Dear Referee 1!

We thank you for careful reading of the manuscript and the thoughtful questions, comments and suggestions. We have addressed the comments in this reply letter and in the revised version of the manuscript. Most changes are aimed to improve the clarity and presentation of the manuscript.

A version of the revised manuscript with tracked changes is attached to this reply letter. All changes in the revised manuscript are given in BLUE. The old passages that were removed are marked in RED.

In this reply letter, the comments of Reviewer 1 are given in BLACK, our answers are given in GREEN.

General comment

This paper demonstrates the detection capability of fluorescence lidar in revealing and identifying high-altitude smoke layers, based on the measurements collected during the Canadian wildfire season in 2023. These layers, due to their low particle concentrations and high altitudes, may be beyond the detection limits of conventional Mie-Raman lidar systems, yet they still influence the ice cloud formation and cloud properties. The authors first introduce the calibration method of the fluorescence channel, followed by an analysis of several scenarios of measurements. These results demonstrate that the fluorescence channel enhances the detection capability of a conventional Mie-Raman lidar, brings new information about aerosol characterization and provides new opportunities for the study of aerosol-cloud interactions. The study aligns well with the scope of <Atmospheric Chemistry and Physics> and the measurements presented are valuable. The presentation of data is clear and the conclusions are scientifically sound. Therefore, I recommend the paper for acceptance. However, major revisions regarding the following aspects are needed:

1. In the calibration procedure, the calculation of detector efficiency contains two terms: the PMT quantum efficiency and the PMT gain ratio. According the text in line 148-153, the quantum efficiency is determined using the data provide by Hamamatsu and the PMT gain ratio is derived by swapping the detectors in the nitrogen Raman and fluorescence channels. However, the description about how this experiment was conducted remains unclear. Please clarify and verify whether the PMT gain ratio calculation has already accounted for the ratio of quantum efficiency.

➔ The experiment was conducted as follows: For better clarity we'll name the PMT used in the nitrogen Raman channel as "PMT_R" and the PMT in the fluorescence channel as "PMT_F". First, we measured 5 minutes with the standard configuration (PMT_R in Raman channel and PMT_F in fluorescence channel). Secondly, PMT_R and PMT_F were swapped and again, 5 minutes of measurement were taken. Thirdly, the PMTs were returned to their initial position in the standard configuration. Another 5 minutes of measurement were taken to ensure that the atmospheric conditions stayed rather stable during the time period of the experiment, so that the signals of the consecutive 5-minute periods are comparable. Now, the temporal average of the signals in both channels were calculated for the standard configuration (P_387_0, P_466_0) and the swapped configuration (P_387_swapped, P_466_swapped). For each channel, the ratio of the signals for both configurations was calculated, i.e., P_387_0/P_387_swapped and P_466_swapped/P_466_0. These ratios account for electrical gain only, because each signal ratio is evaluated at a certain spectral range (=

the filter bandwidth). The mean of both ratios was used as PMT gain ratio for fluorescence backscatter calculations.

The ratio of quantum efficiency can't be obtained with this experiment because an absolute reference for the incident signal intensity in the respective detection channel is missing. To obtain the quantum efficiency experimentally, a calibrated lamp with a known spectrum would have to be used. In fact, with such an approach the whole ratio of the channel optical efficiencies could be determined all at once.

2. The writing needs improvements, as some sentence and expressions lack of the precision expected in scientific papers. The paper contains many colloquial phrases that should be avoided to ensure the writing is concise and accurate. Please refer to specific comment for details.

➔ Thank you for your detailed comments and recommendations for improving of the writing.

3. Section 3.2.4 is lengthy and lacks of clear structure, making it difficult to follow. Please consider condensing the section for better readability. The authors could begin by introducing the observations, followed by the analysis that progressively address key aspects.

Thank you for this comment. We rearranged the structure of Section 3.2.4 in order to increase readability and shortened the discussion at some points.

Specific comments:

L20: A logic issue with 'By serving as cloud condensation nuclei (CCN) or ice nucleating particles (INPs)…, the microphysical properties of water clouds…

➔ To avoid confusion, the "water" was removed, now generally speaking of clouds. References were added to show that there is also an aerosol effect on ice clouds similar as the Twomey effect for liquid water clouds.

