Reply to Referee #1

We appreciate the reviewer's encouraging comments and valuable suggestions. Moreover, the bibliographical list provided by the reviewer was particularly relevant for interpreting our results in a more comprehensive context. Our responses to the reviewer's comments are addressed below (in blue):

Details:
P4, L10-12: Please be a bit more specific, more quantitative, if the extinction coefficient is below 25 Mm-1 or the AOT is below 0.05 CALIOP will no detect this aerosol? Please, provide some kind of threshold numbers.

The reviewer's request is addressed in the following sentence:

"Typically, the detection threshold is range-dependent, and varies as a function of molecular density, solar background and other instrument noise, and signal averaging (Vaughan et al., 2009). In terms of AOD, global analysis of CALIOP V3 daytime data by Toth et al. (2018) show that the "aerosol-free" columns reported by the CALIOP algorithm correspond to a mean MODIS AOD of 0.03-0.05. A similar analysis by Kim et al. (2017) shows that, as expected, CALIPSO extinction and AOD retrieval capabilities are substantially better at night than during the day. These authors estimate a maximum mean undetected extinction coefficient of ~0.006 km^{-1} during daytime versus ~0.003 km^{-1} at night (see their Fig. 5c)."

P6, L6-8: Is the two-layer method not similar to the approach of Ansmann, Appl. Opt, 45, 2006 (Ground-truth aerosol lidar observations: can the Klett solutions obtained from ground and space be equal for the same aerosol case?). Should probably be mentioned.

Ansmann (2006) is now properly acknowledged in our revised manuscript.

P6, L12-16: Regarding true marine lidar ratios, you may check and give reference to the papers of Gross et al., Tellus, 2011 (Cabo Verde, SAMUM2), ACP 2015 (Barbados, SALTRACE), Haarig et al., ACP, 2017 (SALTRACE, Barbados, wet and dry sea salt lidar ratios).

We appreciate the detailed list of references. The following lidar-related articles are included in the revised manuscript (more details in page 4):

Ansmann, A.: Ground-truth aerosol lidar observations: can the Klett solutions obtained from ground and space be equal for the same aerosol case?, *Appl. Optics*, 45, 3367–3371, 2006.

Groß, S., Freudenthaler, V., Schepanski, K., Toledano, C., Schäfler, A., Ansmann, A., and Weinzierl, B.: Optical properties of long-range transported Saharan dust over Barbados as measured by dual-wavelength depolarization Raman lidar measurements, *Atmos. Chem. Phys.*, 15, 11067-11080, https://doi.org/10.5194/acp-15-11067-2015, 2015.

Mona, L., Amodeo, A., Pandolfi, M., and Pappalardo, G.: Saharan dust intrusions in the Mediterranean area: Three years of Raman lidar measurements. *J. Geophys. Res.*, 111, D16203, doi:10.1029/2005JD006569, 2006.
Noh, Y. M., Kima, Y. J., and Mu¨ller, D.: Seasonal characteristics of lidar ratios measured with a Raman lidar at Gwangju, Korea in spring and autumn, *Atmos. Environ.*, 42, 2208–2224, doi:10.1016/j.atmosenv.2007.11.045, 2008.
Bohlmann, S., Baars, H., Radenz, M., Engelmann, R., and Macke, A.: Ship-borne aerosol profiling with lidar over the Atlantic Ocean: from pure marine conditions to complex dust–smoke mixtures, *Atmos. Chem. Phys.,* 18, 9661-9679, https://doi.org/10.5194/acp-18-9661-2018, 2018.
Tesche, M., Gross, S., Ansmann, A., Mu¨ller, D., Althausen, D., Freudenthaler, V., and Esselborn, M.: Profiling of Saha- ran dust and biomass-burning smoke with multiwavelength polarization Raman lidar at Cape Verde, *Tellus*, B63, 649–676, doi:10.1111/j.1600-0889.2011.00548.x, 2011.

