



Discussion of the spectral slope of the lidar ratio between 355 nm and 1064 nm from multiwavelength Raman lidar observations

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Abstract. The extinction-to-backscatter ratio also known as lidar ratio is an important quantity in active remote sensing with lidar. It is a key parameter in aerosol typing and an essential quantity to derive the extinction coefficient from elastic backscatter lidars like the spaceborne Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) or ceilometer observations. The present study discusses the spectral dependence of the lidar ratio from the frequently measured lidar ratios at 355 and 532 nm to the rarely observed lidar ratio at 1064 nm. Therefore, a special focus is put on the rotational Raman lidar method applied to the emission wavelength of 1064 nm. We present a literature review on existing triple-wavelength lidar ratio observations and add new measurements for marine and continental aerosol. For cirrus clouds, marine and continental aerosol the spectral behavior is neutral; for sulfate aerosol, we see a decrease from 532 to 1064 nm. In the case of mineral dust, the lidar ratio slightly increases towards 1064 nm and in the case of aged smoke an increase with wavelength is found. Furthermore, we observe an increase of the lidar ratio with relative humidity for continental aerosol and report hygroscopic growth factors. The findings are compared to the assumptions made in the CALIPSO version 4 automated aerosol classification and lidar ratio selection algorithm (Kim et al., 2018). We could confirm these assumptions for marine and sulfate aerosol. However, we see slight differences for mineral dust and continental aerosol and pronounced differences for elevated smoke.

1 Introduction

Aerosol particles are a major constituent of the atmosphere. They influence the Earth's radiation budget directly by scattering and absorbing long- and shortwave radiation; and indirectly by influencing cloud formation and precipitation. They are highly variable in space and time. This behavior challenges the observations of aerosol particles. Ground-based in-situ measurements help to estimate aerosol's influence on human health, but do not provide a reasonable assessment on the aerosol impact on cloud formation and radiation. Therefore, vertically resolved observations are needed. Light detection and ranging (lidar) systems are proven to be most suitable to provide the required vertical profile of aerosol optical properties from the ground up to the stratosphere. In the past decades, multi-wavelength lidar systems became widely used (e.g., Althausen et al., 2000;



Burton et al., 2015; Engelmann et al., 2016; Haarig et al., 2017a). Usually, light pulses are emitted at 355, 532 and 1064 nm and the elastically backscattered light is detected (Mie lidar principle). In order to assess the mass concentration and optical depth of the aerosol load, measurements of the extinction coefficient are necessary (e.g., Mamouri and Ansmann, 2014). Additional detection of the elastically backscattered and Doppler-broadened light from air molecules (high-spectral resolution lidar, HSRL, Shipley et al., 1983) or Raman backscattered by nitrogen molecules (vibrational-rotational Raman transitions, Ansmann et al., 1992) enables the direct measurement of the extinction coefficient. Alternatively, the pure rotational Raman transitions can be used to derive the extinction coefficient (e.g., Achtert et al., 2013; Veselovskii et al., 2015; Haarig et al., 2016). The HSRL and Raman lidar methods enable the independent measurement of the backscatter and extinction coefficient and thus the calculation of the extinction-to-backscatter ratio (lidar ratio). This ratio depends on the aerosol type and therefore is a favorable and useful quantity in state-of-the-art aerosol typing schemes (e.g., Burton et al., 2012; Floutsi et al., 2024). All backscatter lidars (Mie lidars) including ceilometers (e.g., E-Profile) and Micro-pulse lidars (Campbell et al., 2002) need to assume an aerosol-type dependent lidar ratio to derive the extinction coefficient. At 355 and 532 nm, there is a large number of Raman and HSRL measurements available and lidar-ratio statistics are reported (e.g., by Müller et al., 2007; Floutsi et al., 2023). However, at 1064 nm almost all lidars are simple backscatter lidars, e.g., the lidars in PollyNET (Baars et al., 2016), the European Research Lidar Network (EARLINET) and Asian dust and aerosol lidar observation network (AD-Net, Nishizawa et al., 2017) or the spaceborne lidars such as the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP, Omar et al., 2009; Kim et al., 2018), the Cloud-Aerosol Transport System (CATS, Yorks et al., 2016) or the Aerosol and Carbon Detection Lidar (ACDL/DQ-1, Dai et al., 2024). Furthermore, a great number of ceilometers are operated at 1064 nm (e.g., Wiegner et al., 2019). Therefore, there is a high need to know the lidar ratio at this wavelength to derive extinction coefficients from these numerous backscatter lidar observations. Lidar ratio retrievals from the Aerosol Robotic Network (AERONET) provide global statistics for mineral dust (Shin et al., 2018) but the retrievals at the near infrared wavelengths are not validated by measurements yet. They are based on modelling optical properties of spheroids (Dubovik et al., 2006) which may fail in predicting the correct spectral slope at 180° scattering angle (e.g., Zubko et al., 2013; Wandinger et al., 2023).

First rotational Raman lidar measurements at 1064 nm were reported by Haarig et al. (2016) with a cirrus case study. Since then, measurements in smoke layers (Haarig et al., 2018), desert dust (Haarig et al., 2022; Gebauer et al., 2024) and sulfate-marine mixtures (Gebauer et al., 2024) were reported. Additionally, new lidar systems are designed to measure the extinction at 1064 nm (Wang et al., 2024a, b).

One aim of this study is to check the assumptions of the lidar ratio at 1064 nm made for Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) version 4 (v4) aerosol typing and lidar-ratio-selection algorithm presented in Kim et al. (2018). For this reason, we gathered rotational Raman observations for the CALIPSO aerosol types clean marine, dust, clean continental, elevated smoke and sulfate/other and discuss them to the respective values used by Kim et al. (2018). In the present study, we first shortly present the rotational Raman technique applied in the near infrared wavelength range (Sect. 2). Some corrections and limitations are discussed in the Appendix. Then, we show two case studies of marine and continental aerosol as an addition to the rare lidar ratio observations at 1064 nm already existing (Sect. 3). Furthermore, we investigate the influence of relative humidity (RH) on the lidar ratio of continental aerosol and derive hygroscopic growth factors. The main

part is the discussion of the spectral dependence of the lidar ratio of different aerosol types and the comparison to CALIPSO v4 values in Sect. 4 which is followed by a conclusion in Section 5.

60 2 Rotational Raman measurements at 1064 nm

Extinction coefficients and lidar ratios at 355 and 532 nm were derived with the vibrational-rotational Raman lidar technique. The signals of the inelastically backscattered light at 387 and 607 nm (vibrational-rotational Raman lines of nitrogen) were used. This technique is widely known and needs no further description at this point (see e.g., Ansmann et al., 1992).

Recently, the pure rotational Raman technique (Whiteman, 2003a, b) has been applied to derive the extinction coefficient, e.g.,
 65 at 532 nm (Veselovskii et al., 2015) or 355 nm (Ortiz-Amezcuca et al., 2020), but also allows to obtain the extinction coefficient at 1064 nm. The first direct extinction measurement at 1064 nm ever was performed in a cirrus cloud (Haarig et al., 2016) with the Backscatter Extinction lidar-Ratio Temperature Humidity profiling Apparatus (BERTHA).

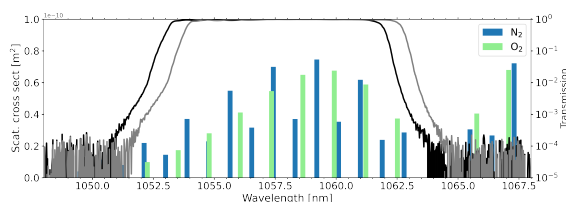


Figure 1. The scattering cross section of the rotational Raman lines of nitrogen (N₂, blue) and oxygen (O₂, green) excited at a wavelength of 1064.14 nm (left y-axis). Additionally, the transmission curves of both interference filters are shown on a logarithmic scale (right y-axis). The suppression of the elastic backscatter at 1064.14 nm is better for interference filter 1 (black line) than for interference filter 2 (grey line).

The same technique introduced in BERTHA is now also applied for the Polly^{XT} observations at Mindelo, Cabo Verde. We
 70 will give a short description here. A full technical description of the implementation of the rotational Raman technique was already presented by Haarig et al. (2016). Interference filters (Alluxa, Stanta Rosa, USA, <https://alluxa.com/>) centered at 1058 nm with transmissions from 1053 to 1062 nm (9 nm band width) were used to detect the rotational Raman lines of nitrogen and oxygen as shown in Fig. 1. Wang et al. (2024b) recently presented a slightly different approach by using an interference filter centered at 1056 nm with a width of 6 nm to reduce the spectral cross talk. Fig. 1 presents the calculated Raman lines
 75 and the transmission curves of the interference filters as measured by the manufacturer. The filters are slightly different in their transmission properties. The suppression at the emitted laser wavelength of 1064.14 nm is crucial and is better achieved for filter 1 (shown as black line in Fig. 1). By using two interference filters we were able to achieve a suppression of 8 orders of magnitude of the elastically backscattered light. The correction of the still remaining spectral cross talk and of the temperature dependence of the rotational Raman lines is described in the Appendix A1. The molecular signal at 1064 nm is 81 times
 80 weaker compared to the one at 355 nm due to the strong wavelength dependence of Rayleigh scattering. This is one of the main limitations of the rotational Raman method at 1064 nm. Therefore, long temporal and vertical averaging is necessary, which

limits our current approach to the analysis of case studies only. The limitations of the rotational Raman method at 1064 nm are discussed in greater detail in Appendix A2.

