial ratio ε_s = 1.3, monomodal, log-normal size distribution with r_g = 0.25, σ_g = 0.4, and complex refractive index m = 1.55 – i0.03. Blue lines in the plots: calculations considering the shape distribution of the AERONET non-spherical model, and monomodal, log-normal size distributions with r_g = 0.1μm and refractive indices of m_r = 1.35, 1.40, 1.44, 1.50, 1.54, 1.60, 1.65, 1.69 for the real part (different line styles in the plot) and m_i = 0, 0.005, 0.015, 0.06, 0.11, 0.3, 0.5 for the imaginary part (different line colors in the plot).



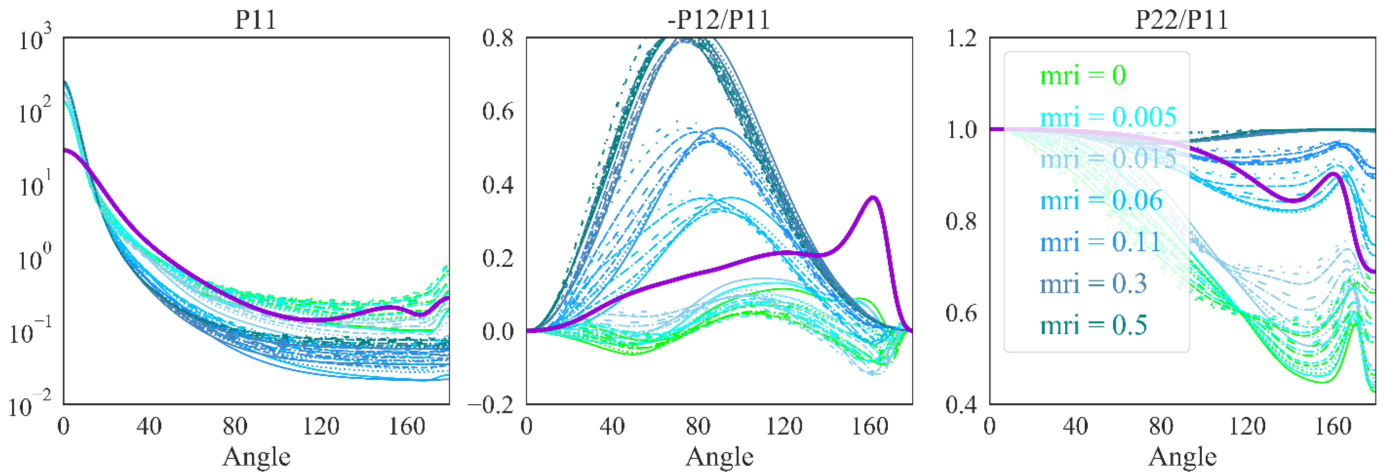
mrr: 1.35 "___", 1.4 ".....", 1.44 "----", 1.5 "-.-.", 1.54 "- - -", 1.6 "----", 1.65 "- . - .", 1.69 "- . . -"

Figure 5. Same as Fig. 4. For the calculations with the AERONET non-spherical model (blue lines) we consider r_g= 0.25µm.



mrr: 1.35 "___", 1.4 ".....", 1.44 "----", 1.5 "-.-.", 1.54 "- - -", 1.6 "----", 1.65 "- . - .", 1.69 "- . . -"

Figure 6. Same as Fig. 4. For the calculations with the AERONET non-spherical model (blue lines) we consider r_g= 0.5µm.



mrr: 1.35 "___", 1.4 ".....", 1.44 "----", 1.5 "-.-.", 1.54 "- - -", 1.6 "----", 1.65 "- . - .", 1.69 "- . . -"

Figure 7. Same as Fig. 4. For the calculations with the AERONET non-spherical model (blue lines) we consider r_g = 1.0μm.

In order to quantify the residual of fitting we calculate the "residual error" (Err) as shown in Eq. 3 (https://aeronet.gsfc.nasa.gov/new_web/Documents/Inversion_products_for_V3.pdf).

$$Err = \sqrt{\frac{\sum_{i=1}^{n}(lnf^{*} - lnf)^2}{N}} * 100 = \%Error \qquad \textbf{(Eq. 3)}$$

where $f*$ denotes the P_{11} values calculated with the near-spherical model, f denotes the P_{11} values calculated with the AERONET non-spherical model, and N is the number of values, in terms of wavelengths and scattering angles. Specifically, we consider the four wavelengths of 440, 670, 870 and 1020nm and the scattering angles from 0 to 150 degrees.

