The authors use an example of transported stratospheric smoke from the 2017 Canadian wildfire pyroCB to model the spectral dependence of depolarization and lidar ratio at 355, 532, and 1064 nm. Near-spherical, Chebyshev, and fractal shapes are used, but only the near-spherical shapes are able to match the results obtained from lidar measurements. The subject is quite relevant and the results could be very significant for scientific community. However, there are some issues with the manuscript which must be clarified before it is suitable for publication. Examples are provided below:

REPLY: Thank you very much for your helpful comments. Please find our point-by-point response below.

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

1. Firstly, the title implies that the near spherical shape may be the "new black" for smoke. However, the case study focuses on a stratospheric smoke case. It follows to ask if this is only the "new black" for smoke in the stratosphere only or should we assume this might apply to the troposphere also? From the example shown, my guess is no.

REPLY: Thank you for this question. To the best of our knowledge, up to now the majority of the cases reported for smoke particle linear depolarization ratio (PLDR) approximating 20% at 532nm, refer to smoke found in the stratosphere. The sole exception is the case study reported in Burton et al. (2015) for a smoke plume found at 8km height. All the cases were associated with PyroCb activity and all are indicative of high depolarization values in both troposphere and stratosphere, this is why we didn’t separate in the title. We make sure we mention this throughout the revised manuscript and we further included the following to make this more obvious to the reader (**page 7, line 24**):

“To the best of our knowledge, up to now the majority of observations for such smoke PLDR values, refer to smoke particles found in the stratosphere (i.e. Ohneiser et al., 2020). The sole exception is the case study reported by Burton et al. (2015) (see also Table 2). “

2. Page 3, Line 10: Is the spectral dependence really non-typical? The authors do a good job of convincing us that the high depolarization values are non-typical for smoke, but spectral dependence seems the opposite.

In fact, decreasing depolarization with increasing wavelength is closer to typical from the references cited by the authors.

REPLY: We agree with the reviewer that this statement should be reworded, primarily because there is still limited amount of information on such case studies for smoke. The only cases that up to now have reported observations at three lidar wavelengths are Haarig et al. (2018), Hu et al. (2019) and Burton et al. (2015). The first two refer to the same case of British Columbia fires of 2017, while the last one refers to the Pacific Northwest fires of 2014. There is a notable difference in PLDR values at 532nm reported from Haarig and Hu (~18%), compared to those reported from Burton (~9%), but still this may not be sufficient information to characterize the spectral dependence as non-typical.

The following has been re-worded in the manuscript in order to highlight this comment (**page 3, line 10**): "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."

3. Page 3, Line 12: Something is missing from this sentence or perhaps the wording is intended to be: The starting point and main assumption of our investigation is that the particle near-spherical-shape can be highly depolarizing as shown in the work of Mishchenko and Hovenier (1995); Mishchenko et al. (2016); Bi et al. (2018) and Ishimoto et al. (2019).

REPLY: Thank you, we have rephrased the following in the revised manuscript (**page 3, line 12**):

"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). "

4. Figure 2: The images seem to be incorrectly placed, given the aspect ratios. Oblate sphere should be flattened and prolate stretched.

REPLY: Thank you for noticing this, the figure has been updated.

5. Page 6, Line 20 - 26 and Figure 6: The latitude limits in the text do not match those in the Figure and the red dashed lines do not match the section highlighted in Figure 4. But even more importantly, the corresponding browse images indicate a complex mixture of aerosol and ice from cirrus clouds which would explain the high depolarization ratios in these cases.

https://www.calipso.larc.nasa.gov/products/lidar/browse_images/show_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

https://wwwcalipso.larc.nasa.gov/products/lidar/browse_images/show_detail.php?s=production&v=V4-10&browse_date=2017-08-15&orbit_time=17-55-33&page=4&granule_name=CAL_LID_L1-Standard-V4-10.2017-08-15T17-55-33ZD.hdf

REPLY: Figure 6 has been corrected. Regarding the next part of the question, 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). 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. Indeed, at such low temperatures homogenous ice formation can occur (Wallace and Hobbs, 2006; Fig. 6.29). However**,** 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 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 Angstrom exponent values close to zero are expected, although, as indicated by the large standard deviation, CALIPSO data are highly noisy at these altitudes

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.