P7, L3: Great design of the campaign is visible in Fig.1! Well planned!

The campaign was primarily intended for acquiring HSRL measurement to validate CALIOP data. We were fortunate of having these measurements for our study.

P7, L16: What is the truth? HSRL? How do you know, what the true AOD is? P7, L27-29: column lidar ratio. . .. also given in Ansmann, Appl, Opt, 45, 2006.

We added the following to explain that the HSRL is the truth in the context of our study:

" HSRL 532 nm AOD and aerosol extinction coefficient have been regularly validated against other airborne instruments, with biases less than 6% and 3%, respectively (Rogers et al. 2009), and generally to within 0.03 in comparison with AERONET AOD (Sawamura et al., 2017). The AOD product from the HSRL instrument makes use of the molecular channel which is a direct observation of atmospheric attenuation between the aircraft and the surface when compared against the GEOS-5 molecular density profile (Rogers et al. 2009). Since this method requires no assumptions about the lidar ratio or assumptions that the lidar ratio is constant, it provides a useful truth measurement in the context of this study."

P8, L1-8: Please explain better, first: 1L approach: all details, ... afterwards 2L approach, i.e., explain 1L and 2L separately, one after another. At the moment, too many details are given at the same time. . . , it took me some time to 'disentangle' the information properly.

We updated this section to read:

"The conventional method to solve eq. (3) follows Fernald (1984) and consists of iteratively solving for β_a, assuming a functional form of the lidar ratio LR(z). The LR selection is physically constrained by comparing the retrieved aerosol optical depth. ($AOD_{ret} = \int_0^z \sigma_a(z')dz'$) with SODA AOD ($AOD_{SODA}$), and LR is iteratively adjusted until the retrieved AOD matches the SODA AOD to within 0.001 or less (i.e., when $|AOD_{ret} - AOD_{SODA}| \leq 0.001$). While the shape of LR with height can be selected in different ways (e.g. Ansmann, 2006), here we opt for two assumptions, which in turn yield two independent sets of aerosol extinction and lidar ratio retrievals:

A. 1-layer lidar ratio (1LR): The simplest assumption is to consider one constant lidar ratio with height. This method is expected to perform well for atmospheric profiles characterized by only one aerosol type.
B. 2-layer lidar ratio (2LR): We also consider an additional scenario, which consists of treating the atmospheric column as two layers, that is, the marine atmospheric boundary layer (MBL) and a second aerosol layer of as-yet-undetermined composition. This method is intended to better capture specific events with two predominant aerosol types, particularly smoke over marine aerosols and dust over marine aerosols, which are particularly frequent over the Atlantic Ocean. The LR for the MBL is assumed constant at 25 sr, as suggested by HSRL measurements over the ocean (Burton et al., 2012; 2013). This lidar ratio is slightly higher than the one compiled by Kim et al. (2018, 23 sr). In contrast, 532 nm Raman lidar observations at Barbados encompass lidar ratios between 21 and 35 sr, with magnitudes primarily controlled by free tropospheric intrusion of dust (Groß et al., 2015). Similar range of MBL lidar ratio were observed in the eastern Atlantic by Bohlmann et al. (2018), with values modulated by the presence of dust-smoke aerosols. Without a-priori knowledge of MBL lidar ratio, the value prescribed here (25 sr) is within the range reported in previous studies over the ocean. $\sigma_a(z)$ and the upper layer LR are iteratively calculated using the Fernald method with the constraint provided by AOD_{SODA}, and LR =25 sr in MBL. MBL height is computed by applying the bulk Richardson number method (McGraw-Spangler and Molod, 2014). "

P8, L14-19: Overestimation. . ., is that caused by the use of the Klett forward integration method? Could be mentioned. . .

The aforementioned sentence is referring to the overestimation of CALIOP V4 aerosol extinction coefficient. The bias is primarily explained by the constant lidar ratio utilized by V4, which is higher than that observed during the field campaign.