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3 Measurements for marine and continental aerosol

Extinction and lidar ratio measurements at 1064 nm of marine and continental aerosol are still lacking in the literature. Therefore, we report these cases in greater detail in the following section, before the spectral slope of various aerosol types is discussed in Section 4. Additionally, we investigate the effect of hygroscopic growth on the lidar ratio of continental aerosol in

90 Section 3.3.

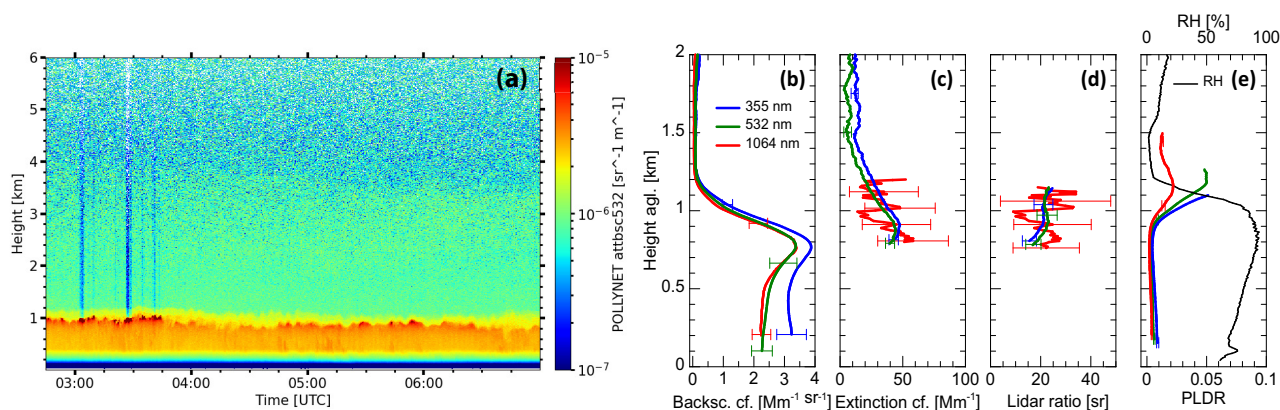


Figure 2. Lidar observations of marine aerosol below 1 km height at Mindelo, Cabo Verde, on 26 March 2022: (a) Time-height display of Polly^{XT} observations (attenuated backscatter coefficient at 532 nm). The vertical profiles are provided above ground level (agl.) between 03:45 and 05:50 UTC: (b) Particle backscatter coefficient. (c) Particle extinction coefficient. (d) Lidar ratio. (e) Particle linear depolarization ratio (PLDR) and relative humidity (RH, black line, upper x-axis). A gliding average of 200 m was applied for the backscatter coefficient and depolarization ratio and of 750 m for the extinction coefficient and lidar ratio.

3.1 Triple-wavelength lidar ratio observations of marine aerosol at Cabo Verde

Since the summer of 2021, TROPOS operates a new Polly^{XT} lidar at the Ocean Science Centre Mindelo (16.878°N, 24.995°W) at São Vicente Island, Cabo Verde. Additionally to previous Polly^{XT} systems described in Engelmann et al. (2016), this lidar has the capability to measure the depolarization ratio and the extinction coefficient at 1064 nm. These capabilities were used

95 for the first time in the study of Gebauer et al. (2024) to derive the so-called 3+3+3 data set (3 backscatter coefficients, 3 extinction coefficients, 3 depolarization ratios) of Saharan dust and sulfate aerosol from the Cumbre Vieja eruption in autumn of 2021.



Here, we present a case of marine aerosol observed on 26 March 2022 (Fig. 2). Cumulus clouds were detected on top of the marine boundary layer at around 1 km height until 3:45 UTC. These optically dense water clouds cause significant laser light attenuation indicated by the vertical blue columns above the marine boundary layer. We use the cloud-free observations after 03:45 UTC for aerosol profiling. Another aerosol layer reaching up to 4 km height can be seen above the marine boundary layer. This faint layer has a much weaker backscatter signal of $0.2 \text{ Mm}^{-1} \text{ sr}^{-1}$ compared to a maximum backscatter coefficient of $3.1 \text{ Mm}^{-1} \text{ sr}^{-1}$ (at 532 nm) in the marine aerosol layer (Fig. 2b). It is a mixed layer (PLDR of 0.13 ± 0.02 at 532 nm) of mineral dust, urban haze and biomass burning smoke originating from the African continent as already often observed during SAMUM-2 (Saharan Mineral Dust Experiment) campaign in 2008 (Tesche et al., 2011). Focusing on the layer below 1 km height, the low depolarization ratio (< 0.01) and a lidar ratio of 20–25 sr (at 355 and 532 nm) plus the location in the Atlantic Ocean undoubtedly characterize this layer as a clean marine boundary layer. The optical properties are summarized in Table 1. The extinction and lidar ratio data at 1064 nm are quite noisy and the uncertainties are high. Nevertheless, the spectrally neutral behavior of the lidar ratio is obvious and will be further discussed in Section 4.1. An interesting feature is the increase in the depolarization ratio on top of the marine aerosol layer between 1.0 and 1.5 km height up to 0.05 at 532 nm. We have observed in the past that the increase of the depolarization ratio is linked to drying marine aerosol (Haarig et al., 2017b; Bohlmann et al., 2018). Drier air masses from above mixed with the humid marine layer are causing this process and also lead to the dissolving of the cumulus clouds. However, the presence of some mineral dust particles and its effect on the depolarization ratio cannot be fully excluded here.

3.2 Triple-wavelength lidar ratio observations of continental aerosol at Leipzig

The summer of 2018 was one of the hottest and driest summers in Germany since the beginning of systematic weather reporting. These conditions favored the development of vertically extended planetary boundary layers of continental aerosol up to 4 km height. An example is shown in Fig. 4. The measurement was performed at Leipzig (51.12°N , 12.3°E) on 7 August 2018. In the night of 7 August 2018, five Raman polarization lidars were operated simultaneously at TROPOS: BERTHA, MARTHA (Multiwavelength Atmospheric Raman lidar for Temperature, Humidity and Aerosol profiling, e.g., Jimenez et al., 2019), Polly^{XT}–OCEANET (e.g., Engelmann et al., 2021), Polly^{XT}–LACROS (Leipzig Aerosol and Cloud Remote Observations System, e.g., Radenz et al., 2021) and Polly 1st (e.g., Engelmann et al., 2016). A picture with the five lidar beams (visible at 532 nm) is shown in Fig. 3. The lidar ratio at 532 nm was observed with all systems, the lidar ratio at 355 nm with all lidars except of Polly 1st, and the lidar ratio at 1064 nm only with BERTHA. The BERTHA lidar was manually operated for 2 hours and 22 minutes (Fig. 4a). The essential cross-talk correction (see Sect. A1) was done using the liquid clouds between 3 and 4 km height before 21:00 UTC. A spectral cross-talk correction factor of $c_s = 0.125 \pm 0.005$ was determined. As already mentioned, the same interference filters have been used as at Cabo Verde (since 2021). However, the suppression of the elastic signal by neutral density filters was much stronger (optical thickness of 3.1 compared to 1.0 at Cabo Verde, Section 3.1). The measurement of the extinction coefficient at 1064 nm with BERTHA was only possible for heights > 2.2 km because of remaining overlap effects in the near-infrared channels at heights below.

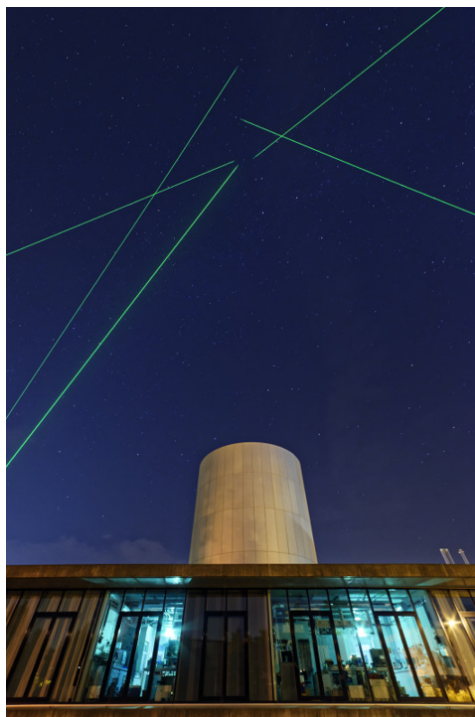


Figure 3. 5 Lidar beams over the Leibniz Institute for Tropospheric Research in the night of 7 August 2018 (Photo by Tilo Arnhold, public relations of TROPOS).

The recorded profiles are shown in Fig. 4b-e for the time between 20:55 and 22:51 UTC in order to exclude the liquid clouds at the beginning of the observations. At around 2.8 km height, a significant increase in the backscatter and extinction coefficient was observed (Fig. 4b,c). This increase is linked to an increase in relative humidity and therefore to hygroscopic growth of the continental aerosol particles. The relative humidity (shown in Fig. 4e) was calculated from the water vapor measurements of Polly^{XT}-LACROS calibrated with the integrated water vapor derived from a microwave radiometer (HATPRO, RPG, Mecken-

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heim, Germany) and the temperature profile of ECMWF (European Centre for medium range weather forecast). The formation of the liquid clouds before 21:00 UTC in this layer and the decrease in the depolarization ratio are further indications of the increase in relative humidity with height as obvious from the derived RH profile (Fig. 4e). A detailed discussion about the

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hygroscopic growth and the calculation of growth factors of the backscatter coefficient, extinction coefficient and lidar ratio at all three lidar wavelengths is given in the next section (Section 3.3), because here we want to focus on the spectral behavior of the lidar ratio.

The intensive optical properties are summarized in Table 1. They are separated for the conditions below 80% RH (reached at 2.61 km height) and above 85% RH (observed between 2.94 and 3.30 km height). This separation is motivated by the find-

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ings of Skupin et al. (2016), who showed the extinction enhancement factor starts to increase drastically at an RH of 85%. A similarly strong enhancement for the extinction at around 2.9 km height can be seen also in our measurements in Fig. 4c. The

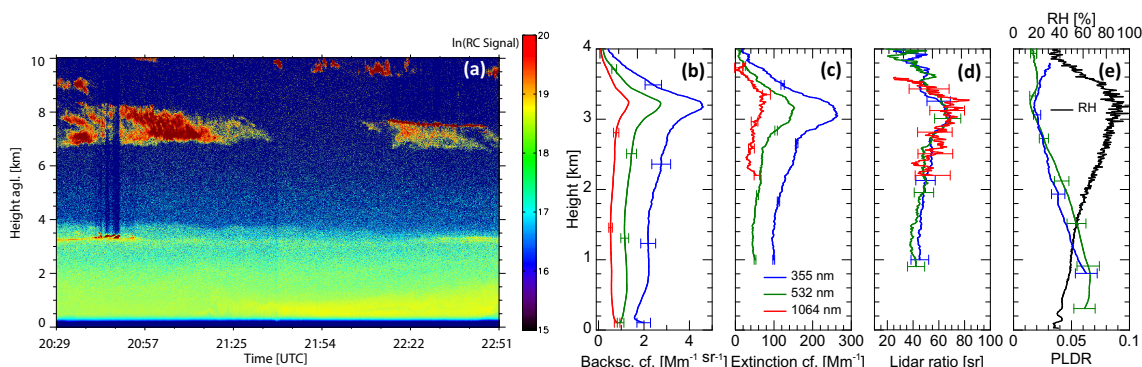


Figure 4. Lidar observations of continental aerosol at Leipzig on 7 August 2018: (a) Time-height display of BERTHA observations (range-corrected signal at 532 nm). The vertical profiles of the optical properties are shown above ground level (agl) between 20:55 and 22:51 UTC: (b) Particle backscatter coefficient. (c) Particle extinction coefficient. (d) Lidar ratio. (e) Particle linear depolarization ratio (PLDR) and relative humidity (RH, black line, upper x-axis). The data at 1064 nm (red) are recorded with BERTHA, the data at 355 (blue) and 532 nm (green) are shown from the Polly^{XT}–LACROS measurements. A gliding average of 200 m was applied for the backscatter coefficient and depolarization ratio and of 500 m (355 and 532 nm) for the extinction coefficient and lidar ratio and for 1064 nm of 750 m.

increase of the extinction coefficient is stronger than the increase of the backscatter coefficient, leading to an increase of the lidar ratio with increasing RH. Our observations show that the lidar ratios for hygroscopically grown continental aerosol are in the range of 65 – 69 sr for all three lidar wavelengths while lidar ratios at lower RH are in the range of 48 – 54 sr. An increase of the lidar ratio with RH for continental aerosol was predicted by Ackermann (1998), simulated by Zhao et al. (2017) in the North China Plain and retrieved from in situ observations by Düsing et al. (2021) in central Germany.

