Response to Anonymous Reviewer 1

The lidar group in Leipzig began measuring the extinction coefficient at 1064 nm in 2016, and this manuscript presents a comprehensive summary of observations. Until recently, information on the lidar ratio at 1064 nm and the Ångström exponent at 532/1064 nm was unavailable, and this study addresses that gap. For different aerosol types, the manuscript provides valuable new insights into the dependence of key parameters—backscattering and extinction coefficients, lidar ratio, and growth factors—on relative humidity.

The measurements were conducted at three wavelengths, allowing for an analysis of the spectral dependence of these parameters. The authors are well-recognized experts in the field of lidar measurements. The manuscript is well-written, includes a thorough review of existing literature, and is, in principle, suitable for publication as is.

The only minor comment concerns Fig. 3. While visually appealing, its necessity in the manuscript is unclear.

Dear Anonymous Reviewer,

We thank you for your time and effort spent to check our manuscript. And we are delighted that it meets your expectations.

Concerning Figure 3, I completely agree with you, that the photograph of the 5 lidar beams at our institute is not necessary to understand the content of the study. It is more a kind of ornament or decoration, it is an additional illustration as it was done in old books centuries ago. And as such I would like to keep it in the manuscript.

We went over the text and improved some formulations (see track changes) and found a minor bug in the fitting of the hygroscopic growth parameter of the lidar ratio (now 0.45 instead of 0.49, the uncertainty changed only below the given significant digits). The optical profiles used for the two presented case studies were made available via zenodo.

Kind regards,
Moritz Haarig in the name of all co-authors

Response to Anonymous Reviewer 2

This straightforward manuscript summarizes Raman lidar ratio measurements made by researchers at the Leibniz Institute for Tropospheric Research. These measurements are made at three wavelengths: 355 nm, 532 nm, and 1064 nm. Of these, the 1064 nm measurements are by far the most interesting and significant. Intrinsic aerosol optical properties at 355 nm and 532 nm, including lidar ratios, are reasonably well characterized in the existing literature (e.g., Floutsi et al., 2023). However, to my knowledge, this paper is the first to report *measured* 1064 nm lidar ratios for a wide variety of aerosol types. I expect this information to be especially valuable for space-based lidars – e.g., LITE, GLAS, CALIPSO, CATS, ACDL, and (eventually) CALIGOLA/Luce – all of which deploy an elastic backscatter channel at 1064 nm. The paper is well-referenced and clearly written and its subject matter is appropriate for ACP. I recommend publication. Though prior to publication, I ask that the authors consider the points raised in the "minor comments" below.

Dear Anonymous Reviewer,

We thank you for your time and effort spent to check our manuscript. Your comments are very helpful in improving the study. Especially, the discussion about the smoke lidar ratios and what we know and what not, profited from your input.
The manuscript was improved according to your suggestions. Additionally, we went over the text and improved some formulations (see track changes) and found a minor bug in the fitting of the hygroscopic growth parameter of the lidar ratio (now 0.45 instead of 0.49, the uncertainty changed only below the given significant digits). The optical profiles used for the two presented case studies were made available via zenodo.

Kind regards,

Moritz Haarig in the name of all co-authors

Minor Comments

This first remark is more a quibble than a useful comment. On lines 32–34 the authors say, "All backscatter lidars [...] need to assume an aerosol-type dependent lidar ratio to derive the extinction coefficient". While this is generally true, it is not (as implied) always true; e.g., see the papers by Dawson et al., 2015, Liu et al., 2015, and Li et al., 2022.

You're right. We removed the "all" and simply state "Backscatter lidars …". Later in the introduction, we introduce the SODA technique, which unfortunately is only applicable to spaceborne lidars. However, the general idea by constraining the extinction coefficient by independent measurements of the AOD is applicable to ground-based lidars as well. Furthermore, I added Li et al., 2022 in the context of the lidar ratios at 532 nm presented in Kim et al., 2018 (beginning of Sect. 4).