L41: "Laser-induced fluorescence is a known process and several remote-sensing applications are based on it." ➔ Laser-induced fluorescence is a well-established technique, serving as the basis of several remote sensing applications.

➔ Change has been applied

L43: In the atmosphere ➔ In the domain of atmospheric research

➔ Change has been applied

L51-52: "but most were small…" do you mean these lidars were with simple configuration? And please add reference.

➔ Yes, it was meant, that these lidars described by Rao et al. (2018) and Li et al. (2019) were with simpler configuration.
➔ Also due to a comment of the other reviewer, the whole paragraph was rearranged, so that this statement was not necessary anymore.

L111: "and only a little loss of fluorescence return" ➔ "with minimal loss of the fluorescence return"

➔ Change has been applied

L114: only if no rain is expected ➔ only when no rain is expected

➔ Change has been applied

L114: Complete nights were collected since 2022 … ➔ Complete night measurements have been collected since 2022…

➔ Change has been applied

L116: the derivation of ~~the~~ new products

➔ Change has been applied

L117: similarly as ➔ similarly to

➔ The phrase has been changed as follows: "similar to Veselovskii et al. (2020)".

L119, L120: The transmission at elastic wavelenght is missing. It is important for the validity of lidar equations, although the 2 terms cancel out in Equation 3.

➔ This is correct, of course. The atmospheric transmission at the elastic wavelength was added to Eqs. (1) and (2) as T_L (transmission at the laser wavelength)

L122: "and the CR and CF represent the corresponding lidar calibration constants."

➔ Change has been applied

L210: …polluted troposphere, with overall aerosol optical depth (AOD) of around 0.8 at 532 nm.

➔ Change has been applied

L212: …and 17 UTC on 04 July… ➔ …**at 17:00 UTC** on 04 July 2023. Please use standard time notation and keep it consistent everywhere in the manuscript.

➔ We revised the notation of times and dates to keep it consistent throughout the whole manuscript.

Figure 2: what are the white points in lidar quicklooks?

➔ On 4-5 July 2023 there were optically thick clouds present. Thus, we have no reliable lidar data above (low and/or negative signals in elastic channels). This leads to a very noisy appearance (high values) of the fluorescence backscatter coefficient. Therefore, these data points were filtered by the following criterion for the range-corrected signal at 1064 nm: $P_{1064} < 5 \times 10^4$. Thus, also some noisy points in the cloud-free periods were excluded. This led to some white points in the time-height plots.

Figure 2(d) is too crowded and the profile of fluorescence backscatter coefficient is truncated, please split this figure into 2, for clarity and better presentation.

➔ Thank you for the comment. We revised the figure and split the vertical profiles into two subfigures.

L217-220: Please indicate the vertical range which you are describing. Such lidar ratios were detected in tropospheric layer or UTLS layer?

➔ The aerosol layer described here is the tropospheric layer from 3.4 to 5.8 km height. The corresponding height information has been added in the manuscript as well.

L243: 22 UTC…. indicated aerosol presence ➔ 22:00 UTC…were presented in the atmosphere. Just for logic consistency, since a cloud layer also existed.

➔ The sentence was changed to:
"Only the polluted boundary layer and some thin layers up to 4 km height indicated aerosol presence, and a thin cloud was visible at around 4 km height from 21:00 to 22:00 UTC."

L245: This already illustrates ~~already~~ that with measurements…

➔ Change has been applied

L252: the 532 nm backscatter coefficient is ~~already~~ only slightly enhanced

➔ Change has been applied

Figure 3: the curves are too crowded, if you split them into 2: elastic backscattering + fluorescence backscatter with capacity, it will look better. And the peak at about 2.2 km seems to be cloud at around 19:10 UTC.

➔ Thank you for the comment. We revised the figure and split the vertical profiles into two subfigures. Furthermore, we chose a smaller height range for the vertical profiles to present the relevant height ranges clearer.

L251-257: It seems, in this case, the detection ability of elastic channels is already at their limit. The description about maxima is not quite evident according to the figure.
How did you smooth the profiles? Is the smoothing method wavelength dependent or vertically varying? How come the backscattering at 532 fails to resolve the aerosol layer below 6.5 km but is still able to resolve the maxima at above 9 km?
This case shows the detection limit of 532 nm backcattering is about 0.2 Mm-1sr-1, is it fair to say so? Do you have any comment about the vertical variation of the fluorescence capacity? Are they real and link to smoke properties/compositions or more likely to be artifacts produced by the vertical variation of elastic and/or fluorescencence backscattering and/or smoothing method?

