***Summary:** The authors report on measurements of biomass burning aerosol (BBA) layers in the free troposphere and in the UTLS region in the summer of 2023 using two multiparameter lidars at two different sites. Emphasis is put on the fluorescence properties of the BBA which are measured with, respectively, one and five discrete fluorescence detection channels. Firstly, a short description of the instruments is given and the calibration procedure for the fluorescence signals is described. Importantly, to make measurements with different fluorescence lidars more comparable, the spectral fluorescence backscatter coefficient and capacity are used as parameters. Secondly, four BBA cases are discussed in detail. Thirdly, a compilation of BBA measurements at the two sites performed between May and September 2023 is presented and analyzed. One topic is the comparison of spectral fluorescence backscatter capacities, supplemented by data obtained at two other measurement sites. Relations between various elastic and inelastic aerosol properties, height, and relative humidity are also investigated.*

***General comments:** The manuscript provides interesting results. In particular, the comparison of fluorescence measurements with two or even four instruments highlights the potential for future comparative research. Ideally, BBA plumes that pass consecutively over several lidar stations (known as lidar match) should then be investigated. Other findings of the study corroborate the conclusions drawn in previous publications, for example the redshift of the BBA fluorescence spectrum with height. The manuscript is well written. After corrections, the paper can be published.*

Specific comments:

l.44: The potential of fluorescence measurements for studying aerosol-cloud interaction was first identified by Reichardt (2014). A citation seems appropriate here.

l.46: The reference list should also include as landmark publications Reichardt et al. (2018) [Reichardt, J., Leinweber, R., and Schwebe, A.: Fluorescing aerosols and clouds: investigations of co-existence, EPJ Web Conf., 176, 05010, https://doi.org/10.1051/epjconf/201817605010, 20189] and Reichardt et al. (2025).

Reply: The two references are now added to the revised manuscript.

l.51: 'as a proxy to aerosol chemical composition.' This is a bold claim. Because the fluorescence spectra of atmospheric aerosols are spectrally broad and without notable features, it is rather difficult to draw conclusions about their chemical composition. However, it does appear possible to distinguish between different aerosol classes. Perhaps the wording should be a little more cautious.

Reply: We agree that fluorescence spectra of atmospheric aerosols are generally broad and lack distinct spectral features, which makes it difficult to infer detailed chemical composition directly from fluorescence measurements. Our intention was to explain that fluorescence signals can provide new information related to aerosol types or classes, because it is more directly related their chemical composition. To avoid overinterpretation, we have revised the sentence to adopt more cautious wording.

"Therefore, the LIF lidar can provide a new dimension of information to aerosol characterization, as the fluorescence signatures (i.e., the fluorescence capacity and spectrum) provide a link to aerosol chemical composition."

l.109: The text 'in this study, we redefine… where…' is quite confusing. There is no redefinition of old but a definition of new parameters. Please, consider writing something like 'we use in this study the spectral fluorescence backscatter coefficient, B_F,lambda, which is the ratio of the spectrally integrated fluorescence backscatter coefficient, b_F,lambda, and the full-width-half-maximum (FWHM) of the interference filters, DD, in the fluorescence channels:

Equation B_F,lambda.

Furthermore, the spectral fluorescence capacity, G_F,lambda, will be used:

Equation G_F_lambda.'

Unfortunately, the notation is a bit different from the one used in Reichardt et al. (2023) and quite confusing because the meaning of 'b' and 'B' are switched around. The authors should consider mentioning this problem.

Reply: Thanks for pointing out the confusion in terminology. Historically it was more natural to use β_F to represent spectrally integrated fluorescence backscatter coefficient, and then we defined B_F to spectrally differentiated fluorescence . Now, as our data processing algorithm evolves, it is more convenient and appropriate to use β_F as the spectral fluorescence backscatter, consistent with Reichardt et al. (2023). The text and equation (1), (2), (3) have been adapted accordingly.