We added a paragraph in the introduction (L44-55 in track changes document): "Furthermore, additional instruments can be used to constrain the extinction coefficient and to estimate the lidar ratio. Ground-based backscatter lidar observations can make use of the columnar extinction coefficient provided by a sun photometer as aerosol optical depth (AOD) (e.g., Mattis et al., 2004; Tesche et al., 2009). For CALIPSO, a solution was found by using opaque water clouds for AOD estimation (Hu et al., 2007) or the ocean surface reflectance

and additional cloud radar returns, the so-called Synergized Optical Depth of Aerosols (SODA) technique to derive independently the AOD (e.g., Josset et al., 2011; Painemal et al., 2019). The retrieval algorithms from passive sensors and the columnar AOD to constrain the extinction coefficient are helpful, but here we will present direct measurements."

In Figure 1, consider adding a (red?) vertical line to indicate exactly where 1064.14 nm is in relation to the Raman shifted lines.

It is a good idea. We added the line at the emission wavelength.

On lines 327–329 the authors correctly note that, "An early CALIPSO study by Liu et al. (2008) revealed an increase of 27 – 34 % in the lidar ratio from 532 nm to 1064 nm for Saharan dust over the Atlantic Ocean". Based on the 2008 publication data, this paper almost certainly used version 2 of the CALIPSO data products. (Version 3 CALIPSO data was not released until November 2009.) Between version 2 and version 4, CALIPSO made a large number of improvements to their 1064 nm calibration algorithm, resulting in substantial changes to the 1064 nm calibration coefficients over most of the globe (Vaughan et al., 2019). In particular, between versions 3 and 4 the changes in the Saharan dust belt are on the order of 20%, with V4 being lower than V3. Because calibration changes of this magnitude will introduce significant changes in the lidar ratios retrieved at 1064 nm, I do not think the Liu et al., 2008 paper offers reliable evidence for a spectral difference in dust lidar ratios.

Thank you for this insight comment. I see that an explicit number like the 27-34% increase can not be supported from the CALIPSO v2 data. Therefore, we decided to change it to a more general statement and explicitly mention that the data version of CALIPSO.

"An early CALIPSO study (data version 2) by Liu et al. (2008) pointed to an increase in the lidar ratio from 532 nm to 1064 nm for Saharan dust over the Atlantic Ocean."

I very much appreciate the list of "limitations" given by the authors in Appendix 2. Their points should be required reading for all practitioners, as retrievals at 1064 nm are much, much trickier than those at shorter wavelengths.

Thank you for your support. In my first version, the limitations were listed in Section 2. Then, I decided for a publication in ACP and moved the more technical part of corrections and limitations to the appendix. Nevertheless, I consider it very important to speak about the limitations of the technique.

My final comment has to do with the authors' recommendation (see lines 474–477) that the CALIPSO project "update the lidar ratio at 1064 nm for these aerosol types [i.e., *"elevated smoke and stratospheric smoke"*] for a CALIPSO v5 algorithm". Whether this is a sound idea or not is, I believe, open to some debate. If so motivated, the authors could make a better case for adopting their recommendation by testing these higher 1064 nm lidar ratios using CALIOP measurements to determine what value(s) might be most appropriate. I illustrate this approach with the example shown in Figure 1, taken from Rajapakshe et al., 2017. The lefthand side replicates panels (c) and (d) from Figure 1 in the Rajapakshe paper, which show CALIOP attenuated backscatter coefficients measured at 532 nm (upper panel) and 1064 nm (lower panel) in a biomass burning plume lofted over an opaque stratus off the west coast of Africa on 8 August 2016. The righthand side shows vertical profiles of the attenuated backscatter coefficients averaged over a 20 km (60 single shot profiles) along track distance centered at 01:38 UTC, as shown by the white vertical line in the Rajapakshe image. Profile