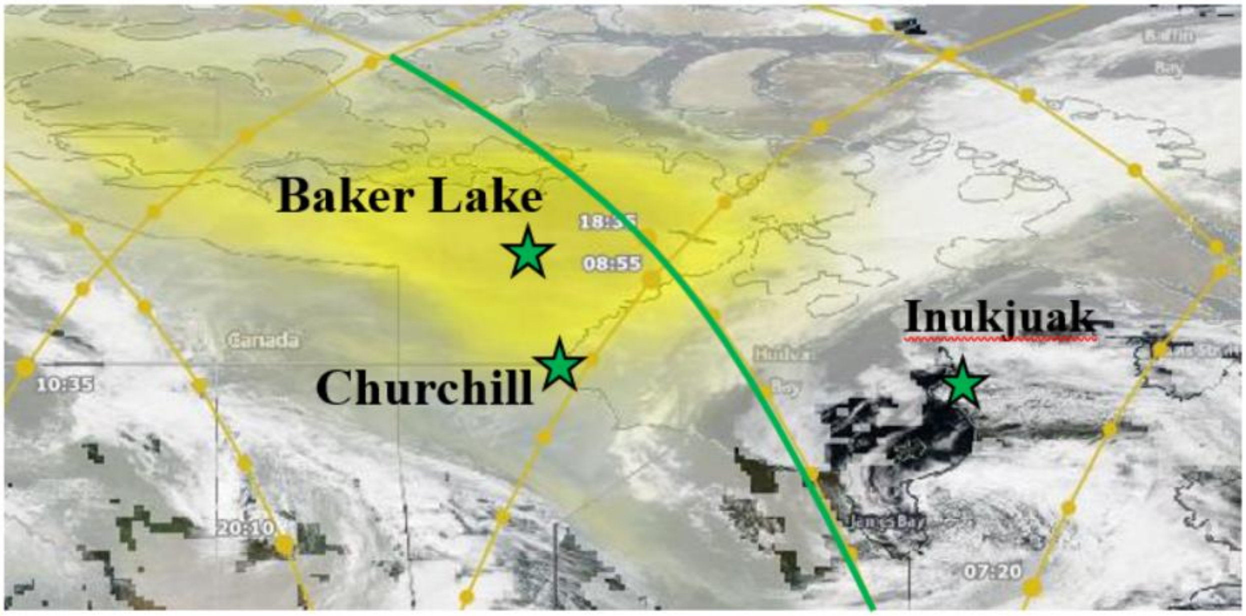

Figure 1. Corrected surface reflectance from MODIS on 15 August 2017, over-plotted with the PyroCb aerosol index product from Suomi NPP/OMPS (in yellow). Green stars indicate the position of the radiosonde stations used, while the green line marks the CALIPSO overflight during 18:22 – 18:35 UTC. The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

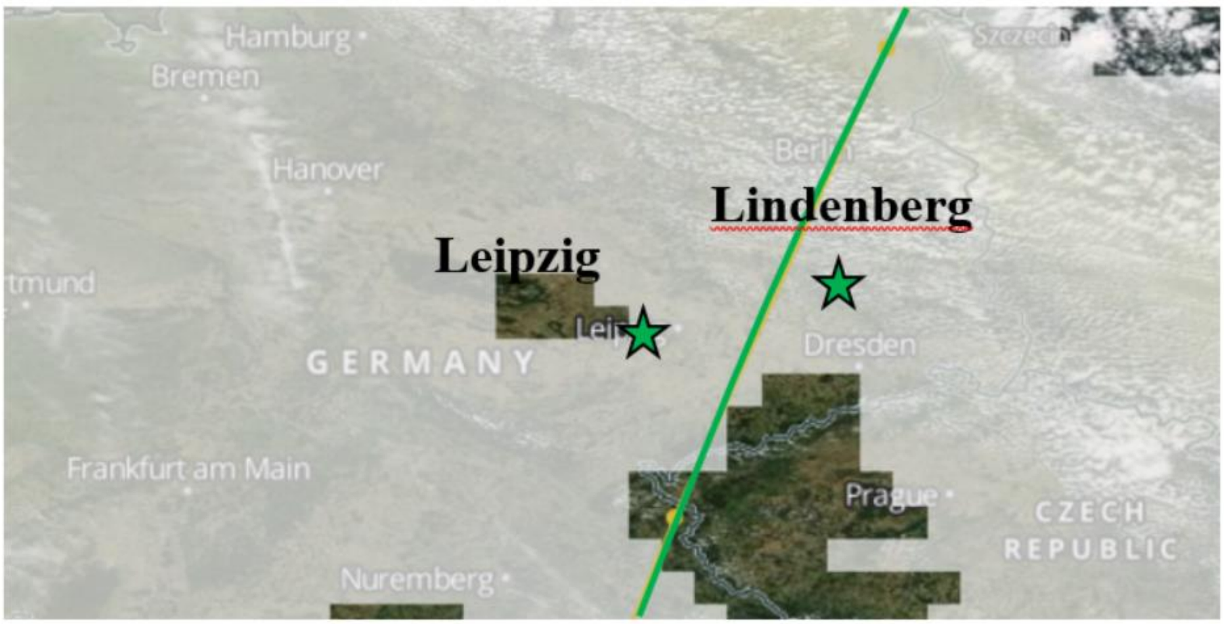

Figure 2. Same as Fig. 1 but for 23 August 2017. The CALIPSO overflight is approximately 90 km from Leipzig station, at 01:23 – 01:48 UTC, 1 hour after the end of the ground based lidar measurements.