P10, L18-21: Here, more comparisons with literature lidar ratio values would be good: Franke et al., GRL 2001, JGR 2003 (Indian Ocean, INDOEX, Maldives, Indian pollu- tion aerosol, 2L structures. . .), Gross et al., , Tesche et al., both in Tellus 2011(eastern Atlantic, SAMUM2, Cabo Verde, summer and also winter, Tellus, 2011), Gross et al., 2015, Haarig et al. 2017, both in ACP (Caribbean, SALTRACE, Barbados, dust lidar ratios), Bohlmann et al, ACP, 2018, Polarstern cruises from the North to the South At- lantic, with Raman lidar aboard, also Kanitz et al., JGR 2013, issue 6. . And please check also . . . all the papers from Japanese, Chinese, and Korean groups. A good starting point may be the following recent paper in ACP: Vertical

variation of optical properties of mixed Asian dust/pollution plumes according to pathway of air mass trans- port over East Asia S.-K. Shin, D. Müller, C. Lee, K. H. Lee, D. Shin, Y. J. Kim, and Y. M. Noh Atmos. Chem. Phys., 15, 6707-6720, https://doi.org/10.5194/acp-15-6707-2015, 2015.
Please check the reference list in this paper for more lidar ratio papers.
P12, L10: limited number of observations of lidar ratios. . .. As mentioned please check the available literature. . ., and then 'update' this statement a little bit. Again, discuss the literature values (P13, L6, Kanitz, Bohlmann), L11-12, Franke et al., L15-16, Franke et al., L20-21, Haarig et al., Bohlmann et al.

We have included some of the suggested references in the manuscript. The inclusion of new references is primarily reflected in the revised discussion:

"As different aerosol types can be, to some extent, characterized by their lidar ratio, the reliability of CALIOP-SODA LR retrievals is qualitatively assessed by analyzing the consistency between the CALIOP-SODA LR spatial pattern and the regional occurrence of aerosol types as well as lidar measurements from several field campaigns over the ocean. Burton et al. (2012), using HSRL measurements over North America and the adjacent Atlantic Ocean, provide the following lidar ratios for a number of aerosol types: the highest LR (45-80 sr) are typically attributed to smoke and urban aerosols, LR of 25-50 sr and 40 sr are associated with dust and polluted maritime aerosols (respectively), and maritime aerosols are characterized by lidar ratios of less than 30 sr. For simplicity, we will primarily interpret daytime LR_{1L} in Figures 9a and c for the following regions of interest:

6.1 Southeast Atlantic: The SODA LR peak in the southeast Atlantic is explained by the well-documented biomass burning season over southern Africa, with massive fires events from May to September during the dry season (Roberts et al., 2009), and smoke being transported offshore by the prevailing winds during July to October (Adebiyi et al, 2015). HSRL airborne measurements collected in September 2016 (Burton et al., 2018) show 532 nm LR in the range 58-76 sr in the free troposphere, with CALIOP-SODA yielding values in the lower bound of the HSRL measurements (55-60 sr). In addition, shipborne Raman lidar observations south of the region dominated by biomass burning aerosols (30˚S, near the South African coast) reveal a transition from a lower troposphere dominated by smoke to one mainly composed of maritime aerosols (lidar ratios less than 25 sr, Bohlmann et al., 2018). This southward reduction in LR is reproduced by CALIOP-SODA.

6.2. Mediterranean Sea: The high spring-summer SODA LR over the Mediterranean Sea (~ 50 sr) is also expected given the southward pollution transport from Europe which is maximized in summer in the boundary layer (Duncan and Bey, 2004). Moreover, lidar observations show a maximum dust AOD over the Mediterranean Sea (southern Italy) in summer (Mona et al., 2006), in connection with a Saharan dust layer in the free troposphere. The higher presence of pollution and dust in spring would explain the high CALIOP-SODA LR in spring-summer (MAMJJA).