Figure 8 shows the residual error "Err" for fitting the phase function of the near-spherical particles presented in the manuscript with the phase functions calculated with the AERONET non-spherical model, considering the big suite of sizes and refractive indices discussed above. The minimum residual error is 9.4%, whereas the limit of a successful AERONET retrieval is 5% (Holben et al., 2006). However, the latter threshold denotes to the multiple-scattered light, which may mask the big differences seen in the single-scattering properties in Fig. 8.

Similar 2D plots for the residual error considering only the wavelengths at 440, 670, 870 and 1020nm are provided in the Supplement.

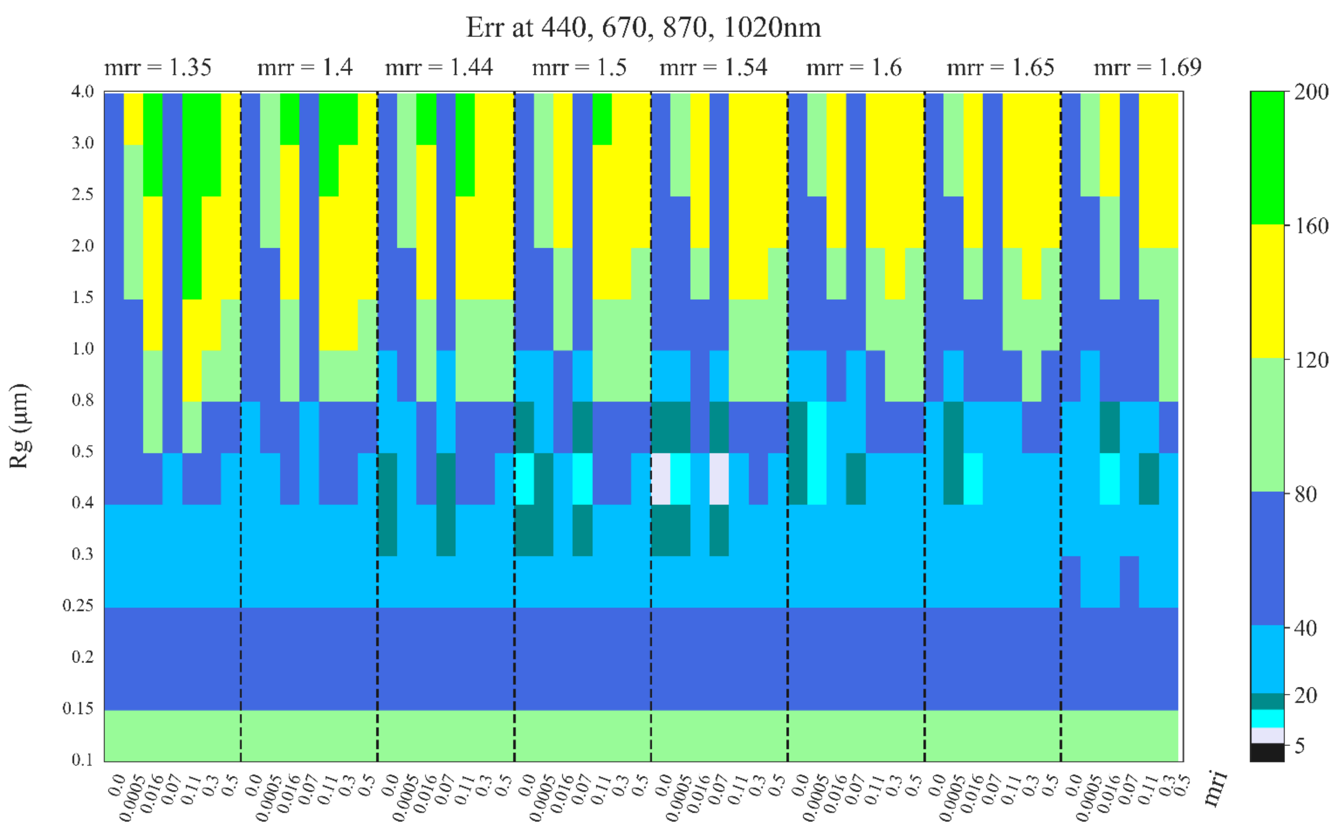


Figure 8. The residual error (Err) of fitting the phase functions at 440, 670, 870 and 1020nm of the near-spherical particles presented in the manuscript, with the phase functions calculated with the AERONET non-spherical model, for r_g and m shown in y- and x-axis, respectively.