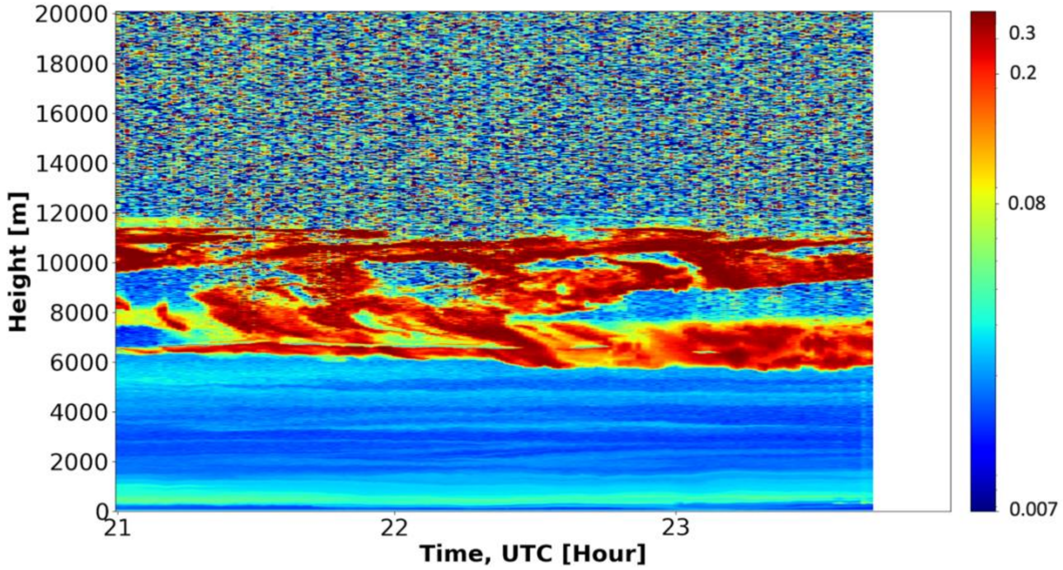
l.128: 'The spectral…' See comment on l.51.

Reply: To avoid overinterpretation, the sentence has been changed to "The fluorescence capacity is influenced by the presence of fluorescent compounds within the particles and reflects their ability to emit fluorescence signals when exposed to radiation."

l.146: 'thick mixed… cirrus clouds…'. How do the authors know? You would need at least particle depolarization ratio and lidar ratio for cloud typing, you cannot tell from the current Fig. 2. By the way, is the LILAS lidar tilted? If not, Figure 3 could, for example, reveal a cirrus layer with horizontally oriented particles at 6.7 km which is embedded in a broader ice cloud with randomly oriented particles.

Reply: 1. LILAS is tilted by 4 degrees to the zenith, so the low depolarization ratio at 6.7 km cannot be explained by horizontally oriented ice particles. 2. We prefer to show the quicklook the volume depolarization ratio in publications since it shows better the tempral and vertical variation of aerosol and cloud signatures, comared with particle depolarization ratio which requires stronger smoothing and/or time averaging in order to surpress the noise or extreme values resulted from the calculation. Ideally, one would need particle linear depolarization ratio to distinguish mixed phase cloud and ice cloud. However, in real aerosol lidar observation, particle depolarization ratio is not always accessible, because signal saturation in cloud layers. Volume depolarization ratio allows a qualitative diagnostic of mixed phase cloud if low or intermediate volume depolarization ratios are observed in cloud layers. We think the cloud layers in Figure 3 would better be described as <mixed-

phase clouds and cirrus clouds>, instead of pure ice-clouds, because volume depolarization ratios lower than 0.1 were detected. The orignal Figure 3(c) does not allow to clearly see the intermediate values of depolarization, so in the revised manuscript, this figure is replaced by the figure below, which shows the volume depolarization in logarithmic scale.



l. 164: 'a sharp RH increase above 5000 m, … water uptake.' The apparent rise in RH is probably caused to some degree by fluorescence interference as it coincides with an increase in the spectral fluorescence backscatter coefficient. See comment on l.182.