6.3. Bay of Bengal and western Pacific Ocean: A major LR maximum in autumn-winter (SONDJF) is observed south of India, over the Bay of Bengal and part of the Arabian Sea. This pattern is concomitant with the pervasive presence of pollution and biomass burning during the winter and pre-monsoon season (October to April, Krishnamurti et al., 2009). In contrast, during the monsoon season (June-September), dust aerosols become the dominant species over the Bay of Bengal (Das et al., 2013), which is manifested in the reduction in SODA LR in spring-summer (MAMJJA). Further east, off the coast of eastern China and Korea, a semi-annual contrast is

retrieved by SODA, with maximum LR>55 sr for SONDJF. Changes between autumn and spring were also observed over the Korean peninsula in the lidar ratios retrieved with a Raman lidar (Noh et al., 2008), with layer-mean of 56 sr and 63 sr for spring and autumn, respectively, and larger differences in the free troposphere. These changes are thought to be primarily explained by seasonal changes in the composition of dust and smoke.

6.4. Eastern Pacific and Southern Ocean: Regions with intermediate CALIOP-SODA LR (35 sr< LR< 50 sr) are located over broad regions of the eastern Pacific and the east coast of North America. These regions are likely influenced by a combination of maritime aerosols and pollution from the continents. It is nevertheless surprising the high SODA lidar ratios retrieved over rather pristine regions, especially over the Southern Ocean, where maritime aerosols are expected to be the dominant aerosol type. A plausible factor that may help reconcile high LR for maritime aerosols is a lidar ratio increase with relative humidity (Ackerman 1998). Relative humidity could also explain the presence of LR >30 sr over stratocumulus cloud regimes, where high relative humidity is confined in the boundary layer.

6.5. Central Pacific and northern Atlantic: The regions with the lowest LR are located over the tropical Pacific Ocean, where AOD is the lowest (Figure 11). An unanticipated result is the absence of a zonal band across the Atlantic that could be attributed to the westward transport of Saharan dust across the Atlantic Ocean. Unfortunately, due to the lack of in-situ observations along the Saharan dust pathways, the assessment of SODA LR over this region is challenging. Raman lidar data over the eastern Atlantic (Cape Verde), off the coast of western Africa, in spring show dust and smoke in the free troposphere and boundary layer with a mean LR of 54 sr (Tesche et al., 2011), and a dust layer thickness of about 4 km. Over the same region, SODA LR is 40 sr, which increases up to 45-50 sr when LR is estimated using the 2L assumption. Ground-based lidar observations over the western Atlantic (Barbados, 13.14° N, 59.62° W) in summer reveal the presence of maritime aerosols and dust, with lidar ratios of less than 40 sr in the boundary layer, and pure dust aerosols generally confined to the free troposphere (Groß et al., 2015). This suggests that the relatively low CALIOP-SODA LR over the Altantic basin may be explained by the contribution of maritime aerosols in the boundary layer. A more quantitative assessment, which includes the analysis of specific dust events, is left for future work. Lastly, the interpretation the 1064 nm CALIOP-SODA is not attempted here due to the lack of independent measurements and calibration uncertainties associated with the use of CALIPSO V3 for deriving SODA AOD. A future release of SODA based on CALIPSO V4 will benefit from the improved calibration of V4, which is estimated to be within 3% (Vaughan et al., 2018)."

P12, L25 to P13, L30. . . and one has always to be careful with column lidar ratios, when marine particles are involved (so in the case of the SODA approach). The lidar ratio of sea salt is partly below 20sr, So these particles are rather efficient in backscattering of laser photons. As a consequence, their weight in the backscatter-weighted column integration. . . controls or can dominate the result. . .

The reviewer raises an interesting point. This is the primary motivation of why we also retrieve CALIOP-SODA extinction using the the 2-layer method (2L), in which the lidar ratio is assumed constant at 25 sr in the boundary layer, and the one of the upper layer is estimated using the Fernald-Klett method.

All in all: An excellent paper!

We appreciate the reviewer's kind words.