Based on the above w a new subsection in Discussion section of the paper, entitled:

5.1 Comparison with AERONET products, elements of the scattering matrix

We understand that this analysis is not conclusive and in order to have a clear understanding of whether the near-spherical shape model could in fact improve the AERONET retrieval for stratospheric smoke, further analysis is imperative. We consider this only as a first step and we make sure to emphasize this in the manuscript.

References *(that are not included in the initial version of the manuscript)*:

Holben, B. N., T. F. Eck, I. Slutsker, A. Smirnov, A Sinyuk, J. Schafer, D. Giles, O. Dubovik, 2006: Aeronet's Version 2.0 quality assurance criteria, Proc. SPIE 6408, Remote Sensing of the Atmosphere and Clouds, 64080Q, doi:10.1117/12.706524.

Lopatin, A., Dubovik, O., Chaikovsky, A., Goloub, P., Lapyonok, T., Tanré, D., and Litvinov, P.: Enhancement of aerosol characterization using synergy of lidar and sun-photometer coincident observations: the GARRLiC algorithm, Atmos. Meas. Tech., 6, 2065–2088, https://doi.org/10.5194/amt-6-2065-2013, 2013.

14. Fractal aggregates: *Fractal aggregates require a lot of parameters to describe them, and some of them you held fixed instead of varying. If you cannot explore the full parameter space, how do you know that fractal aggregates can't fit the observations? At line 25, you then point out that previous authors (not just Ishimoto et al. 2019, but see also Kahnert 2018, Kanngeisser and Kahnert 2018, Luo et al. 2019 and Ceolato et al. 2018, as noted above) have already established that bare aggregates are not able to reproduce measurements as well as coated aggregates, so I don't see a lot of value in running a model type that has already been shown not to work without also running a related model type that has been used with some success in the past (granted with less complete measurements in the past; indeed, the new measurements are the real strength of this contribution and where you have the opportunity to go beyond prior work).*

REPLY: We included this analysis in order to emphasize the fact that fractal aggregates do not reproduce the measured optical properties of stratospheric smoke from the Canadian fires. Considering the limited parameter space by using only fixed, typical values for the fractal dimension D_f, we refer the reviewer to the comprehensive study of Paulien et. al. (2019), which shows that this parameter has only weak influence on the lidar-relevant optical properties. Moreover, in order to define the different monomer number N_m = 244, 579 and 1953 and monomer radius r_m = 40, 60 and 80 nm (volume equivalent radius of R_v= 500 nm) we followed the work of Ceolato et. al (2018). However, we see the reviewer's point on the limited range of parameters used to describe fractal aggregates and we agree this is not an excessive analysis, so we decided to remove this section from the revised version of the manuscript.

References *(that are not included in the initial version of the manuscript)*:

Paulien, L., Ceolato, R., Soucasse, L., Enguehard, F., Soufiani, A.: Lidar-relevant radiative properties of soot fractal aggregate ensembles. Journal of Quantitative Spectroscopy and Radiative Transfer, Elsevier, 2019, 241, pp.106706, DOI: 10.1016/j.jqsrt.2019.106706.

Ceolato, R., Gaudfrin, F., Pujol, O., Riviere, N., Berg, M. J., Sorensen, C. M.: Lidar cross-sections of soot fractal aggregates: Assessment of equivalent-sphere models, Journal of Quantitative Spectroscopy and Radiative Transfer, Volume 212, 2018, Pages 39-44, ISSN 0022-4073, https://doi.org/10.1016/j.jqsrt.2017.12.004.

15. page 9, line 16. See also Kablick et al. 2018 for another case discussing ice.

REPLY: Thank you, this part has been removed from the manuscript.

16. page 9, line 17. In 2015, Burton et al. could not have extensively discussed studies that were published 3 or 4 years later. Specifically, there's no discussion in Burton et al. 2015 about the ice hypothesis. It would be better for this manuscript's authors to take the opportunity to address these newer theories more completely here.