and

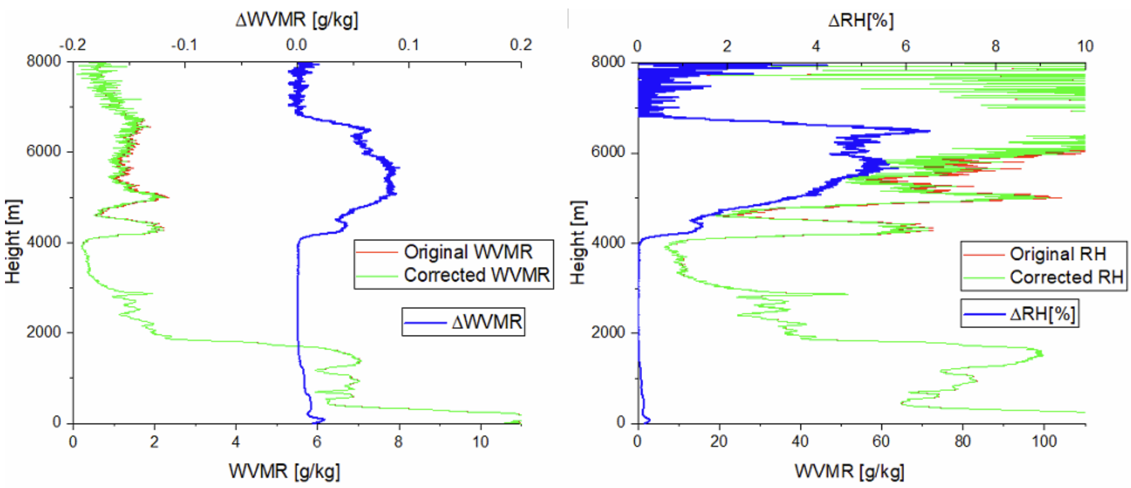
l.182: 'It is worth noting… lidar-derived humidity measurements.' The issue of RH measurements in the presence of fluorescing aerosols must be discussed in more detail. Even with a bandwidth of 0.3 nm the LILAS instrument is very well affected by fluorescence, as Reichardt et al. (2023) have shown; and the comparison between ERA-5 and LILAS RH profiles in Fig. 3 supports this assessment. Above the base of the aerosol layer, lidar RH is consistently higher than the model data and follows roughly the 466-nm spectral fluorescence backscatter coefficient. Unfortunately, the mixing ratio (MR) profile is not given, but probably its values are low within the aerosol layer, and so only a relatively small fluorescence contribution to the measured water vapor Raman signal makes a big difference in RH. The authors might try a 0-order correction of their MR, and then RH, profile: Reichardt et al. (2023) find for a detector bandwidth of 0.22 nm a fluorescence-induced effect on MR of roughly 0.017 g/kg per mean spectral fluorescence backscatter coefficient between 430 to 450 nm (in Tm^-1sr-1nm-1). A spectral fluorescence backscatter coefficient of 6 Tm^-1sr-1nm-1 would therefore result in an MR error of approx. +0.1 g/kg. The broader bandwidth of LILAS would exacerbate the effect, on the other hand it measures fluorescence at a slightly longer wavelength, which would partially compensate for this.

Reply: We are aware that the fluorescence signal influence the measurement accuracy of water vapor mixing ratio (WVMR) and this effect cannot be correct if the lidar system has only a single fluorescence channel. However, this influence is not the dominant factor for the variation of water vapor mixing ratio or the RH. Firstly, the filter of interference filter is very narrow, 0.3 nm, which is about 1/146 of the width of the fluorescence filter.

A quick estimation is done in order to estimate the contribution of fluorescence signal to the water vapor measurements, i.e., WVMR [g/kg] and relative humidity (RH) [%].

Assume the spectral fluorescence backscatter at 408 nm is as strong as at 466 nm (actually it is with a factor of 0.5-0.8, but we use 1.0 as a conservative value), and $\frac{1}{146}$ of the fluorescence goes into the water vapor channel. The following figures show the original water vapor mixing ratio and the corrected one, as well as the relative humidity. One can see the maximum difference of WVMR is about 0.1g/kg in the smoke layer, which result in a difference of 5-6% in the RH.

After correction, the sharp increases of RH at 4200 at 5000 m are still presented, which demonstrates clearly that the influence of fluorescence on water vapor measurements is not significant and the increase of RH was real.



l.174: 'The absence of fluorescence quenching…' This is a question difficult to answer with a single fluorescence detection channel instrument. First, one must rule out that the air masses within the cloud have a different origin than those outside, second, one has to show that the aerosol and cloud particles not merely coexist but actually interact.

Reply: Thanks for the insightful and constructive comment. We fully agree that a single fluorescence channel has inherent limitations in accessing fluorescence quenching. And we cannot identify whether the smoke and cloud particles coexisted or interacted. The original sentence has been modified to soften the claim and explicitly acknowledge these limitations.

"The fluorescence quenching….(Reichardt et al., 2025; Gast et al., 2025). In Case 1, despite the sharp increase in RH to 90 %–100 % near the cloud base, the fluorescence capacity showed only minor variations (Figure 3b). This observation contrasts with other events during the 2023 fire season, where quenching was clearly detected using a five-channel fluorescence lidar (Veselovskii et al., 2024a). However, with only a single broadband fluorescence channel at ATOLL, we cannot identify the spectra of the aerosols at low and high humidities, to check if they are from the same origin. And lidar observation cannot prove if aerosol and cloud particles interact or they only spatially coexist. Consequently, the ”

l.251: ‘during the … in 2023’. The redshift has been consistently observed over the years, it thus seems to be a general feature of transported BBA.