To avoid the influence of hygroscopic growth in our discussion about the spectral slope of the lidar ratio, we will focus on the aerosol layer below 2.61 km height as representative for continental aerosol. Here, the lidar ratio ranges between 48 and 54 sr at all three lidar wavelengths. At 355 and 532 nm, a variety of aerosol typing schemes exist. The low depolarization ratios (around 0.03), the intermediate lidar ratios (45 – 55 sr) and the enhanced Ångström exponents would result in "Anthropogenic pollution" according to Groß et al. (2013) and are in perfect agreement with "Central European background" discussed in Floutsi et al. (2023). Furthermore, it is inline with long-term observations of planetary boundary layer (PBL) aerosol properties in central Europe (Mattis et al., 2004). Thus, we are confident that these measurements represent continental aerosol and can be used to compare it to the category of "Clean continental" in CALIPSO v4 aerosol typing which will be done in Section 4.2.

3.3 Hygroscopic growth of continental aerosol at Leipzig

In this section we describe the hygroscopic growth of continental aerosol which was observed on 7 August 2018 at Leipzig. The measurements are described in the previous section, where we focused on the spectral lidar ratio. Here, we derive the hygroscopic growth factors for the backscatter coefficient, the extinction coefficient and the lidar ratio. To our knowledge, it is



Table 1. The optical properties observed in the continental (Leipzig, 7 August 2018) and marine (Mindelo, 26 March 2022) boundary layer. The range of relative humidity (RH) is provided additionally. In the case of continental aerosol, the values at 1064 nm were measured with BERTHA, and the values at 355 and 532 nm were measured with Polly^{XT}–LACROS. λ – wavelength, LR – lidar ratio, PLDR – particle linear depolarization ratio, EAE – extinction-related Ångström exponent, BAE – backscatter-related Ångström exponent

	λ (nm)	Continental		Marine
RH		70 – 80%	85 – 92%	> 80%
LR (sr)	355	54.2 ± 8.2	65.0 ± 9.8	20.4 ± 3.4
	532	48.1 ± 7.4	65.3 ± 9.9	21.8 ± 3.9
	1064	48.7 ± 12.8	69.0 ± 14.1	21.2 ± 17.4
PLDR	355	0.029 ± 0.01	0.019 ± 0.01	0.007 ± 0.005
	532	0.033 ± 0.01	0.017 ± 0.01	0.005 ± 0.005
	1064	–	–	0.004 ± 0.005
EAE	355/532	1.95 ± 0.10	1.38 ± 0.07	0.24 ± 0.29
	532/1064	0.97 ± 0.32	1.15 ± 0.20	0.08 ± 1.11
	355/1064	1.33 ± 0.20	1.23 ± 0.13	0.14 ± 0.70
BAE	355/532	1.64 ± 0.52	1.36 ± 0.52	0.45 ± 0.52
	532/1064	1.00 ± 0.31	1.28 ± 0.31	0.04 ± 0.30
	355/1064	1.23 ± 0.19	1.31 ± 0.19	0.19 ± 0.19

165 the first lidar study which reports the hygroscopic growth factors of these 3 quantities at 355, 532 and 1064 nm simultaneously. Therefore, it offers the possibility to study the spectral dependence of the hygroscopic behavior of the aerosol optical properties. In order to link these changes in the optical properties to hygroscopic growth, it is important to check first that the same air mass and aerosol load was present in this height range. The HYSPLIT backtrajectories (Fig. 5a) for the heights of 2.5, 3.0 and 3.5 km indicate the same origin of the air masses. The three days prior to the observation, the trajectories followed exactly
 170 the same path from northern France to central Germany. The origin of the air masses from the Atlantic Ocean might result in some remaining marine particles in the observed air masses. Additionally, we checked the water vapor mixing ratio from the Polly^{XT}–LACROS observations and the potential temperature from the Global Data Assimilation System (GDAS 1.0). Both are constant in the height range from 2.0 to 3.3 km, as shown in Fig. 5b. This behavior clearly indicates a well mixed atmosphere, where the same aerosol load was present. All changes in the optical properties in this height range can therefore be attributed
 175 to changes in RH.

In order to study the hygroscopic growth, we compute enhancement factors f_χ of quantity χ (backscatter coefficient (bsc), extinction coefficient (ext) and lidar ratio (LR)) at wavelength λ

$$f_\chi(\text{RH}, \lambda) = \frac{\chi(\text{RH}, \lambda)}{\chi(\text{RH}_{\text{ref}}, \lambda)}. \quad (1)$$

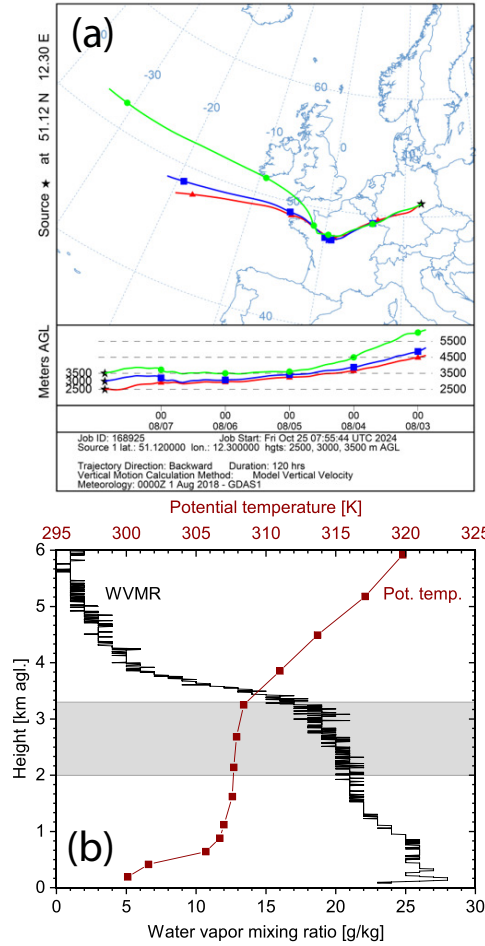


Figure 5. (a) 5-day backtrajectory calculated by HYSPLIT (Stein et al., 2015). The trajectories arriving at 2.5, 3.0 and 3.5 km height shared the same atmospheric path for the 48 – 72 h prior to their arrival at Leipzig. (b) Water vapor mixing ratio (WVMR, lower x-axis) from the Polly^{XT}–LACROS measurements and potential temperature (upper x-axis) from GDAS 1.0 profiles. The height range (2.0 – 3.3 km above ground level (agl.)) used for the hygroscopic study is shaded in grey. The constant potential temperature and WVMR are clear indications for a stable atmospheric layering.

The enhancement factors are normalized to the reference relative humidity RH_{ref} . It is 60% for all quantities except of the
 180 extinction coefficient and lidar ratio at 1064 nm, where a RH_{ref} of 75% is used because of overlap issues in the 1064 nm Raman signals. The hygroscopic growth of aerosol optical properties is commonly described by the Hanel parameterization (Hanel, 1976) which introduces the hygroscopic growth factor $\gamma_{\chi}(\lambda)$ which describes the dependency of a quantity χ with RH. It depends on aerosol type (in this case continental aerosol) and wavelength. The Hanel parameterization is given by

$$f_{\chi}(RH, \lambda) = \left(\frac{1 - RH/100}{1 - RH_{ref}/100} \right)^{-\gamma_{\chi}(\lambda)} \quad (2)$$

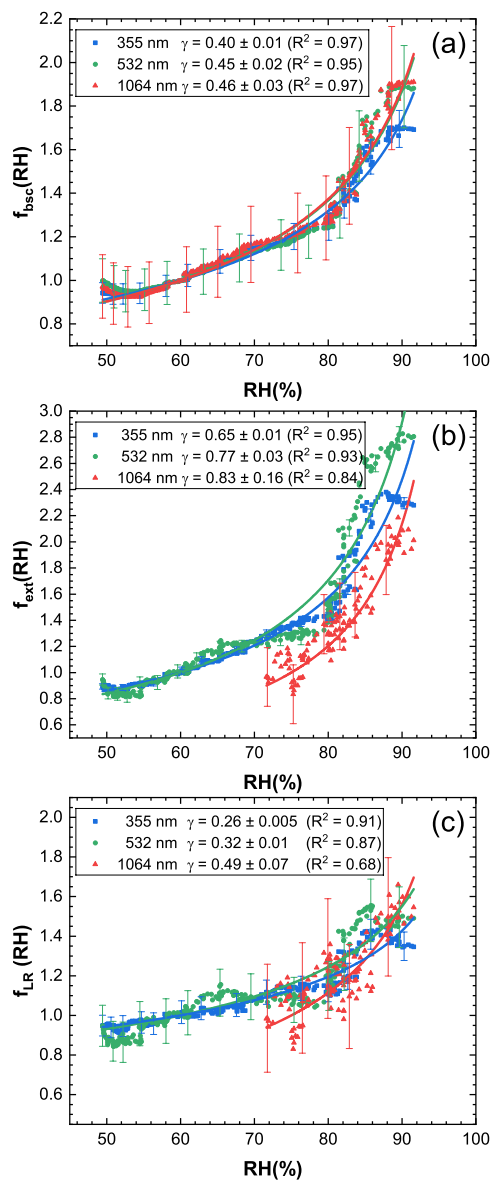


Figure 6. (a) Backscatter enhancement factor f_{bsc} , (b) extinction enhancement factor f_{ext} and (c) lidar ratio enhancement factor f_{LR} . A relative humidity (RH) of 60% is used to normalize the enhancement factors for all quantities except of the extinction coefficient and lidar ratio at 1064 nm, there 75% RH are used. The hygroscopic growth parameter γ at 355, 532 and 1064 nm is given for all quantities. Additionally, the R^2 value of the fit following equation 2 is listed. The uncertainty already includes the fit error and the uncertainty of the quantity itself (see last column of Table 2).