➔ Thank you for your two comments concerning the presentation of Figure 3. It was reshaped to make it clearer and that the maxima described in the text are better recognizable in the new (sub-)figures.
➔ The description of the maxima at 6.5 km was changed to the following:
"For the layer at 6.5 km height, their corresponding maxima are at higher altitudes than the distinct maximum in the fluorescence backscatter coefficient. The 355 nm backscatter coefficient even fails to resolve the aerosol layer at 6.5 km. However, all

→ Yes, the smoothing is done wavelength-dependent. Generally, 355 nm and 1064 nm profiles are smoothed more strongly than the one at 532 nm. The fluorescence backscatter is always smoothed in the same way as the 532 nm backscatter. In this particular case, 532 and 1064 nm are smoothed in the same way. 355 is smoothed a bit stronger. However, we tested it with the same smoothing for all 3 wavelengths. But this did not improve the 355 nm backscatter ability to resolve the layer maxima.

→ Regarding the resolution of the aerosol layers: To resolve these layers is generally difficult, as they are optically thin. But in principle, β_{532} resolves all the layers, even if sometimes only with a broad increase rather than a distinct peak.
Regarding your particular comment to the layer at around 6.5 km: Probably you refer to the fact that for this layer, the maxima of the 532 nm and 1064 nm backscatter coefficients are at slightly higher altitudes than the distinct peak in the fluorescence backscatter coefficient (this remark was also added in the manuscript). Here is an attempt to explain: Both elastic backscatter coefficients show an increase from around 6 km on, where also the fluorescence backscatter coefficient starts to increase. While β_F already peaks at 6.4 km, β_{532} reaches its maximum at 6.46 km and β_{1064} only at 6.8 km. Above their maxima, β_{532} and β_{1064} stay enhanced compared to the background up to an altitude of 7.7 km and so does β_F as well, but at clearly lower values than at the peak at 6.4 km. In combination, this sequence of the backscatter profiles could indicate the presence and increasing influence of less fluorescing aerosol particles from 6.5 to 7.7 km height. So, the different altitudes of the maxima could be due to changing aerosol characteristics at this altitude range. But this is just a hypothesis.

→ In summary, we would argue that also this layer at 6.5 km is resolved by β_{532} due to its broad increase at this altitude range and the shift in the maximum may be due to the presence of less fluorescent aerosol.

→ **Detection limit:** For the layer at 3.3 km, which is still clearly resolved, β_{532} peaks at $0.15 \text{ Mm}^{-1} \text{ sr}^{-1}$, the higher layers range around $0.1 \text{ Mm}^{-1} \text{ sr}^{-1}$. Thus, we would assume a detection limit of the 532 nm backscatter coefficient of $0.1 - 0.15 \text{ Mm}^{-1} \text{ sr}^{-1}$. These values also agree well with the two other case studies (15 May and 29 May 2023).

→ **Vertical variation of the fluorescence capacity:** As β_F and β_{532} are smoothed in the same way, smoothing effects on the fluorescence capacity can be ruled out.
In this particular case, the fluorescence capacity is difficult to evaluate as the aerosol layers are optically very thin and the elastic backscatter values are close to the background noise level and can hardly be analyzed. Thus, in this case, the fluorescence backscatter coefficient is the more interesting quantity. In the other cases presented in this manuscript with optically thicker aerosol layers, the variations in fluorescence capacity should link to the aerosol properties. For further details, please refer to our reply to your corresponding comment to the case study of 15 May 2023.

L258: (also because the particle depolarization ratio is also quite low with 2 %, not shown) → especially because the particle depolarization ratio (not shown) is also quite low, around 2 %.

→ Change has been applied

L269: (again in agreement with AERONET: 0.1 at 500 nm) → which is consistent with AERONET data showing 0.1 at 500 nm

➜ Change has been applied

L264: Another example of such "unnoticeable" layers is the night of (from) 15-16 May 2023. Similar error in L266.