REPLY: Thank you, this part has been removed from the manuscript.

17. page 9, line 17-18. I'm not following the statement "soil lifting ...could explain the...observations presented in this study". Do you mean to eliminate this possibility or support this possibility?

REPLY: This part is now removed from the original version of the manuscript.

18. page 7, line 18. The Angstrom exponent is indeed confusing, primarily because we don't know what wavelengths you're referring to either in the measurements or in the comparison dataset that causes you to say this is "low". It's not uncommon for smoke measurements to have significant curvature in the spectral AOD (or extinction) (see Eck et al. 1999) and in fact Haarig et al. 2018 show a significant difference between the 355-532 nm and 532-1064 nm Angstrom exponents for their analysis of this smoke plume. Taking this into account, do you still believe the Angstrom exponent for this case indicates coarse mode particles? Please add a more complete discussion.

REPLY: We thank the reviewer for noticing this. Indeed, as the reviewer points out, there is a strong curvature between the AE at 355/532nm (-0.3 ± 0.4) and the corresponding values at 532/1064nm (0.85 ± 0.3). According to Eck et al. (1999) this behavior can be associated with a size distribution presenting a pronounced accumulation mode, as well as a spectral dependence in the absorption, with higher absorption at the smaller wavelengths. This is in agreement with the retrieved size distribution for near-spherical particles of r_{eff} = 0.38 μm. The spectral dependence of the absorption can indeed be another reason, although it is not shown in our results due to the assumed spectrally-independent value of the imaginary part of refractive index. To address this comment we rephrased this part of the manuscript (**page 8, line 22)**:

"A common feature observed in our results is that the ground based lidar measurements are adequately reproduced by the assumed monomodal size distributions for which the mean particle geometric radius did not exceed 0.35μm. This fits well with in-situ measurements of aged smoke particles (i.e. Dahlkoetter et al., 2014). The presence of a pronounced accumulation mode is also suggested by the extinction related Angstrom exponent (EAE) values measured in Leipzig. According to Eck et al. (1999), a strong spectral slope in EAE can be associated with a prominent accumulation mode of the size distribution for smoke particles. Similar simulations presented by Bi et al., (2018; Fig 2), suggest that for near-spherical particles the measured spectral dependence of PLDR (steeply decreasing from the UV to the Near-IR) could not be reproduced by coarse mode particles. Thus, the possibility of an optically significant coarse mode would have to be investigated with a different shape model. In any case though, the retrieved fine mode is in good agreement with in-situ measurements of aged smoke particles (i.e. Dahlkoetter et al., 2014). The presence of a pronounced accumulation mode is also suggested by the extinction related Angstrom exponent (EAE) measured in Leipzig (-0.3±0.4 at 355/532nm and 0.85 0.3 at 532/1064nm). According to Eck et al. (1999), a strong spectral slope in EAE can be associated with a prominent accumulation mode of the size distribution for smoke particles."

References *(that are not included in the initial version of the manuscript)*:

Eck, T. F., Holben, B. N., Reid, J. S., Dubovik, O., Smirnov, A., O'Neill, N. T., Slutsker, I., and Kinne, S.: Wavelength dependence of the optical depth of biomass burning, urban, and desert dust aerosols, Journal of Geophysical Research: Atmospheres, 104, 31333-31349, 10.1029/1999JD900923, 1999.

19. page 7, line 21 "surface roughness alterations"? I don't follow where this idea comes in the paper. Is this what the Chebyshev particle shape is meant to represent? There are other model representations of surface roughness (e.g. Liu et al. 2013, Kemppinen et al. 2015) so this label is probably too non-specific (vague) in this context and should be made more precise.

REPLY: This part is now removed from the original version of the manuscript.

20. For the comments 20 to 24: At the current stage the discussion on the possible physical explanation for the smoke PLDR values is removed from the manuscript. More efforts will be made to investigate this issue and re-assess the possible coexistence of smoke particles and particles of sulfuring nature and whether this could affect the former in such a way to form near-spherical particles.