185 The results for all 9 quantities are presented in Fig. 6 where the $\gamma_{\chi}(\lambda)$ parameter is given. The first finding is that the extinction enhancement is stronger (γ_{ext} between 0.65 and 0.83) than the backscatter enhancement (γ_{bsc} between 0.40 and 0.46) which



leads to an enhancement of the lidar ratio (γ_{LR} between 0.26 and 0.49), because the lidar ratio is the extinction-to-backscatter ratio. The second finding is that the lidar ratio enhancement increases with wavelength from $\gamma_{LR} = 0.26 \pm 0.005$ at 355 nm to 0.32 ± 0.01 at 532 nm and further to 0.49 ± 0.07 at 1064 nm. This increase with wavelength is already visible in the backscatter and extinction enhancement where the lowest γ values are observed at 355 nm.

To estimate the uncertainty of γ we consider the error of the fit and the uncertainty of the quantity itself (Table 2). The uncertainty of the backscatter coefficient is assumed to be 5, 10 and 15% at 355, 532 and 1064 nm, respectively, as outlined in the appendix of Haarig et al. (2017a). The relative uncertainty of the extinction coefficient and the lidar ratio in the studied layer is given in Table 2. There, the overall absolute uncertainty of γ is calculated from the square root of the quadratic sum of both uncertainties. This uncertainty is provided with the plots in Figure 6.

In the following, we compare our findings to previous observations.

Table 2. The hygroscopic growth factors γ of quantity χ , the absolute uncertainty of the fit σ_{fit} , the relative uncertainty of the quantity σ_χ in the studied RH interval (height interval) and the resulting absolute uncertainty of σ_γ of the hygroscopic growth parameter.

χ	γ	σ_{fit}	σ_χ	σ_γ
Bsc355	0.40	0.002	5%	0.01
Bsc532	0.45	0.004	10%	0.02
Bsc1064	0.46	0.003	15%	0.03
Ext355	0.65	0.005	2.7%	0.01
Ext532	0.77	0.007	4.9%	0.03
Ext1064	0.83	0.016	22.8%	0.16
LR355	0.26	0.003	5.7%	0.005
LR532	0.32	0.005	11.2%	0.01
LR1064	0.49	0.015	27.3%	0.07

Hygroscopic growth of the backscatter coefficient

Sicard et al. (2022) studied the hygroscopic growth of the backscatter coefficient on a statistical basis at 355, 532 and 1064 nm. Their measurements were performed at Barcelona, Spain, at the Mediterranean Sea, which is much stronger influenced by highly hygroscopic sea salt particles than continental Leipzig. They observed backscatter hygroscopic growth parameters of 0.65 to 0.81. Therefore, these measurements are not comparable to our observations of continental aerosol at Leipzig (400 km away from the ocean). Our backscatter hygroscopic growth parameters and their spectral dependence are comparable to those measured by Bedoya-Velázquez et al. (2018) in the Sierra Nevada in southern Spain: $\gamma_{bsc}(355 \text{ nm}) = 0.40 \pm 0.01$ and $\gamma_{bsc}(532 \text{ nm}) = 0.48 \pm 0.01$. Although the site is also close to the Ocean, the station is located at the northern slopes of the Sierra Nevada mountains which limits the influence of marine air masses. Miri et al. (2024) used the fluorescence backscatter coefficient to normalize the backscatter coefficient in order to ensure that the aerosol load does not change during the measurements. In the case of urban pollution over Lille, northern France, they report a $\gamma_{bsc}(532 \text{ nm})$ of 0.47 ± 0.03 , which aligns well with our



value of 0.45 ± 0.02 . Navas-Guzmán et al. (2019) studied a smoke mixture in Switzerland and found similar values at 355 nm ($\gamma_{\text{bsc}} = 0.48 \pm 0.08$), but then a spectral decrease towards the wavelength of 1064 nm ($\gamma_{\text{bsc}} = 0.29 \pm 0.08$). Pérez-Ramírez et al. (2021) studied mixtures of sulfate and organics at the East coast of North America and found a spectral decrease of the γ_{bsc} parameter but with values in the same range (0.31 till 0.46) and one value at 355 nm which reached up to 0.65 ± 0.10 . It can be concluded that there is a slight wavelength dependence on the γ_{bsc} parameter for continental aerosol. However, this dependence sensitively depends on the composition of the aerosol mixture, which is not well defined in the case of continental aerosol.

215 **Hygroscopic growth of the extinction coefficient**

The hygroscopic growth of the extinction coefficient was mostly studied with in situ instrumentation on ground (for details see Zieger et al., 2013; Titos et al., 2016, 2021). Measurements with a spectral aerosol extinction monitoring system (Skupin et al., 2016) at ground-level found $\gamma_{\text{ext}}(550 \text{ nm}) = 0.46 \pm 0.30$ for 4 years of measurements at Leipzig, so our observations of $\gamma_{\text{ext}}(532 \text{ nm}) = 0.77 \pm 0.03$ would be on the upper edge of the long-term observations and represent a strong hygroscopic case. As mentioned above, a slight marine influence in the observed air masses cannot be completely excluded. Jefferson et al. (2017) found in a 7-year study over the southern Great Plains in North America values of $\gamma_{\text{ext}}(550 \text{ nm}) = 0.44 \pm 0.16$ for sub-1 μm particles and 0.40 ± 0.15 for sub-10 μm particles.

Hygroscopic growth of the lidar ratio

Previous studies of the lidar ratio enhancement used airborne particle properties to calculate the lidar ratio enhancement. Ackermann (1998) modeled the lidar ratio enhancement for continental aerosol and found that the lidar ratios vary between 40 and 80 sr. However, his modeled spectral behavior is not consistent with our observations. Already Ferrare et al. (1998) studied comprehensively the hygroscopic growth of the backscatter and extinction coefficient by using ground based Raman lidar measurements at the Southern Great Plains (Central USA, continental aerosol) with collocated airborne in situ observations. They derived the hygroscopic growth parameter γ_r from the in situ measurements to describe the changes in particle radius r . A γ_r of 0.3 ± 0.05 was in good agreement with Raman lidar observations. In these observations, they found a strong increase in the extinction coefficient at 351 nm with RH and a weaker increase in the lidar ratio ($f_{\text{LR}} = 1.26$ from 60% to 90–95% RH). For the same RH range the lidar ratio enhancement factor in our observations was 1.34 at 355 nm. The study of Zhao et al. (2017) was motivated by the investigation of the influence of the lidar ratio with RH on the retrieval of elastic lidars where a lidar ratio has to be assumed. However, they used a different parameterization of the hygroscopic growth that makes it difficult to compare to our results. The hygroscopic growth of the lidar ratio was studied by Düsing et al. (2021) for the rural site of Melpitz, Germany. They used in situ measurements from an airborne platform as input into Mie calculations to derive the lidar ratio enhancement factors. The strongest enhancement was found at 532 nm ($\gamma_{\text{LR}} = 0.48 \pm 0.01$) and lower values at 355 ($\gamma_{\text{LR}} = 0.29 \pm 0.01$) and 1064 nm ($\gamma_{\text{LR}} = 0.31 \pm 0.01$). In contrast to their modeling results, we observe from Raman lidar observations an increase in the lidar ratio enhancement factor with wavelength. Only the value at 355 nm ($\gamma_{\text{LR}} = 0.26 \pm 0.005$) is comparable to their study. However, in general the agreement of the modeled values at 532 and 1064 nm with the lidar observations was poor, so that closure could not be reached by Düsing et al. (2021).

With our study, we could show with direct measurements that the lidar ratio of continental aerosol depends on RH and we



could derive hygroscopic growth factors for each wavelength for the backscatter coefficient, extinction coefficient and lidar ratio.

245 4 Discussion of the spectral slope of the lidar ratio

In this section, we discuss the spectral dependence of the lidar ratio based on the observations at 355, 532 and 1064 nm. An extensive literature study to constrain the lidar ratio at 1064 nm from auxiliary observations and calculations was presented by Kim et al. (2018) for the CALIPSO v4 automated aerosol classification and lidar ratio selection algorithm. They estimated the lidar ratio at 1064 nm in order to analyze the CALIPSO observations at this wavelength. Floutsi et al. (2023) provided a comprehensive study about the lidar ratios at 355 and 532 nm in the collection of depolarization ratios, lidar ratios and the Ångström exponents (so-called DeLiAn). Hence, the special focus of the present study is on the spectral slope from 532 to 1064 nm. So, we subdivide our findings according to the spectral behavior of the lidar ratio from 532 to 1064 nm in spectrally neutral (Sect. 4.1), decreasing (Sect. 4.2) and increasing behavior (Sect. 4.3). Then, we want to shed light on the connection of the extinction and backscatter-related Ångström exponent to the Ångström exponent of the lidar ratio (Sect. 4.4).

255 Figure 7 summarizes the spectral behavior of the lidar ratio observed with Raman lidars for five aerosol types and ice clouds and compares the results to the CALIPSO v4 algorithm. Table 3 lists the references and their abbreviations used in the following and Tables 4 and 5 provide the exact values for the results presented in Fig. 7.

Table 3. References and their abbreviations used in the following tables and figures.

