The manuscript topic fits well within the journal scope is providing new insights on biomass burning aerosol layers. Nevertheless, it needs major revisions before being ready for publication.

REPLY: Thanks for your helpful comments. Corrections have been made considering your suggestions as well as other reviewers'. Please find our point-by-point response and first revised version in the supplement. Remark: The figure numbers and the page numbers in the referee comments and in our replies correspond to the original manuscript.

Major comments:

1. A substantiated and consolidated verification of the measurement quality and the potential role of systematic errors affecting the measurements is a preliminary paramount step when such high PLDR values are measured. This is particularly true for stratospheric aerosols as calibration of aerosol depolarization measurements of stratospheric particles is quite difficult and cannot rely on molecular calibration approach.

REPLY: Thank you for this comment. Indeed, the calibration of the depolarization measurements is very crucial for any aerosol study. For the calibration of the depolarization measurements used in this study we followed the "*Δ±45 depolarization calibration*" method proposed by Freudenthaler et al. (2009). Specifically, for the PLDR measurements used here, the systematic errors are 0.015 at 355nm, 0.006 at 532nm and 0.007 at 1064nm as presented in Haarig et al. (2018). A detailed discussion on the parameters affecting the depolarization measurements of the BERTHA lidar system is presented in Haarig et al. (2017) (APPENDIX A).

To highlight this comment, we added the following paragraph to the manuscript (**page 7, line 14**):

"To ensure the high quality of depolarization measurements, the Δ±45 depolarization calibration method proposed by Freudenthaler et al. (2009) was followed, while the effect of different parameters on the depolarization measurements of the BERTHA lidar system has been carefully assessed and is presented in detail in Haarig et al. (2017)."

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

Freudenthaler, V., Esselborn, M., Wiegner, M., Heese, B., Tesche, M., Ansmann, A., Müller, D., Althausen, D., Wirth, M., Fix, A., Ehret, G., Knippertz, P., Toledano, C., Gasteiger, J., Garhammer, M., Seefeldner, M.: Depolarization ratio profiling at several wavelengths in pure Saharan dust during SAMUM 2006, Tellus B: Chemical and Physical Meteorology, 61:1, 165-179, DOI: 10.1111/j.1600-0889.2008.00396.x, 2009.

Haarig, M., Ansmann, A., Althausen, D., Klepel, A., Groß, S., Freudenthaler, V., Toledano, C., Mamouri, R.-E., Farrell, D. A., Prescod, D. A., Marinou, E., Burton, S. P., Gasteiger, J., Engelmann, R., and Baars, H.: Triple-wavelength depolarization-ratio profiling of Saharan dust over Barbados during SALTRACE in 2013 and 2014, Atmos. Chem. Phys., 17, 10767–10794, https://doi.org/10.5194/acp-17-10767-2017, 2017.

2.The fact that such high PLDR values were reproduced using T-matrix simulations, assuming near-spherical shapes, for biomass burning is not itself a verification of the fact that observed particles were indeed transported stratospheric smoke plumes. More information on possible particle composition, and its possible organic origin, should be inferred from other optical measurements (multi-wavelength particle extinction and backscatter measurements).

REPLY: Thank you very much for this comment. Indeed, the origin/composition of the particles cannot be deduced only from the measurements presented in the manuscript (multi-wavelength PLDR and LR measurements). Detailed discussion on the transport of the smoke plumes that are presented in our analysis is included in several previous studies referring to the Canadian wildfires of August 2017. For example, Khaykin et al. (2018) present CALIPSO data that are used to follow the evolution of the plume since two days after the PyroCb eruption on 14 August 2017 (Peterson et al., 2017) to 30 August 2017 (see Fig. 3a in supplement S2 from Khaykin et al., 2018). The ground-based lidar observations at Leipzig on 23 August 2017 presented in the manuscript, observe the smoke plume, which was located above Germany during 21 – 24 August 2017 (Khaykin et al., 2018). In Ansmann et al. (2018), HYSPLIT backward and forward trajectories were used to depict the route of the smoke plume from North America to central Europe and identify the smoke source regions. Results were found to be in good agreement with CALIPSO observations and UV aerosol index maps from OMPS presented in Khaykin et al. (2018). In Hu et al. (2019) MODIS maps, UV aerosol index from OMPS as the CO product from AIRS were used to determine whether the observed aerosol plumes over northern France were indeed smoke transported from Canada. Indeed, the strong spatio-temporal correlation between UV aerosol index and CO revealed the smoke presence. Apart from the high PLDR values measured from the ground-based lidar system in Leipzig, lidar ratio (LR) values are also available at 3 wavelengths and used in our simulations: 40 ± 16sr, 66 ± 12sr, 92 ± 27sr at 355, 532 and 1064nm. Although LR of smoke presents a large variability due to different particle characteristics between fresh and aged smoke particles, these LR values are in good agreement with past measurements for smoke LR at 355 and 532nm (i.e. Fiebig et al., 2002; Muller et al., 2005; Ortiz-Amezua et al., 2017).