data are smoothed vertically using a 150 m (5 range bin) sliding average. Extinction coefficients for the smoke layer are retrieved between the base and top altitudes indicated by yellow circles in the righthand image in Figure 1.
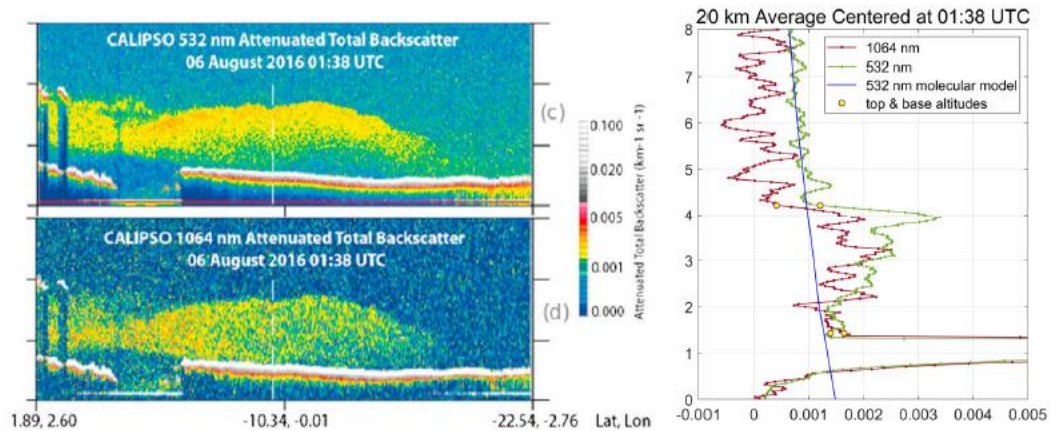
Figure 1: The lefthand side replicates panels (c) and (d) from Figure 1 in Rajapakshe et al., 2017, showing CALIOP attenuated backscatter measurements at 532 nm (upper panel) and 1064 nm (lower panel) of a biomass burning plume lofted over an opaque stratus off the west coast of Africa on 8 August 2016. The righthand side shows vertical profiles of the attenuated backscatter coefficients averaged over a 20 km (60 single shot profiles) distance centered at 01:38 UTC, as shown by the white vertical line in the Rajapakshe image. Top and base altitudes of the smoke plume (identified manually) are indicated using yellow circles. Data are smoothed vertically using a 150 m (5 range bin) sliding average.

Scenes like this are ubiquitous in the CALIPSO measurements during the African biomass burning season. The applicability of the author's recommendation could be tested using the following procedure: (a) estimate 532 nm smoke lidar ratios using measurements of above cloud optical depths provided in the CALIOP level 2 products (e.g., see Hu et al., 2007), and then (b) retrieve backscatter and extinction profiles at 1064 nm lidar ratio using a range of different lidar ratios. The most reasonable choice for a revised 1064 nm lidar ratio would presumably be the one that returns values that compare best with the extinction and backscatter Ångström exponents previously reported for smoke.

Table 2 shows the results of this experiment using the two averaged profiles shown in Figure 1, with 1064 nm lidar ratios varying between 20 sr and 85 sr. For the 20 km average in the Rajapakshe scene, CALIOP retrieves a mean above cloud optical depth of ~0.629, suggesting an approximate 532 nm lidar ratio of 90 sr rather than the CALIOP default of 70 sr. In this simple test, using a 1064 nm lidar ratio of 82 sr (i.e., as in Table 4) yields an EAE of 0.04, which is very different from the authors' retrieved value of 0.6 (Table 6). Instead, a 1064 nm lidar ratio just under 65 sr should give the closest match. Alternatively, based on the very sparse information available in the existing literature, one might expect the 1064–to–532 EAE to be in the neighborhood of 1.4 to 1.8, as in Chand et al., 2008 and Wu et al., 2012. For this range of values, 1064 nm lidar ratios between 35 sr to 45 sr provide the closest matches. Nowhere in this table does the BAE get close to the authors' retrieved value of 0.8. The largest value (~0.59) is found for a lidar ratio of 20 sr. When the 1064 nm lidar ratio is fixed at 82 sr, the resulting BAE is an order of magnitude smaller than the authors' value and has the opposite sign (i.e., -0.09). Also contrary to the authors' mean value, the SAE in Table 1 never drops below zero. I can only speculate that the reasons for these disparities lie in differences in the optical properties for the relatively fresh biomass burning aerosol CALIOP measured off the coast of Africa versus those for aged smoke transported to the interior of Europe. So, while it may well be that a higher 1064 nm smoke lidar ratio is warranted for the