Abbreviation	Reference
Fl23	Floutsi et al. (2023)
Ge24	Gebauer et al. (2024)
Ha16	Haarig et al. (2016)
Ha18	Haarig et al. (2018)
Ha22	Haarig et al. (2022)
Ho20	Hofer et al. (2020)
Ki18	Kim et al. (2018)
MA17	Mamouri and Ansmann (2017)
Ma04	Mattis et al. (2004)
Oh20	Ohneiser et al. (2020)
Om10	Omar et al. (2010)
Te09	Tesche et al. (2009)

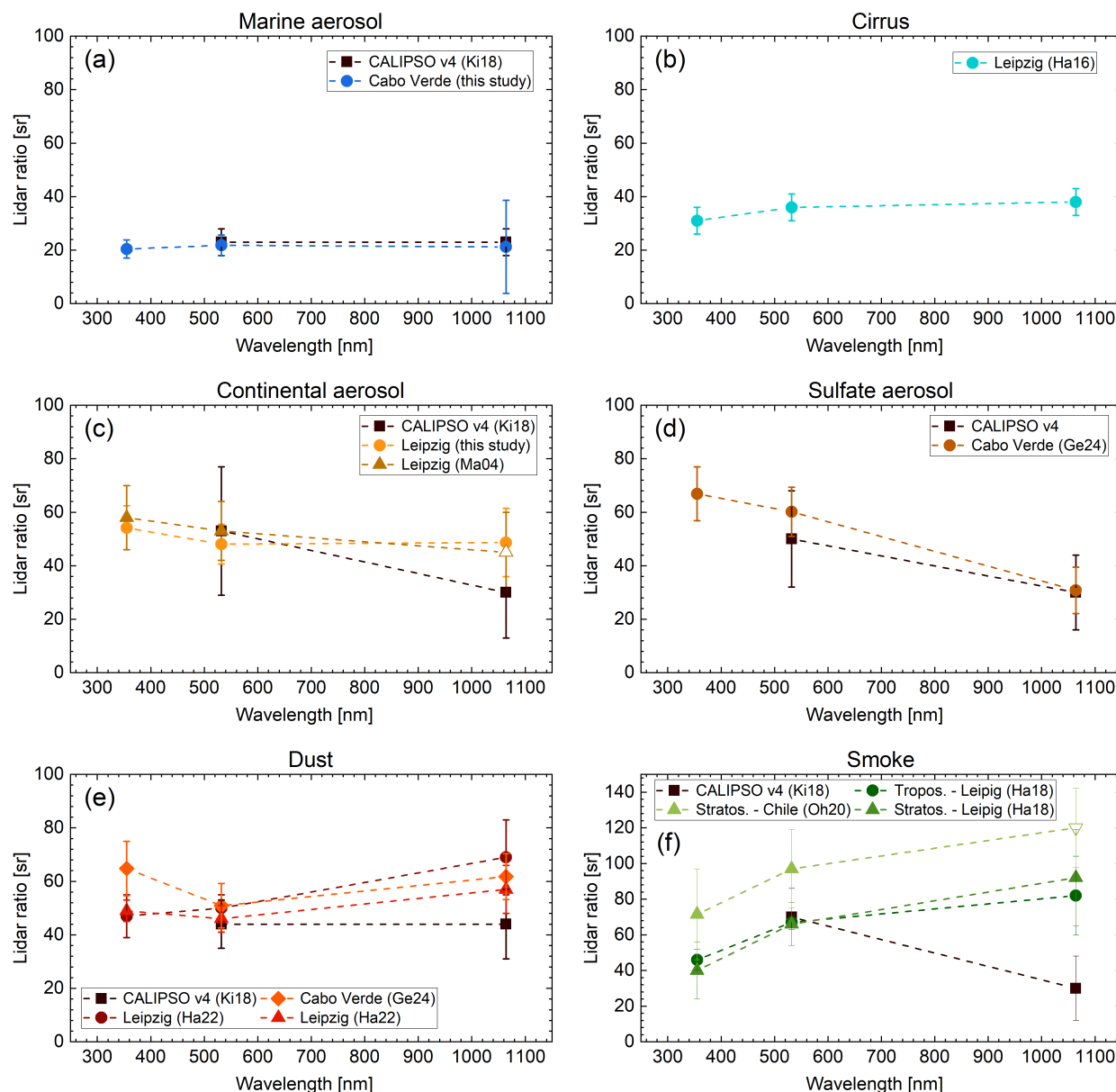


Figure 7. Spectral slope of the lidar ratio for (a) marine aerosol, (b) cirrus clouds, (c) continental aerosol, (d) sulfate aerosol, (e) mineral dust and (f) smoke (in the troposphere and stratosphere). The Raman lidar observations shown in filled colored symbols are compared to the assumptions made in the CALIPSO v4 aerosol typing and lidar ratio assignment algorithm (black squares). Open symbols refer to a combination of lidar and photometer observations. The references are listed in Tab. 3 and the values are provided for convenience in Table 4 and separately for mineral dust in Table 5.



Table 4. The spectral lidar ratio and the ratio of lidar ratios discussed in this study and compared to CALIPSO v4 (values at 532 and 1064 nm, Kim et al., 2018) and DeLiAn (values at 355 and 532 nm Floutsis et al., 2023). The abbreviations for the references are given in Tab. 3.

Aerosol information	Lidar ratio (sr)			Ratio of lidar ratios		Reference
	355 nm	532 nm	1064 nm	532/355	1064/532	
Marine aerosol						
Cabo Verde	20.4 ± 3.4	21.8 ± 3.9	21.2 ± 17.4	1.07 ± 0.26	0.97 ± 0.82	this study
Clean marine (CALIPSO v4)	–	23 ± 5	23 ± 5	–	1.00 ± 0.31	Ki18
Clean marine (DeLiAn)	22.4 ± 5.6	21.9 ± 13.9	–	0.98 ± 0.67	–	Fl23
Ice clouds						
Leipzig	31 ± 5	36 ± 5	38 ± 5	1.16 ± 0.25	1.06 ± 0.20	Ha16
Continental aerosol						
Leipzig	54.2 ± 8.2	48.1 ± 7.4	48.7 ± 12.8	0.89 ± 0.19	1.01 ± 0.31	this study
Leipzig	58 ± 12	53 ± 11	45 ± 15 ^a	0.91 ± 0.27	0.85 ± 0.33 ^a	Ma04
Clean continental (CALIPSO v4)	–	53 ± 24	30 ± 17	–	0.57 ± 0.41	Ki18
Central Europ. background (DeLiAn)	57.0 ± 4.7	56.2 ± 8.3	–	0.99 ± 0.17	–	Fl23
Sulfate						
Cabo Verde	66.9 ± 10.1	60.2 ± 9.2	30.8 ± 8.7	0.90 ± 0.19	0.51 ± 0.16	Ge24
Sulfate/other (CALIPSO v4)	–	50 ± 18	30 ± 14	–	0.60 ± 0.35	Ki18
Smoke						
Troposphere, Leipzig	46 ± 6	67 ± 4	82 ± 22	1.46 ± 0.21	1.22 ± 0.34	Ha18
Stratosphere, Leipzig	40 ± 16	66 ± 12	92 ± 27	1.65 ± 0.72	1.39 ± 0.48	Ha18
Stratosphere, Punta Arenas	72 ± 26	97 ± 22	120 ± 22 ^a	1.36 ± 0.57	1.24 ± 0.36 ^a	Oh20
Elevated smoke (CALIPSO v4)	–	70 ± 16	30 ± 18	–	0.43 ± 0.28	Ki18
Smoke (DeLiAn)	68.2 ± 7.4	71.8 ± 11.1	–	1.05 ± 0.20	–	Fl23

^a Estimated using lidar and photometer.

4.1 Spectrally neutral behavior of the lidar ratio

For three case studies, a spectrally neutral behavior of the lidar ratio from 532 to 1064 nm is shown in Fig. 7a–c. The values are provided in Table 4.

Marine aerosol

The spectrally neutral behavior of the lidar ratio from 355 to 532 nm was already observed before as collected by Floutsis et al. (2023). According to our observations at Cabo Verde (Sect. 3.1), the lidar ratio at 1064 nm is not increasing compared to the lidar ratio at 532 nm (Fig. 7a). This finding contradicts previous modeling results (e.g., Ackermann, 1998; Haarig et al., 2017b; Kahnert and Kanngießer, 2024), but it confirms the assumptions introduced for CALIPSO v4 (Kim et al., 2018). The lidar ratio



at 1064 nm was decreased from 45 ± 23 sr (CALIPSO v3) to 23 ± 5 sr (CALIPSO v4) and is now the same value as for 532 nm. This reduction was based on the SODA (Synergized Optical depth of Aerosols) observations (Josset et al., 2011). The SODA technique was applied to marine aerosol lidar ratios at 1064 nm (Josset et al., 2012). Also, the marine aerosol network observations presented by Sayer et al. (2012) and AERONET observations at marine sites (Cattrall et al., 2005) indicated a spectrally neutral behavior of the marine lidar ratio, which could be now confirmed by Raman lidar observations for marine aerosol under humid conditions.

Cirrus clouds

The large ice crystals of cirrus clouds with sizes typically $> 100 \mu\text{m}$ introduce a spectrally neutral behavior in the wavelength range of 355 to 1064 nm, both in the backscatter coefficient and the extinction coefficient and therefore in the lidar ratio as well (Fig. 7b, Sect. 4.4). Voudouri et al. (2020) studied the lidar ratios of cirrus clouds at three different locations and found no spectral dependence in the wavelength range from 355 to 532 nm. Haarig et al. (2016) presented the first measurements of the lidar ratio at 1064 nm in a cirrus cloud (at Leipzig, Germany) and found no spectral dependence up to 1064 nm. The backscatter color ratio between 1064 and 532 nm was intensively studied by Vaughan et al. (2010) who used a large data set of cirrus observations from airborne lidar measurements. They found a color ratio of 1.01 ± 0.25 . Under the assumption that the extinction coefficient does not change in the given wavelength range (because the ice crystals are much larger than the wavelengths), the lidar ratio should not change as well. However, the study of Vaughan et al. (2010) pointed out that natural variability (25%) of the backscatter color ratio is higher than expected and thus in some cases a spectral dependence is possible.

Continental aerosol

Continental aerosol is a mixture of anthropogenic pollution, biological particles and soil dust and may vary with location and season. Therefore, its optical properties can vary considerably for different mixtures of light absorbing and scattering aerosol fractions.

Mattis et al. (2004) studied the aerosol properties in the PBL above Leipzig, Germany, in the years of 2000 to 2003. She and her coauthors found mean lidar ratios of 58 ± 12 sr at 355 nm and 53 ± 11 sr at 532 nm. Furthermore, they used a combination of sun photometer aerosol optical depth (AOD) and lidar column-integrated backscatter coefficient to estimate the lidar ratio at 1064 nm, which was found to be 45 ± 15 sr. These values are in line with the measurements of continental aerosol presented in our study (Sect. 3.2).