21. While informal titles can catch people's attention, I wonder if this one is really a good idea? "The new black" means "fashionable" or "popular", which is probably not quite what you're hoping for from your new smoke particle model. In general, I appreciate funny titles but if it is a pun, I'm not getting it, since I don't see what scientific meaning "black" has in this title. Also, the phrase "the new black" is itself something of a fad which may fade quickly, leaving readers 5 or 10 years from now completely confused about what the title means. But of course it is up to the author; this isn't a comment that needs a response, just a perspective on how one reader sees it, in case you find this helpful.

REPLY: We thank the reviewer for this comment.

22. page 2, line 26. The sentence starting "implication for enrichment of smoke plumes with dust particles" should probably be deleted. Later on, you mention several possible explanations from previous literature, but here you only mention one without discussion that you later dismiss. Better to delete it here and hold the discussion until you are ready for it in the later section.

REPLY: Thank you, we removed this phrase from the introduction.

23. page 7, line 21 "not currently supported by other observation evidence found in the literature". Probably should be reworded. It seems like you're saying there is other evidence found in the literature that doesn't support your finding, but my understanding is that there is no contradictory observational evidence because lidar ratio at 1064 nm for this kind of observation has never been reported before! Please make this clearer. Having unique measurements is a real strength! No need to muddy the picture with imagined controversy.

REPLY: The following has been rephrased (**page 7, line 21**): "…although there are currently no other available measurements of the LR of smoke particles at this wavelength. "

24. page 7, line 11, probably delete the brief mention of pollen, which has not been addressed at all in this paper. We have no way of knowing whether the near-spherical model has any success in modeling pollen.

REPLY: Thank you, it has been deleted.

25. page 9, line 12, "results in near-spherical shapes" should be reworded. Logically, the finding that the near-spherical particle model reproduces a set of measurements better than a few other models could still be merely a very useful approximation. It does not necessarily mean the particles are literally shaped like Figure 2.

REPLY: This part has been removed from the manuscript.

26. page 9, line 12, "previous studies" should be "some previous studies" (i.e. but not all, see for example Murayama et al. 2004, Burton et al. 2015 who specifically dispute it for certain other cases).

REPLY: This part has been removed from the manuscript.

27. page 9, line 14, Sugiomoto et al. what year? (typo)

REPLY: Thank you, this part has been removed from the manuscript.

28. page 9, line 23, "advocate dissuasive towards" should be changed to (e.g.) "argues against"

REPLY: Thank you, this part has been removed from the manuscript.

29*. page 9, line 26, "while up to now the LR values are not reproduced either". Again, I feel like this should be reworded, because it's making a confusing (or perhaps just incomplete) point when a much stronger one is indicated. Most previous modeling papers did not have the opportunity to reproduce three-wavelength lidar ratios because these observations have only just recently been published. The strength of the current manuscript is these new and unique measurements.*

REPLY: Actually in the study we are referring to, it is explicitly stated that tar ball aggregates were not found to reproduce the PLDR values at 532nm, as presented by Haarig et al. (2018) and Hu et al. (2019). However, we deleted this statement.

30. page 9 line 30. I think 15 micrometer monomers must be a typo.

REPLY: Thank you, this part has been removed from the manuscript.

31. page 10 line 4. "Thought" should be "Although"'

REPLY: Thank you, this part has been removed from the manuscript.

32. Table 4 caption. Please specify the instrument that made these measurements.

REPLY: We added the instrument, it is the BERTHA (Backscatter Extinction lidar-Ratio Temperature Humidity profiling Apparatus) multi-wavelength polarization Raman lidar system.

33. Figure 6. The red dashed lines appear to be in the wrong place.

REPLY: An updated the figure has been provided.

34. It would be good to include a figure explaining what Chebyshev particles look like.

REPLY: Thank you, we added a figure.

35. Figure 7. What do the white pixels near the top of the layer signify, in both the linear depolarization ratio and the water vapor mixing ratio? What do the black down-arrows on the latitude and longitude axes represent? Please indicate in 7c the portions of the track that are represented in 7a and 7b.

REPLY: The figure has been updated.

36. Figure 14. Please mark the location of the smoke plume and consider plotting this on an altitude (rather than pressure) scale and with altitude range more comparable to the ranges shown in Figures 6 and 7. Please also consider putting an indicator of distance scale on Figures 6, 7, and 14.

REPLY: The figure has been removed from the manuscript.