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

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.

Ortiz-Amezcua, P., Guerrero-Rascado, J. L., Granados-Muñoz, M. J., Benavent-Oltra, J. A., Böckmann, C., Samaras, S., Stachlewska, I. S., Janicka, L., Baars, H., Bohlmann, S., and AladosArboledas, L.: Microphysical characterization of long-range transported biomass burning particles from North America at three EARLINET stations, Atmos. Chem. Phys., 17, 5931–5946, https://doi.org/10.5194/acp-17-5931-2017, 2017.

3. Because of 2) the proposed approach is rater weak. It is not possible to generalize statements just from a single case study. Moreover, it seems a sort of ill-posed problem and the minimum in Eq. 8 might be relative, i.e. what happens if instead a mono-modal distribution a bimodal is chosen? or a gamma instead of normal distribution? Probably Eq. 8 will provide independently a solution.

REPLY: As discussed in Hansen and Travis (1974), the sensitivity of the optical properties of the particles to different types of size distributions (e.g. standard gamma, log normal, bimodal and a power-law) is limited. Maybe the reviewer is also interested in the answer provided for a similar comment (Comment 11) made by anonymous Reviewer 3.

In the revised version of the manuscript, we have included the following (**page 4, line 20**):

"The fixed width of the size distribution σ_g is again 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). Choosing a log-normal size distribution over any other plausible type of distribution is not expected to alter our results significantly (Hansen and Travis, 1974)."

Regarding the first part of the comment, we agree with the reviewer. For this reason, we updated the manuscript, including the retrievals for all available measurements of stratospheric smoke in the literature, using the proposed near-spherical model. Figure 1 below presents some examples of successful reproduction of the measurements for all the cases assuming near-spherical shapes, and Table 2 below presents the retrieved values for the mean axial ratio ε_s of the near-spherical shapes, the complex refractive index m and the effective radius $reff$ of the particles. All the retrievals (using near-spherical and Chebyshev particles) are available in the manuscript Supplement (for Hu et al., 2019 fitting of the measurements of 31 August 2017 are presented. For Ohneiser et al., 2020 fitting of the measurements of 8 January 2020 are presented).

Furthermore, we added the following section to the text (**page 8, line 23**):

"Although the available literature on the PLDR and LR values of stratospheric smoke is for now limited, we see that we can reproduce all reported of PLDR and LR using the near-spherical shape model (Table 1-9 and Fig. 1-9 in the Supplement). All cases listed in Table 2 are associated with Pyro-cumulonimbus activity. As already mentioned the case studies of Burton et al. (2015), Hu et al. (2019) and Haarig et al. (2018) refer to Canadian smoke, while the most recent case study presented by Ohneiser et al. (2020) refer to Australian wildfires of 2019-2020. Table 5 present the retrieved mean axial ratio, complex refractive index and geometric radius of the size distribution. For Hu et al. (2019), measurements on 24, 29 and 31 August were reported. For Ohneiser et al. (2020) measurements on 8, 9 and 10 January 2020 were reported."

Table. 1: Reported PLDR and LR values for UTLS smoke. For Hu et al. (2019) and Ohneiser et al. (2020), one of the available observations is included in the table.