The presented case study of continental aerosol shows a spectrally neutral behavior of the lidar ratio. The lidar ratio values for $\text{RH} < 80\%$ are provided in Table 4. The lidar ratio increases with increasing RH. However, the spectrally neutral behavior remains even under increasing relative humidity as discussed in Sections 3.2 and 3.3. In contrast, CALIPSO v4 assumes a lower lidar ratio at 1064 nm (30 ± 17 sr) compared to 532 nm (53 ± 24 sr). However, considering the large variety of continental aerosol particles, we refrain from advising a change in the CALIPSO lidar ratio selection algorithm. The case presented here was characterized by enhanced RH (70 – 80 %) and following the discussion of the hygroscopic growth of the lidar ratio (Sect. 3.3), dry continental aerosol might have lower lidar ratios. The hygroscopic growth of the lidar ratio at 1064 nm (hygroscopic growth parameter $\gamma_{\text{LR}} = 0.49 \pm 0.07$) was stronger compared to 355 nm ($\gamma_{\text{LR}} = 0.26 \pm 0.005$). An extrapolation of the hygroscopic growth curve using equations 1 and 2 to a dry RH of 40% would lead to a lidar ratio of 30 sr at 1064 nm, 39 sr at



532 nm and 43 sr at 355 nm. This extrapolation is linked to a high uncertainty and therefore it is shown here just to describe the trend that can be expected for dry continental aerosol. Furthermore, it highlights the importance of considering the relative humidity in the lidar ratio selection in the case of continental aerosol as discussed by Zhao et al. (2017).

4.2 Decreasing lidar ratio from 532 to 1064 nm

For sulfate-dominated aerosol discussed in Gebauer et al. (2024), a decrease in the lidar ratio towards 1064 nm was observed, as shown in Fig. 7d. The values are provided in Table 4.

Sulfate aerosol

Gebauer et al. (2024) observed the volcanic sulfate plume from the Cumbre Vieja eruption on La Palma (Canary Islands, Spain) in early fall of 2021. The Raman lidar measurements were performed at Mindelo, Cabo Verde, with the same Polly^{XT} as used for the observations of marine aerosol in Section 3.1. Here, the case study in Fig. 7d presents a mixture of sulfate and marine aerosol, because the sulfate aerosol arrived in the marine aerosol layer at the Cabo Verdian islands. However, this mixture was strongly dominated by sulfate aerosol with >80 % contribution to the AOD at 532 nm in the layer (Gebauer et al., 2024). The lidar ratio of the mixture decreased from 60.2 ± 9.2 sr at 532 nm to 30.8 ± 8.7 sr at 1064 nm. The lidar ratios are measured in the troposphere, whereas the assumptions of for the aerosol type "sulfate/other" in the CALIPSO v4 lidar ratio selection algorithm (50 ± 18 sr at 532 nm and 30 ± 18 sr at 1064 nm) is considered for the stratosphere (Kim et al., 2018). Nevertheless, the ground-based measurements confirm the assumptions about the lidar ratios of sulfate aerosol.

4.3 Increasing lidar ratio from 532 to 1064 nm

The observations in dust and smoke layers point to an increase of the lidar ratio at 1064 nm compared to the values at 532 nm, as shown in Fig. 7e,f. The values for smoke are provided in Table 4 and for dust in Table 5.

Mineral dust

The Raman lidar measurements point to an increase of the dust lidar ratio towards the wavelength of 1064 nm in plumes of Saharan dust (Fig. 7e). Haerig et al. (2022) observed two cases of Saharan dust at Leipzig, Germany, in February and March 2021. Gebauer et al. (2024) reported a case of Saharan dust at Mindelo, Cabo Verde, measured in September 2021 (Fig. 7e and Table 5). Previous studies based on a combination of the lidar backscatter coefficient at 1064 nm and the photometer AOD at 1020 nm point to an increase in the lidar ratio as well (Mamouri and Ansmann, 2017): Long-range-transported Saharan dust at Barbados showed a lidar ratio of 67 ± 15 sr at 1064 nm compared to 55 ± 8 sr at 532 nm. However, the combination of lidar and sun photometer used in Tesche et al. (2009) resulted in a spectral neutral behavior of the dust lidar ratio. 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. Furthermore, Omar et al. (2010) used airborne in-situ observations at Cabo Verde to constrain the CALIPSO extinction profile and provided dust lidar ratios of 39.4 ± 5.9 sr at 532 nm and 56.5 ± 16.5 sr at 1064 nm. These findings are reflected in the previous CALIPSO lidar ratios (v3 and before) of 40 ± 20 sr and 55 ± 17 sr at 532 and 1064 nm, respectively (Omar et al., 2009). In contrast, in version 4 a spectrally neutral lidar ratio of 44 ± 9 and 44 ± 13 sr is assumed at 532 and 1064 nm, respectively.



Table 5. The spectral lidar ratio and the ratio of lidar ratios for mineral dust and compared to CALIPSO v4 (values at 532 and 1064 nm, Kim et al., 2018) and DeLiAn (values at 355 and 532 nm Floutsi et al., 2023). The abbreviations for the references are given in Tab. 3.

Aerosol information	Lidar ratio (sr)			Ratio of lidar ratios		Reference
	355 nm	532 nm	1064 nm	532/355	1064/532	
Mineral dust						
Saharan dust, Leipzig	47 ± 8	50 ± 5	69 ± 14	1.06 ± 0.21	1.38 ± 0.31	Ha22
Saharan dust, Leipzig	49 ± 4	46 ± 5	57 ± 9	0.94 ± 0.13	1.24 ± 0.24	Ha22
Saharan dust, Cabo Verde	64.8 ± 10.2	50.9 ± 8.3	61.8 ± 8.6	0.79 ± 0.18	1.21 ± 0.26	Ge24
Saharan dust, Barbados	55 ± 8	55 ± 8	67 ± 15 ^a	1.00 ± 0.21	1.22 ± 0.33 ^a	MA17
Saharan dust, Morocco	52.9 ± 7.0	54.8 ± 6.7	54.9 ± 12.7 ^a	1.04 ± 0.19	1.00 ± 0.26 ^a	Te09
Saharan dust, Cabo Verde	–	39.4 ± 5.9	56.5 ± 16.5	–	1.43 ± 0.47	Om10
Dust (CALIPSO v4)	–	44 ± 9	44 ± 13	–	1.00 ± 0.36	Ki18
Saharan dust (DeLiAn)	53.5 ± 7.7	53.1 ± 7.9	–	0.99 ± 0.21	–	Fl23
Central Asian dust (DeLiAn)	43.4 ± 1.9	37.7 ± 2.1	–	0.87 ± 0.06	–	Ho20, Fl23
West Asian ^b dust (DeLiAn)	39.5 ± 6.0	37.4 ± 4.3	–	0.95 ± 0.20	–	Fl23

^a Estimated using lidar and photometer. ^b Also sometimes referred to Middle East. However, the "Middle East" is rather a political term whereas "West Asia" is a geographical term.

The Aerosol Robotic Network (AERONET, Holben et al., 1998) data base also includes lidar ratios for the wavelengths from 440 to 1020 nm. Müller et al. (2010) calculated lidar ratios for Saharan dust from AERONET observations, which showed a slight increase towards larger wavelengths. However, the lidar ratios at 440 nm were unrealistically high and not comparable to observations at 355 nor 532 nm. The study of Schuster et al. (2012) uses the dust size distributions and refractive index estimates from several AERONET stations around the desert regions to calculate regional dust lidar ratios, which show a slight spectral decrease from 532 to 1064 nm. However, newer studies based on AERONET data (version 3) point to a sharp decrease of the dust lidar ratio from 440 to 675 nm and then to a slight increase towards 1020 nm (Shin et al., 2018). They studied AERONET-retrieved lidar ratios for several deserts and found the highest lidar ratios for the Sahara. A similar spectral slope but with a more pronounced increase of the lidar ratio towards the near infrared was found by analyzing the AERONET measurements with the Generalized Retrieval of Aerosol and Surface Properties (GRASP, Torres et al., 2017; Dubovik et al., 2021) for 6 and 7 wavelengths, which includes the 1640 nm (Toledano et al., 2019; Haarig et al., 2022). However, all these results depend on the optical model to describe the irregular shape of mineral dust particles.

The modeling of dust lidar ratios (e.g., Gasteiger et al., 2011; Saito et al., 2021; Kong et al., 2022) is a challenging task because the lidar ratio depends on the size and shape distributions and the complex refractive index of the dust particles, which represents the mineralogical composition. Saito et al. (2021) predicted a slight increase of the lidar ratio towards 1064 nm, especially for the small dust particles. Gasteiger et al. (2011) modeled for case studies during the Saharan Mineral Dust Experiment (SAMUM) an U-shaped spectral slope of the lidar ratio with a minimum at 532 and 710 nm and increases towards 355



and 1064 nm.

The spectral slope of the lidar ratio of mineral dust contains information about its source regions. Because measurements at 1064 nm were not available yet, the discussion focuses on the spectral slope from 355 to 532 nm. Often, a spectrally neutral behavior was observed (Tesche et al., 2011; Groß et al., 2015), but sometimes the values in the UV were higher (Mattis et al., 2002; Bohlmann et al., 2018; Gebauer et al., 2024). Veselovskii et al. (2020) found both spectral behaviors in their observations at Dakar, Senegal. They could explain the two scenarios with differences in the imaginary part of the complex refractive index. In some source regions, the absorption, especially in the UV wavelength range, was found to be significantly higher compared to other regions (Di Biagio et al., 2019). Gómez Maqueo Anaya et al. (2024) tried to link these differences in the lidar ratio and the imaginary part of the refractive index to differences in the hematite content in the source region.

Compared to the lidar observations of western Saharan dust presented in Fig. 7e, the CALIPSO v4 algorithm does not capture the increase of the lidar ratio from 532 to 1064 nm. A spectrally neutral behavior is assumed (44 sr), whereas the Raman lidar observations point to an increase of lidar ratio by 21 – 38 %. The presented dust lidar ratios are solely for western Saharan dust. It is known that the lidar ratios at 355 and 532 nm are lower for Arabian dust (Filioglou et al., 2020), Western and Central Asian dust (Hofer et al., 2020) and East Asian dust (Murayama et al., 2004). In Tab. 5, we present Central and West Asian dust lidar ratios collected in the DeLiAn data base (Floutsi et al., 2023). A different mineralogical composition is the most plausible reason for differences in the observed lidar ratio (e.g., Schuster et al., 2012; Zhang et al., 2024; Gómez Maqueo Anaya et al., 2024). These differences might affect the values at 1064 nm and the respective spectral slope as well. There is the need for lidar ratio measurements at 1064 nm of dust from different source regions. The CALIPSO v4 lidar ratio selection algorithm (Kim et al., 2018) uses one lidar ratio for all dust cases and includes the natural variability in its uncertainties. Regional dust lidar ratios are under discussion for the CALIPSO v5 algorithm.