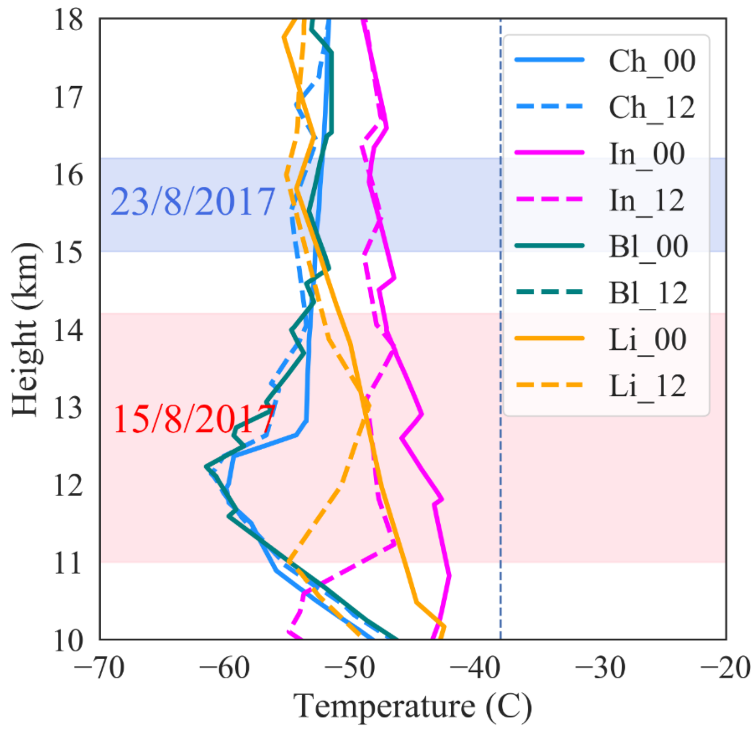

Figure 3. Radiosonde temperature (T) profiles from Churchill (Ch), Inukjuak (In), Baker Lake (Bl) and Lindenberg stations. Solid lines denote the measurements at 00:00 UTC, while dashed lines the measurements at 12:00 UTC. For the first three stations (Ch, In, Bl) measurements from 15 August 2017 are 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).

To highlight this comment, we included the following paragraph in the revised 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). 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."

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.

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

6. The images from Haarig et al 2018 are more convincing for the case the authors are making. Perhaps another CALIPSO image should be used if the authors would like to show satellite measurements.

REPLY: We improved the images to better demonstrate our arguments.

7. Page 8, Line 12-13: The authors state: What is interesting here is that the retrieved sizes for near-spherical smoke particles are absent in the AERONET climatology product. However, the difference in using a mono-modal versus bi-modal distribution is something that should be explored or at least discussed more in the text. Are we seeing the result of an "effective" radius in a mon-modal distribution that presents a possible solution for the high depolarization and spectral dependence, when there could also be another solution obtained from a mixture in a bi-modal distribution?

REPLY: Thank you for this comment. We have provided a reply to a similar comment made by anonymous Reviewer 1: Regarding the kind of the distribution used, as discussed in Hansen and Travis (1974), the sensitivity of the optical properties of the particles to the type of the size distribution is limited. Regarding the second (coarse) mode, similar simulations presented by Bi et al., (2018; Fig 2), suggest that for near-spherical particles the measured spectral dependence of PLDR could not be reproduced by coarse mode particles. Thus, an optically significant coarse mode would have to be investigated with a different shape model. Maybe the reviewer is further interested in the answer provided for Comment 11 made by anonymous Reviewer 3.

In the revised version of the manuscript, we have also included the following paragraph (**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)."

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

8. Sections 4.2 and 4.3: Is this all that can be said for these solutions... we tried these shapes and they didn't work? It would seem that more could be gleaned from these failed attempts. For instance, these shapes show the correct trend in spectral dependence even though the absolute values/relative differences do not fit the measurements.

REPLY: We would like to thank the reviewer for this comment. Updated discussion on the results for Chebyshev particles has been included in the manuscript ***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."

We decided to remove fractal aggregates from the present study, since as pointed out by Anonymous Reviewer 3 the range of the parameters used in our study to model fractal aggregates was limited.

9. Page 9, Line 18: Can the authors offer an explanation for the low angstrom exponents other than coarse particles?

REPLY: We have provided an answer to a similar comment made by anonymous Reviewer 3. We summarize also here: According to Eck et al. (1999), the strong curvature between the extinction related Angstrom exponent (EAE) at 355/532nm (-0.3 ± 0.4) and the corresponding values at 532/1064nm (0.85 ± 0.3) can be attributed to the pronounced accumulation mode of the size distribution, which is in good agreement with the retrieved size distribution for near-spherical particles of $reff$ = 0.38μm. Another possible reason could be a spectrally-dependent absorption, although this 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 added the following paragraph to the manuscript (**page 8, line 22**):

"We note here that all the retrievals indicate fine particles, with mean geometric radius that does not exceed the value of 0.35μm. The simulations presented by Bi et al., (2018; Fig. 2) suggest that for the near-spherical particles the measured spectral dependence of PLDR (steeply decreasing from the UV to the Near-IR) could not be reproduced by coarse 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.