	PLDR$_{355}$ (%)	PLDR $_{532}$ (%)	PLDR $_{1064}$ (%)	LR$_{355}$ (sr)	LR$_{532}$ (sr)	LR$_{1064}$ (sr)
Burton et al. (2015)	20.3 ± 3.6	9.3 ± 1.5	1.8 ± 0.2	X	X	X
Hu et al. (2019)	24 ± 4	19 ± 3	5 ± 1	41 ± 7	54 ± 9	X

Ohneiser et al. (2020)	26 ± 5.2	15 ± 1.5	X	53 ± 15.9	76 ± 15.2	X

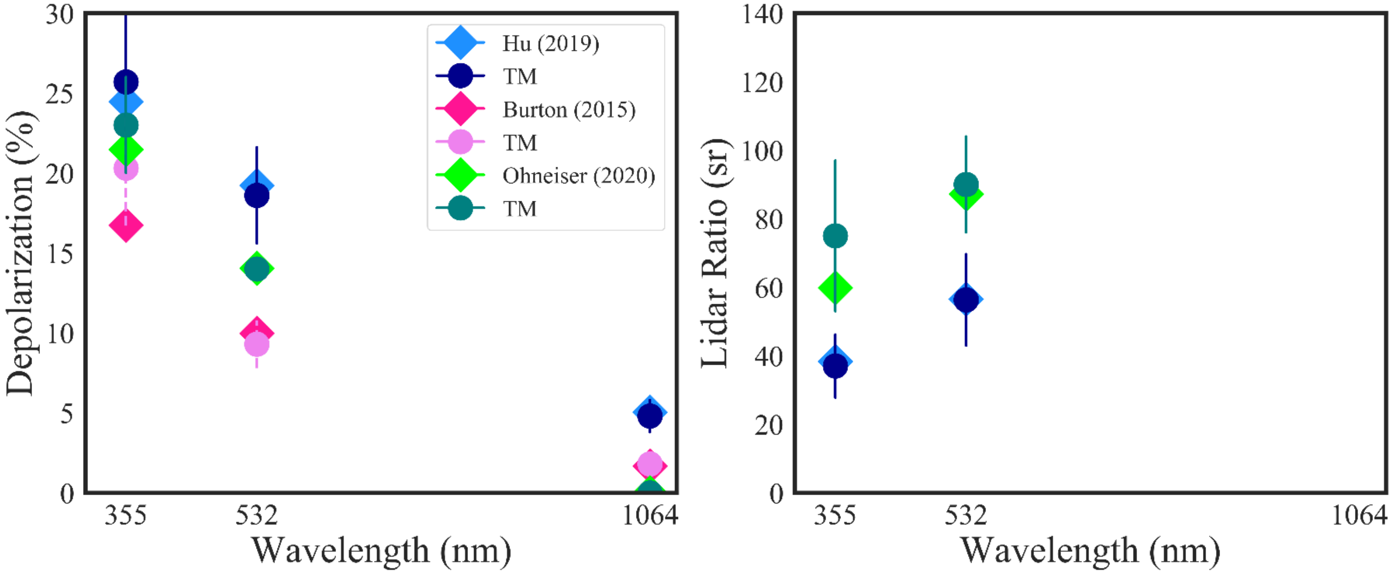

Figure 1. Example fittings of the PLDR and LR measurements presented in Hu et al. (2019), Burton et al. (2015) and Ohneiser at al. (2020), using the near-spherical model. First two cases refer to Canadian wildfires of 2017 and 2014, respectively. The third case refers to the Australian wildfires of last 2019 – 2020. All cases are associated with PyroCb activity. TM in the legend stands for the T-matrix simulations with near-spherical particles: blue circles denote to the simulations reproducing the observations of Hu (2019), pink circles denote the simulations reproducing the observations of Burton (2015), and green circles denote to the simulations reproducing the observations of Ohneiser (2020). All of the retrievals are included in the manuscript Supplement.

Table 2. The simulations with the near-spherical shape model, used to reproduce the measurements presented in Table 1.

	r_g (μm)	ε_s	m_i	m_r
Burton et al. (2015)	0.3	1.15	0.005	1.45
Hu et al. (2019)	0.25	1.45	0.02	1.55
Ohneiser et al. (2020)	0.35	0.9	0.035	1.45

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

Ohneiser, K., Ansmann, A., Baars, H., Seifert, P., Barja, B., Jimenez, C., Radenz, M., Teisseire, A., Floutsi, A., Haarig, M., Foth, A., Chudnovsky, A., Engelmann, R., Zamorano, F., Bühl, J., and Wandinger, U.: Smoke of extreme Australian bushfires observed in the stratosphere over Punta Arenas, Chile, in January 2020: optical thickness, lidar ratios, and depolarization ratios at 355 and 532 nm, Atmos. Chem. Phys., 20, 8003–8015, https://doi.org/10.5194/acp-20-8003-2020, 2020.