CALIOP retrievals, it is not at all clear what value would be consistent with the authors' measurements.

Table 1: backscatter and extinction Ångström exponents computed for 1064 nm lidar ratios varying between 20 sr and 85 sr and a 532 nm lidar ratio of 90 sr.

λ	S	τ	EAE	BAE	SAE
532 nm	90 sr	0.6310	N/A	N/A	N/A
1064 nm	20 sr	0.0933	2.7578	0.5894	2.1684
1064 nm	25 sr	0.1198	2.3976	0.5513	1.8463
1064 nm	30 sr	0.1478	2.0945	0.5115	1.5831
1064 nm	35 sr	0.1775	1.8301	0.4697	1.3604
1064 nm	40 sr	0.2091	1.5933	0.4257	1.1675
1064 nm	45 sr	0.2430	1.3767	0.3794	0.9973
1064 nm	50 sr	0.2794	1.1753	0.3303	0.8450
1064 nm	55 sr	0.3188	0.9851	0.2781	0.7070
1064 nm	60 sr	0.3616	0.8033	0.2223	0.5811
1064 nm	65 sr	0.4085	0.6273	0.1623	0.4650
1064 nm	70 sr	0.4604	0.4548	0.0973	0.3575
1064 nm	75 sr	0.5185	0.2835	0.0263	0.2571
1064 nm	80 sr	0.5843	0.1111	-0.0520	0.1630
1064 nm	82 sr	0.6133	0.0412	-0.0858	0.1269
1064 nm	85 sr	0.6603	-0.0653	-0.1396	0.0743

I note too that should the authors conduct tests like this to buttress their recommendation for increasing CALIOP's 1064 nm lidar ratio, the results will, as expected, depend very much on the accuracy of the 532 nm extinction retrievals. Table 2 shows the same set of calculations as Table 1, but using CALIOP's default 532 nm smoke lidar ratio of 70 sr. (Actually, since conducting these tests seems like a long and laborious task, I suspect the authors would be better served by simply omitting their recommendation from their final manuscript. But, included or not, I firmly recommend this paper be published.)

Table 2: backscatter and extinction Ångström exponents computed for 1064 nm lidar ratios varying between 20 sr and 85 sr and a 532 nm lidar ratio of 70 sr.

λ	S	T	EAE	BAE	SAE
532 nm	70 sr	0.3579	N/A	N/A	N/A
1064 nm	20 sr	0.0933	1.9395	0.1372	1.8023
1064 nm	25 sr	0.1198	1.5793	0.0991	1.4802
1064 nm	30 sr	0.1478	1.2763	0.0593	1.2169
1064 nm	35 sr	0.1775	1.0119	0.0175	0.9943
1064 nm	40 sr	0.2091	0.7750	-0.0264	0.8014
1064 nm	45 sr	0.2430	0.5584	-0.0727	0.6312
1064 nm	50 sr	0.2794	0.3570	-0.1218	0.4788
1064 nm	55 sr	0.3188	0.1669	-0.1740	0.3409
1064 nm	60 sr	0.3616	-0.0149	-0.2299	0.2149
1064 nm	65 sr	0.4085	-0.1910	-0.2899	0.0989
1064 nm	70 sr	0.4604	-0.3635	-0.3548	-0.0087
1064 nm	75 sr	0.5185	-0.5348	-0.4258	-0.1090
1064 nm	80 sr	0.5843	-0.7072	-0.5041	-0.2031
1064 nm	82 sr	0.6133	-0.7771	-0.5379	-0.2392
1064 nm	85 sr	0.6603	-0.8836	-0.5917	-0.2918