Reply: Thanks for adding coroboerating information.

l.271: ‘moderate volume linear depolarization ratio.’ The reviewer does not understand why volume depolarization ratio is used in the discussion. It is not a true particle property. Please, consider using particle depolarization ratio instead. Further, are lidar ratios available? Ice particle formation should reveal itself by a drop in lidar ratio.

Reply: In the analysis of lidar observations from the GPI lidar, the volume depolarization ratio is used as a qualitative indicator of particle shape. However, it is not suitable for quantitative interpretation due to the insufficient polarization purity of the laser emission. The presence of ice crystals can be inferred from an increase in both the volume depolarization ratio and the elastic backscatter signal, in contrast to the absence of, or only weak and steady, fluorescence backscatter.
The lidar ratio at 10,000 m is not available, as the system was tilted at 45°, leading to noisy signals at such altitudes.

l.334: ‘In contrast to EAE, … refractive indices.’ This is not correct. EAE is affected by wavelength dependent absorption. Please, reword.

Reply: We thank the reviewer for this valuable comment. We agree that the original wording was misleading and not rigorous, as the EAE is indeed influenced by wavelength-dependent absorption (imaginary part of the refractive index).
Our intention was to emphasize that the BAE exhibits a stronger sensitivity to the imaginary part of the refractive index than the EAE does, while both parameters are also affected by particle shape. The sentence has been changed as follows:
“ In contrast to the EAE, the BAE of aerosol particles shows a stronger dependence on the imaginary part of the refractive index (i.e., wavelength-dependent absorption), in addition to the influence of particle morphology.”

l.351: ‘High … morphology,…’ It also depends on the size parameter!

Reply: Thanks to the reviewer for the precise remark. We are aware that the linear depolarization ratio depends on both the particle morphology (size and shape) and the size parameter. In the original wording, we used ‘morphology’ in a broader sense, implicitly including the size effects (which was the subject of the next paragraph, starting from L354). We fully agree that this formulation could be misinterpreted due to its impricision. Therefore, the sentence was now revised:

High depolarization ratios are usually associated with particle morphology, i.e., their size and irregular shape. In the UTLS, aged BBAs particles were detected with thick organic coating, which may appear semi-solid and glassy in cold and dry conditions in UTLS, thereby enhancing their depolarization ratio.

l.351: 'semi-solid and glassy…' Can the authors provide a reference?

Reply: The reference has been added.

Viscosity and Phase State of Wildfire Smoke Particles in the Stratosphere from Pyrocumulonimbus Events: An Initial Assessment. Mei Fei Zeng, Andreas Zuend, Nealan G. A. Gerrebos, Pengfei Yu, Gregory P. Schill, Daniel M. Murphy, and Allan K. Bertram
Environmental Science & Technology 2025 59 (16), 8037-8047
DOI: 10.1021/acs.est.4c10597

l.381: 'This decrease… BBA layer.' This is very speculative! Unless it is unequivocally shown that the air masses carrying the aerosols and the clouds have the same origin, it can also be a transport phenomenon. Please, argue more cautiously.

Reply: Thanks for pointing out that our interpretation was too speculative. In this case (and some other cases in 2023), back trajectory analysi is only sufficient to identify the major fire sources, not to explain the fine-scale difference in observed aerosol optical properties. Because the smoke plumes were distrbuted over large areas, the exact age and atmospheric processing pathways of individual air mass are difficult to determine. We agree that the argument -- cloud processing, mentioned in the manuscript is just one explanation and the other possibilities should not be excluded. To address this, we have added the following sentence after the original statement:
Nevertheless, other explanations cannot be excluded, such as, different smoke plumes arriving at ATOLL and TROPOS (i.e., originating from the same fire complex but not the same fire, or having experienced different aging conditions) or changes in smoke properties during transport between ATOLL and TROPOS.

l.397: 'This consistency… Figure 12.' The reviewer would prefer a more cautious wording. The two wavelengths are close to the point in the spectrum of least variability (Figs. 7(e) and 9(e)) and therefore not well suited for assessing the overall consistency.