Smoke

The spectrally resolved lidar observations presented in Fig. 7f show a clear increase of the lidar ratio with wavelength. Haarig et al. (2018) observed smoke layers above Central Europe that originated from wildfires in western Canada, both in the troposphere and in the stratosphere. The Raman lidar measurements revealed almost the same lidar ratios in the troposphere and stratosphere (Tab. 4) for this event and explored for the first time the lidar ratio at 1064 nm of smoke. It is challenging to derive 1064 nm extinction in such thin smoke layers. There are some uncertainties involved, but nevertheless, the direction of the spectral slope is clear. Further evidence was provided by Ohneiser et al. (2020), who combined the lidar backscatter coefficient and the AERONET AOD to estimate a lidar ratio at 1064 nm for Australian smoke observed in the stratosphere above Punta Arenas, Chile. They found a lidar ratio of 120 ± 22 sr at 1064 nm, which is quite high. Nevertheless, the ratio of lidar ratios (1064-to-532 nm) is similar to the values observed by Haarig et al. (2018) and provided in Tab. 4.

An overview of lidar ratio measurements in atmospheric smoke layers is presented in the literature (Haarig et al., 2018; Adam et al., 2020; Ansmann et al., 2021). There seem to be two regimes of the spectral lidar ratio (355-to-532 nm) in smoke layers. Studies of aged boreal (Siberian, Canadian) smoke point to a clear increase from values of 30 – 50 sr at 355 nm towards 50 – 70 sr at 532 nm (Murayama et al., 2004; Müller et al., 2005; Ortiz-Amezcuca et al., 2017; Haarig et al., 2018; Hu et al., 2019). Higher lidar ratios but with the same spectral behavior were observed in the Arctic (Ohneiser et al., 2021). Other measurements



for European, Amazonian and fresh smoke report spectrally neutral lidar ratios (Veselovskii et al., 2015; Alados-Arboledas et al., 2011; Baars et al., 2012; Pereira et al., 2014). However, measurements of spectral decreasing lidar ratios are rare (Nicolaie et al., 2013). The DeLiAn collection results in a spectrally neutral lidar ratio (Floutsi et al., 2023). Janicka et al. (2023) present a comprehensive analysis of multi-year smoke observations (2013 – 2022) with a Polly^{XT} lidar at Warsaw, Poland, and separated it by source region. Smoke originated from western Europe (approximately 2 days of atmospheric transport) exhibited a spectrally neutral behavior with lidar ratios of 65 ± 17 sr, whereas smoke from eastern Europe (1 – 3 days of transport) showed a decrease with wavelength from 71 ± 14 sr at 355 nm to 59 ± 12 sr at 532 nm. Smoke that was advected over long distances from the Iberian Peninsula (5 – 7 days) or North America (4.5 – 8 days) exhibited the aforementioned spectral increase from 34 ± 10 sr and 37 ± 16 sr at 355 nm to 55 ± 14 sr and 58 ± 17 sr at 532 nm for Iberian and North American smoke, respectively. According to their observations, the lidar ratio at 532 nm varies less (55 to 65 sr) compared to the lidar ratio at 355 nm (34 to 71 sr).

The observations of Haarig et al. (2018) clearly belong to the category of increasing lidar ratios with wavelength, which seems to be a typical feature for long-range transported smoke, no matter whether in the troposphere or in the stratosphere. The spectral slope of the lidar ratio used in CALIPSO v4 is based on a comprehensive study of smoke-affected AERONET observations (Sayer et al., 2014). These authors found a monotonic decrease of the lidar ratio with wavelength from 355 to 532 and 1064 nm for all stations (close to source and in the transport regime) except the boreal near-source stations for which the values at 355 and 532 nm are almost the same and then the decrease occurred towards the 1064 nm. The spheroid model used for the AERONET retrievals is known to have problems in calculating the spectral slope of the backscattering properties at 180°. It could not reproduce the spectral increase from 355 to 532 nm, which was commonly found with lidar measurements for long-range transported smoke. Therefore, the persistent spectral decrease towards the wavelength of 1064 nm found for all AERONET sites seems questionable, now that direct measurements at this wavelength are available.

In conclusion, we may suggest that a future CALIPSO lidar ratio selection algorithm uses higher values at 1064 nm for "elevated smoke" and the stratospheric aerosol type "smoke". Whereas for aerosol layers below 2.5 km height, which are classified as "polluted continental/smoke", spectrally neutral values might be the best choice. Nevertheless, the spectral slope of the lidar ratio for smoke obviously contains information about the smoke characteristics, such as transport pathways, burning material or source region – a potential which is not fully exploited yet.

4.4 Extinction and backscatter related Ångström exponents

We want to support the discussion about the reasons for the observed spectral slope of the lidar ratio by looking at the spectral slope of the extinction and backscatter coefficient.

The spectral slope of the lidar ratio S between two wavelengths (λ_1 and λ_2) is a result of the spectral slope of the extinction coefficient α and backscatter coefficient β . Therefore, the Ångström exponent of the lidar ratio (SAE) can be expressed by the

extinction-related Ångström exponent (EAE) and the backscatter-related Ångström exponent (BAE) in the following way:

$$\begin{aligned}
 \text{SAE} &= \frac{\ln(S_{\lambda_1}/S_{\lambda_2})}{\ln(\lambda_2/\lambda_1)} \\
 &= \frac{\ln(\alpha_{\lambda_1}/\beta_{\lambda_1} * \beta_{\lambda_2}/\alpha_{\lambda_2})}{\ln(\lambda_2/\lambda_1)} \\
 &= \frac{\ln(\alpha_{\lambda_1}/\alpha_{\lambda_2}) + \ln(\beta_{\lambda_2}/\beta_{\lambda_1})}{\ln(\lambda_2/\lambda_1)} \\
 &= \text{EAE} - \text{BAE}.
 \end{aligned} \tag{3}$$

Table 6. The measured extinction-related Ångström exponent (EAE), backscatter-related Ångström exponent (BAE) and the lidar-ratio-related Ångström exponent (SAE) for the wavelength range 532 to 1064 nm from Raman lidar observations.

Aerosol type	EAE	BAE	SAE	Reference
Marine aerosol	0.08 ± 1.11	0.04 ± 0.30	0.04±1.21	this study
Ice clouds	0.08 ± 0.26	0.21 ± 0.31	−0.08±0.28	Ha16
Continental aerosol	0.97 ± 0.32	1.00 ± 0.31	−0.02±0.44	this study
Sulfate aerosol ^a	1.53±0.26	0.54±0.31	0.97±0.46	Ge24
	−0.08±0.21	0.35±0.26	−0.47±0.33	Ha22
Saharan dust	0.01±0.08	0.35±0.26	−0.31±0.28	Ha22
	−0.06±0.53	0.13±0.31	−0.28±0.31	Ge24
Smoke, troposphere	0.6 ± 0.3	0.8 ± 0.3	−0.29±0.40	Ha18
Smoke, stratosphere	0.85 ± 0.3	1.2 ± 0.6	−0.48±0.50	Ha18

^a Mixed with marine aerosol.

In Table 6, we report the corresponding Ångström exponents (only shown for the 532 to 1064 nm wavelength range) for the discussed Raman lidar observations. The presented results follow equation 3 within the uncertainties. Deviations from the exact agreement can be explained by different vertical smoothing lengths. The backscatter coefficients and the corresponding BAE need less vertical smoothing compared to the extinction coefficient and subsequently the lidar ratio.

The positive SAE of sulfate aerosol indicates the decrease of the lidar ratio with increasing wavelength. For marine aerosol, continental aerosol and ice clouds, the SAE is close to zero ($-0.1 < \text{SAE} < 0.1$), which indicates the spectrally neutral behavior. Whereas, for dust and smoke, the SAE is negative (< -0.25), which indicates the spectrally increasing behavior.

The EAE is close to zero ($-0.1 < \text{EAE} < 0.1$) for large aerosol particles (such as mineral dust and marine aerosol) and ice clouds. Smoke particles show an EAE of 0.6 to 0.85 and the even smaller continental and sulfate aerosol particles exhibit EAE > 0.95 .

The BAE is always above zero, which indicates that the backscatter coefficient always decreases from 532 to 1064 nm. The strongest decrease is observed for smoke and continental aerosol ($\text{BAE} > 0.8$), which exhibit a pronounced wavelength dependence of the backscatter coefficient. In the case of sulfate-dominated aerosol, a mixture of marine and sulfate was observed,



which probably decreased the BAE compared to pure sulfate aerosol. The BAE is smallest but still positive for large, non-absorbing particles such as marine aerosol and ice crystals. We especially want to highlight the high BAE in the 532 to 1064 nm wavelength range for the long-range transported Saharan dust observed at Leipzig (Haarig et al., 2022). The BAE from 355 to 532 nm for the same observations is close to zero or even negative.

440

5 Conclusions

Thanks to the rotational Raman technique at 1064 nm Haarig et al. (2016), first direct measurements of the lidar ratio at 1064 nm have been conducted since 2016. In this work, we reviewed the existing observations to provide an overview lidar ratio values at 1064 nm but also at 355 and 532 nm. The existing triple-wavelength lidar ratio observations at 355, 532 and 1064 nm were discussed with respect to five aerosol types (mineral dust, smoke, sulfate, marine and continental aerosol) and ice crystals in cirrus clouds and its spectral behavior has been investigated. Previous studies (e.g., Müller et al., 2007; Floutsi et al., 2023) could only focus on the widespread observations of the lidar ratio at 355 and 532 nm. Now, we were able to expand the wavelength range from 355 to 1064 nm and to discuss the findings with respect to the spectral behavior of the lidar ratio for different aerosol types. We could identify the following key results:

- 450 – For marine aerosol and ice crystals in cirrus clouds, a spectrally neutral behavior of the lidar ratio was found. Here, the spectrally neutral behavior was present for the entire observed wavelength range (355 to 1064 nm).
- For continental aerosol, a spectrally neutral behavior of the lidar ratio was found as well. The lidar ratio of continental aerosol varies with RH. The dependence of lidar ratio on RH is stronger at longer wavelengths.
- For sulfate aerosol, the lidar ratio decreases with wavelength. In the presented case study of Gebauer et al. (2024), the spectral decrease is more pronounced towards 1064 nm, whereas the lidar ratios at 355 and 532 nm are almost the same with only a slight decrease with wavelength.
- 455 – For mineral dust, an increase of the lidar ratio was found in the wavelength range from 532 to 1064 nm for source regions in the western Sahara. The spectral slope from 355 to 532 nm is neutral or decreasing according to the mineralogical composition in the source region. Differences in the spectral behavior of the imaginary part of the refractive index (absorption) are most probably the reason (Veselovskii et al., 2020).
- 460 – For wildfire smoke, an increasing lidar ratio with wavelength is a typical feature for long-range transported smoke, which extends to 1064 nm as well (Haarig et al., 2018). However, for fresh smoke and medium-range transported smoke (approximately < 3 days), a spectral neutral or rarely decreasing behavior has been observed for the wavelength range 355 to 532 nm (e.g., Janicka et al., 2023).