We sincerely thank the reviewer for taking up the discussion on the spectral behavior of the lidar ratio of smoke. Indeed, our observations are limited to aged, long-range transported smoke plumes from the midlatitudes (Canadian, southern Australia). And even there, we see strong differences in the values with higher values for the Australian smoke plumes. Tropical smoke plumes and fresh smoke plumes do show a different behavior already in the commonly measured wavelength range from 355 to 532 nm. I went back to my own literature overview which I presented in the Haarig et al., 2018 paper (Table 3). Looking at the African smoke in the troposphere Tesche et al., 2011 and Giannakaki et al., 2015, observed values in the range of 80 – 90 sr with slightly higher values at 355 nm compared to 532 nm. These values are much more in line with the CALIPSO observations from Rajapakshe et al., 2017 which you showed and which are originating from African biomass burning sources. In the case of African biomass burning smoke, I would not expect that the values at 1064 nm are 20 – 40 % higher as suggest by our observations. I suspect that the size distribution of the smoke particles is different according to burning conditions, transport path and time. The large BAE observed in our studies might not be the case for tropical biomass burning smoke.

The same holds for fresh biomass burning smoke as observed by Veselovskii et al., 2015 over the eastern US.

From the discussion with you, I conclude that the spectral behavior of the lidar ratio contains valuable information about the smoke plumes. However, we are still far away from a complete understanding of the reasons behind. Herein lies a potential which is not fully exploited yet. A solid data base only exists for the 355 – 532 nm wavelength range as the observations at 1064 nm are very limited.

And I agree that from our limited observations of aged, long-range transported Canadian smoke, we cannot draw global conclusions. We rather deliver another piece of information in the puzzle. Therefore, I removed the recommendation for CALIPSO v5 lidar ratios from the manuscript as CALIPSO has to consider globally occurring smoke plumes. However, I left the recommendation for stratospheric smoke as this is usually aged and long-range transported smoke from wildfires from the northern or southern mid-latitudes.

We should move forward in the characterization of smoke in the atmosphere. A promising approach is the use of fluorescence lidars (Veselovskii et al., 2022), which we recently started in Leipzig as well (Gast et al., 2025).

The text in Section 4 was changed (L419-425 in the track changes manuscript)
"It is difficult to draw firm conclusions for a future CALIPSO lidar ratio selection algorithm, because atmospheric smoke layers show a different spectral behavior of the lidar ratio in the wavelength range of 355 to 532 nm and it can be assumed that the spectral slope towards 1064 nm is affected as well. The presented observations of lidar ratios at 1064 nm cover only the category of long-range transported, aged smoke from extra-tropical source regions. However, the CALIPSO lidar ratio selection algorithm needs to use global lidar ratios per aerosol type to cover for example the smoke plumes originating from tropical Africa (e.g., Rajapakshe et al., 2017). On the other side, ground-based lidar stations might select the best matching lidar ratios according to the dominant smoke influence at their location."

And in the conclusion as well (L499-503)
Stratospheric smoke layers are in most cases transported over large distances from the wildfire source. Therefore, we would recommend to update the lidar ratio at 1064 nm for this aerosol type for a CALIPSO v5 algorithm. The tropospheric smoke layers exhibit a varying spectral behavior in the 355-to-532 nm wavelength range and therefore it difficult to predict the values at 1064 nm and to give recommendations to the CALIPSO lidar ratio selection algorithm, e.g., for elevated smoke.

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