4. As stated by Sassen and Khvorostyanov, smoke can directly act as ice nuclei before liquid clouds form (https://iopscience.iop.org/article/10.1088/1748-9326/3/2/025006). This fact can partially explain the higher PLDR (considering a process in progress). This aspect, very likely is not mentioned in the manuscript and can be the reason of PLDR increase.

REPLY: We agree with the reviewer that the PLDR values alone could indicate the formation of ice crystals inside the stratospheric smoke layer. However, the reported PLDR values of ~20% at 532nm are small compared to those usually observed (>40%) for cirrus clouds containing ice crystals (Chen at al., 2002; Noel et al., 2002; Voudouri et al., 2020). Furthermore, the available data from Leipzig include also the lidar ratio (LR) values of 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 which present values of the order of 25 sr (Gouveia et al., 2017). 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).

We would also like to refer the reviewer to Comment 5 from anonymous Reviewer 3, who raised a similar concern on ice formation.

To highlight this for the reader we included the following in the manuscript **(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 in 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). 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."

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.

Kanji, Z. A., Ladino, L. A., Wex, H., Boose, Y., Burkert-Kohn, M., Cziczo, D. J., and Krämer, M.: Chapter 1: Overview of ice nucleating particles, Meteor Monogr., Am. Meteorol. Soc., 58, 1.1-1.33, https://doi.org/10.1175/amsmonographs-d-16-0006.1, 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.

Phillips, V. T. J., P. J.Demott, C.Andronache, K. A.Pratt, K. A.Prather, R.Subramanian, and C.Twohy, 2013: Improvements to an empirical parameterization of heterogeneous ice nucleation and its comparison with observations. J. Atmos. Sci., 70, 378–409, doi:https://doi.org/10.1175/JAS-D-12-080.1.

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.

5) The simulations themselves are not original as in fact similar simulations were performed in the past by Bi et al. 2018, Mishchenko et al. 2016; Ishimoto et al., 2019, as the authors explicitly admit. What is different with respect to those manuscript?

REPLY: Bi et al. (2018) is an interesting modeling study on the properties of spheroid and super-ellipsoid particles for a large suite of refractive indices and size parameters. It is though a generic study, not focused on stratospheric smoke particles. Also, the simulations in Bi et al. (2018) refer only to PLDR and not to other intensive properties (e.g. LR) as we do in our study. On the other hand, Mishchenko et al. (2016) used four different models to reproduce the PLDR values observed by Burton et al., (2015). Our results are comparable, but the study is only limited to PLDR since there were no available LR measurements at the time. Ishimoto et al. (2019) use fractal aggregates coated by water soluble materials. In this study both the PLDR and LR are examined, but the simulations refer only to monodisperse particles. The results are comparable to ours only for coated fractals, producing a shape that closely resembles the near-spherical shape (i.e. shapes of “Type-B, size 11, Vr = 20” shown in Fig. 4 of Ishimoto et al. 2019).

In our study we propose a simple model of compact near-spherical particles, that can reproduce both the PLDR and LR values measured by sophisticated lidar systems, part of the EARLINET, that are capable of providing quality-assured retrievals for stratospheric smoke particles. We further examine whether this model could be used on an operational level to extend the AERONET retrieval scheme. The introduction of the manuscript has been updated in order to present how our research is differentiated by previous research (**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)."

6) The title (i found it funny) might be misinterpreted and considered inappropriate.

REPLY: Thank you for your comment.

We reply to specific comments in the attached manuscript below:

Page 1, line 15: *We added: "of axial ratio 0.7 to 1.5"*

Page 2, line 1: We rephrased to: "Smoke particles in the atmosphere can be identified with lidar measurements which provide valuable information on the optical properties of aerosol particles, such as the depolarization of the backscattered light in terms of the particle linear depolarization ratio (PLDR)."

Page 9, line 4: please see response to Comment 3.