465 The spectral slope of the lidar ratio from 532 to 1064 nm was compared to the CALIPSO v4 aerosol typing and lidar ratio selection algorithm (Kim et al., 2018). Good agreement was found for the aerosol type of "clean marine". Also for the strato-



spheric aerosol type "sulfate/other", good agreement was found with the tropospheric sulfate observations. In the case of "clean continental", the Raman lidar observations show a spectrally neutral behavior under humid conditions (RH between 70 and 80%), whereas CALIPSO v4 assumes lower values at 1064 nm compared to 532 nm. The study of hygroscopic growth might indicate that this is valid under dry conditions. In the case of mineral dust, the Raman lidar measurements point to a slight increase of the lidar ratio towards 1064 nm while CALIPSO v4 assumes a spectral neutral lidar ratio. Within the uncertainties, these values still agree. The strongest discrepancy was detected for elevated smoke and stratospheric smoke. For these aerosol types, CALIPSO v4 assumes a strong decrease with wavelength ($S_{1064}/S_{532} = 0.43 \pm 0.28$) whereas the Raman lidar observations point to an increase with wavelength. We discussed the variety of spectral lidar ratios for smoke and came to the conclusion that for long-range transported smoke, the spectral increase is a typical feature. Elevated smoke and 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 these aerosol types for a CALIPSO v5 algorithm.

Data availability. The Polly^{XT} data are available at <https://polly.tropos.de/> (PollyNET, 2024). The manual analysis of the extinction coefficient will be made available via zenodo.

Appendix A: Corrections and limitations of the rotational Raman lidar method at 1064 nm

The rotational Raman lidar technique was shortly introduced in Section 2. Here, we firstly describe the applied spectral cross talk and temperature correction and then discuss the limitations of the technique.

A1 Corrections

Additionally to the vibrational-rotational Raman lidar analysis, two corrections were applied to the rotational Raman measurement at 1058 nm: a correction for the spectral cross-talk introduced by the elastically backscattered light at 1064.14 nm and a correction due to the temperature dependence of the intensity of the rotational Raman lines (e.g., Behrendt and Nakamura, 2002; Siomos et al., 2024). The temperature dependence of the vibrational-rotational Raman signals at 387 and 607 nm is weaker and a spectral cross-talk correction has not be considered because the suppression of the elastic backscattered light is easier to achieve due to the larger spectral distance. In our setup, the spectral cross-talk correction clearly dominates over the temperature correction in terms of magnitude for the rotational Raman measurements.

Cross-talk correction

The spectral cross-talk correction was already outlined in Haarig et al. (2022) and is discussed here for the Polly^{XT} at Cabo Verde. A persistent deck of altocumulus clouds, which frequently occur in the upper part of the Saharan Air Layer between approximately 4.0 – 4.5 km altitude, are best suited for the spectral cross-talk correction. The vibrational Raman signals at 387 and 607 nm, which are not influenced by any spectral cross talk, show a pronounced decrease in the cloud layer. However, the



rotational Raman signal at 1058 nm is influenced by the high elastic return signal at 1064 nm from the cloud droplets. The influence of the elastic signal has to be subtracted from the rotational Raman signal. The empirical cross-talk correction factor c_S is chosen in the way that the influence of the elastic signal is removed and the rotational Raman signal decays in a similar way as the vibrational Raman signals. The spectral cross-talk correction is applied to the range-corrected Raman signal of P_{Ra} using the elastic range-corrected signal P_{el} at 1064 nm in the following way:

$$P_{Ra, S-cor} = P_{Ra} - c_S * P_{el}. \quad (A1)$$

The spectral cross-talk depends not only on the interference filters, but also on the sensitivity of the elastic channel, which depends on the applied neutral density filters. Therefore, a new spectral cross-talk correction constant has to be determined if the neutral density filters in the elastic channel are changed. For the Polly^{XT} on Cabo Verde, c_S was 0.00067 ± 0.00003 between 10 September 2021 and 28 January 2022 (see sulfate aerosol case in Gebauer et al., 2024), afterwards c_S was 0.00014 ± 0.00004 until 23 March 2023 (see marine aerosol case in Sect. 3.1). During the first time period, the neutral density filters in the elastic channel were equivalent to an optical thickness of 1.3; in the second period, the neutral density filters were reduced to 1.0. The spectral cross-talk correction is applied before the temperature correction.

510 Temperature correction

The temperature dependence of the transmitted intensity through the two interference filters is shown in Fig. A1. It was calculated from the temperature dependent intensity of the rotational Raman lines (Adam, 2009; Siomos et al., 2024) multiplied with the transmittance curves of both filters (Fig. 1). The same filters were used first in BERTHA and later in the Polly^{XT} at Cabo Verde. The correction of the temperature-dependent transmission of the range-corrected rotational Raman signal P_{Ra} is performed with respect to a temperature T_0 of 288.15 K using the a polynomial fit of the 3rd order:

$$P_{Ra, T-cor}(T) = \frac{P_{Ra, S-cor}}{1 + a_1(T - T_0) + a_2(T - T_0)^2 + a_3(T - T_0)^3}, \quad (A2)$$

where the fit coefficients are $a_1 = -(1.6047 \pm 0.0036) \times 10^{-3}$, $a_2 = -(2.3179 \pm 0.06449) \times 10^{-6}$ and $a_3 = (3.6184 \pm 0.0750) \times 10^{-8}$. A detailed discussion about the temperature dependence of the rotational Raman lines can be found in Behrendt and Nakamura (2002), Veselovskii et al. (2015) and Wang et al. (2024a).

520 A2 Limitations

In this subsection, we discuss the limitations of the rotational Raman lidar technique to its application in the near infrared wavelength range, which currently prevents an automatic analysis. Therefore, we can base our present study on case studies only.

- The molecular signal at 1064 nm is 81 times lower compared to observations at 355 nm, because the scattering cross-section for Rayleigh scattering scales with λ^{-4} (λ – wavelength). Therefore, long averaging periods (> 2 hours) are necessary. Especially the lower energy of the Polly^{XT} laser at 1064 nm (~ 80 mJ at 100 Hz) compared to BERTHA

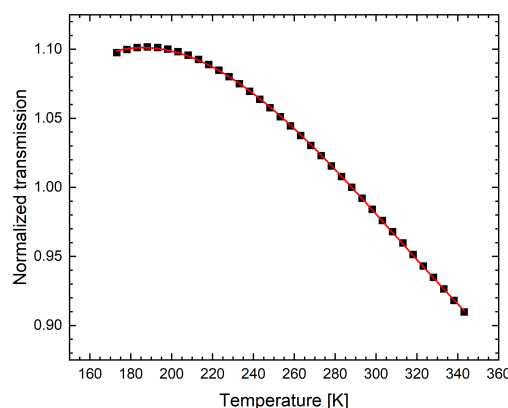


Figure A1. The temperature dependent transmission of the rotational Raman lines when applying both interference filters simultaneously. The transmission is normalized to a temperature of $T_0 = 288.15$ K. The polynomial fit curve to the data points is used to correct the rotational Raman signal for the temperature effect.

(nominal up to 1000 mJ (30 Hz) at 1064 nm, Haarig et al., 2016) together with the smaller telescope (30 cm mirror compared to 53 cm mirror in BERTHA) limits useful signal detection to 4.5 km height on Cabo Verde, whereas strong BERTHA signals allowed us to compute even the extinction coefficient of a stratospheric smoke layer at around 15 km height at Leipzig (Haarig et al., 2018).

- We use photomultiplier tubes (PMTs, R3236 from Hamamatsu Photonics, Japan) to detect the signals in the near infrared to cover several orders of magnitude in the backscattered signal. The single photon quantum efficiency of the PMTs is low (0.08%). Longer temporal and vertical averaging is necessary for our PMTs because of the reduced quantum efficiency. Furthermore, the detectors are aging, which further decreases their sensitivity. Wang et al. (2024b) used silicon single photon avalanche diodes (Excelitas Technologies Corp., Canada) with a quantum efficiency of 2.5%, which is more than 30 times higher. Analog detection with avalanche photodiodes provides an even higher quantum efficiency of around 40%. However, the analog detection has difficulties to accurately detect the signal as oscillations may occur which significantly affect the extinction retrieval. Nevertheless, due to the optical design of Polly^{XT} it is not possible to apply these method currently, but may be an option for future development.
- The large filter width (9 nm) limits the observations to night-time hours only. Vibrational-rotational Raman measurements are as well limited to night-time, whereas rotational Raman observations at shorter wavelengths provide daytime extinction observations as well (e.g., Arshinov et al., 2005). Due to the reasons discussed above, however, its not applicable at 1064 nm yet.



545 – The calibration of the backscatter coefficient at 1064 nm is not trivial (e.g., Vaughan et al., 2009), because of the low
 molecular signal at 1064 nm. The photon counting detection implemented in BERTHA and Polly^{XT} is most sensitive
 to the low molecular signal at 1064 nm. A slight variation in the backscatter coefficient affects the lidar ratio. A cirrus
 cloud helps to constrain the backscatter coefficient by comparison to the observations at 532 and 355 nm and assuming
 a color ratio of 1 (Vaughan et al., 2010, 2019).

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Author contributions. MH analyzed the data at 1064 nm, conceptualized the study and wrote the manuscript. RE conceptualized the rota-
 tional Raman measurements and together with DA strongly supported the measurements with BERTHA and Polly^{XT}. BG performed the
 calculations for the temperature correction. HB supported the analysis of the Polly^{XT} data and AA supported the discussion of the results.
 All co-authors provided valuable input to the manuscript.

555 *Competing interests.* The authors declare that they don't have competing interests.

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 565 lidar beams at Leipzig (Fig. 3).



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