Reply: Thanks for pointing out the need of cautious wording. According to the fluorescence spectra measured by lidar, as shown in Figure 13 and 14, the variability of smoke fluorescence capacity at 466 or 472 nm is less significant at wavelength > 500 nm, yet some differences still exist within/between tropospheric and UTLS smoke particles, as shown in Figure 14. A more robust assessment of geographic variability would therefore require a more comprehensive parameters, such as the full fluorescence spectrum.
Consequently, we have removed the sentence on line 397 that referred to "consistency" between the ATOLL and GPI sites in Figure 12. This change does not affect the overall message of the paragraph.

l.410: ‘rather a recurring feature of BBAs (Reichardt et al., 2025) and …’

Reply: The word ‘rather’ is removed from the sentence, since it is slightly informal.

l.454: ‘The … were used’. Comment: It is quite intriguing that BBA events at both locations go mostly hand in hand with (very) dry air. So it is no wonder that hygroscopic growth is not readily observed.

Reply: We agree that the strong association between the detected BBA layers and dry air masses (mostly RH < 50 %) provides a clear explanation for the lack of observable hygroscopic growth throughout our dataset. However, this statistical correlation alone does not constitute direct evidence of the inherent non-hygroscopicity of aged smoke particles. The most direct evidence comes from Case 1 (14 May 2023 at ATOLL), where BBA particles were embedded in a layer with RH increasing sharply to 90 %–100 % near the cloud base, yet both the particle linear depolarization ratio and the fluorescence capacity remained almost unchanged. More observational cases under high-RH conditions are clearly needed to corroborate this result.

Equations:

All: Sub- or superscripts that are not variables must not be in italic.

Reply: Thanks to the reviwer for careful revision. This has been modified in the revised manuscript.

Figures:

Reply: Figures are modified according to the suggestion of the reviewer.

2, 4: + Axis titles are not centered; increase font size for better readability

+ Show the calibrated fluorescence backscatter coefficient in panel (b)

+ Show particle depolarization ration in panel (c) for easier interpretation

+ Caption: Unit must not be in italic

+ Caption: Provide integration time per profile; was a sliding average applied?

+ Caption: Provide information on the tilt angle of the lidar

3: + Correct legend of panel (e)

+ It would be helpful to see the temperature profile along with RH

+ Caption: Provide vertical resolution

5: + It would be helpful to see the temperature profile along with RH

+ Caption: Provide vertical resolution

6, 8: + Homogenize the styles for Figs. 2, 4, 6, and 8

+ Axis titles have different font sizes

+ Show particle depolarization ratio in panel (c) for easier interpretation

+ Caption: Provide information on the tilt angle of the lidar

11: + Have the legends the same font size?

12: + Caption: 'Lindenberg'

+ Caption: 'Reichardt et al. (2025), respectively.'

13: + Caption: 'at 472 nm…'

+ Caption: 'fluorescence capacity/backscatter' (?)

+ Caption: Please, indicate symbols for CR

A1: + DM must be rotated 90° clockwise

B1: + The blue trajectories are difficult to see in the maps.

+ Caption: 'airmass passed through.'

B2: + The dotted orange lines are difficult to see in the maps.

Tables:

C1: + Header: 'calculated as SFC_2 to SFC_1.'

Reply: Thanks for the careful revision and for pointing out this typo. It has been modified.

Text:

l.87: 'The receiver…'

l.89: '(vibrational …'

l.94: The symbols used for the spectral fluorescence properties should be given

l.98: How about using '/' for the ratio?

l.103: 'channel; and…'

l.103: '(see …'

l.120: 'optical receiver…'

l.155: 'nm^-1 and independent of height, which …'

l.164: '(e.g., Navas-…'

l.179: 'and variability of organic…'

l.197: 'Giant and …' is not a sentence

l.200: 'Figure 4(b) and (c).' (?)

l.214: 'adding more evidence…'

l.240: 'The spectra of fluorescence capacity are…'

l.267: Meaning not clear, please, change wording

l.268: 'from 4000 to…' (?)

l.277: 'coefficients, and the extinction…'

l.341: But lidar ratio does increase with wavelength, so it is correlated, isn't it?

l.346: The shorter wavelengths are missing

l.363: 'with a … downward motion…', meaning not clear, please, change wording

l.425: 'three intensive parameters…'

l.450: 'and the small number of collocated and …' (there are some)

l.555: 'by the gain of …'

l.559: How about using '/' for the ratio? (here and in the remainder of the Appendix)

l.559: 'of optics. In the…'

References:

Veselovskii et al. (2024b) and Veselovskii et al. (2025) refer to the same publication, discard the former.

Reply: The text of the manuscript has been corrected accordingly. We thank the reviewer for the precision, patience and professionalism.