➜ Changes have been applied (also at a few other places in the manuscript)

L268: In the range of 4-6.7 km, the vertical variation of fluorescence capacity is significant, from 2*10-4 to 6*10-4, such strong variation is also observed in the layer at 10.5-12.2 km. The comparability of fluorescence capacity between upper layer and lower layer depends on the selection of vertical range. So, again, the question is: do you think this variation of fluorescence capacity is real? Why would it change so much in the same plume? Is there any explanation for the drop of fluorescence capacity at the edges of the plume? Or it is just some artifacts arising from different sensitivity of detection channel or smoothing ?

➜ In this case, the variations in fluorescence capacity should be real and not systematic. Such problems with artifacts from low signal intensity may only be relevant in the case of optically very thin aerosol layers, as it was the case on 21 September 2022, where the elastic backscatter coefficients were so small that they can hardly be analyzed. Please refer to the corresponding comment above. Smoothing effects have already been ruled out there as well.

➜ At closer inspection, the "layer" in the range 4-6.7 km seems to be composed of 3 individual layers. This impression is given by 3 individual peaks in the vertical profiles of β_F and G_F in Fig. 4(d) and is corroborated by the temporal evolution before the averaging period, which can be seen in the time-height plot of the fluorescence backscatter coefficient in Fig. 4(b). Between these 3 layers, slight variations in the fluorescence capacity can be expected. And when we compare the maxima of these 3 layers, the fluorescence capacity varies from 5.1×10^{-4} (4.5 km) to 6×10^{-4} (5.0 km), which is in our opinion a quite reasonable variation for slightly different smoke layers.

The main variations that you are describing are found at the edges of the smoke plumes. This dropping at the plume edges, which you also mentioned in your comment, could have different causes: Firstly, the number of fluorescing particles could decrease towards the edges of the plume, as the highest concentration of smoke particles is expected in the plume center. Thus, the relative influence of other less or non-fluorescing background aerosol particles could increase, reducing the fluorescence capacity. This may especially be important, in cases where a layer of less or non-fluorescing aerosol particles is present above and below and some mixing occurs at the edges of both plumes. Secondly, and probably more important in this case, is the height constancy of the fluorescing aerosol layer. If the height of the layer varies during the averaging period, data points outside the plume (where the fluorescence capacity is of course lower) are averaged together with data points inside the plume. As a result, the temporal mean value of the capacity at the plume edges is lower than in the center of the plume, where in-plume data points are averaged over the entire averaging period.

This effect can be nicely observed in Fig. 2 for the measurement case on 4 July 2023. The thick smoke layer from 3.4 to 5.8 km height is rather homogeneous and its lower edge is very height-constant throughout the whole averaging period from 21:00 to 22:00 UTC. As a result, the increase in fluorescence capacity at the base of the smoke layer is very steep (cf. Fig. 2d). In contrast, the layer top height clearly decreases

L269,270: What is the travel time of this smoke layer, at 4-6.7 km? And how many days does it need to become so spherical? What is the relative humidity in this layer? The elastic backscatter coefficeint is so low in this layer that there appears to be no evidence of hygroscopic growth. Does lidar ratio tell something?

→ The calculated back trajectory suggests a transport time of approximately 10 days for layer 1 (cf. Fig. A). The relative humidity in layer 1 was in the range of 35-45% according to our lidar data.

→ The hypothesis of hygroscopic growth influencing the fluorescence capacity of layer 1 was removed due to the lack of evidence and relevance for the main message of this manuscript. Therefore, we restricted to the analysis of the aerosol optical properties (fluorescence, depolarization ratio, lidar ratio and Angström exponent) and naming possible causes for the differences.

L275: Does the back trajectory reveal any difference in transport time or pathway between the two layers?

→ The back trajectory analysis for Leipzig at the starting time of 2 UTC is displayed in Fig. A. The main difference lies in the altitude of the back trajectories. Layer 2 seems to be transported constantly at around 10-11 km height (which was at least at our observation place and time slightly above the tropopause). Instead, layer 2 mainly ranged at altitudes from 5-6 km, for a short time reaching 8 km height at the maximum.

→ Also, the back trajectories suggest a shorter travel time of layer 2 (5-6 days) compared to approximately 10 days of layer 1.

→ The shorter travel time and lower humidity (because of higher altitude, shortly above the tropopause) indicate less aging of layer 2 compared to layer 1.
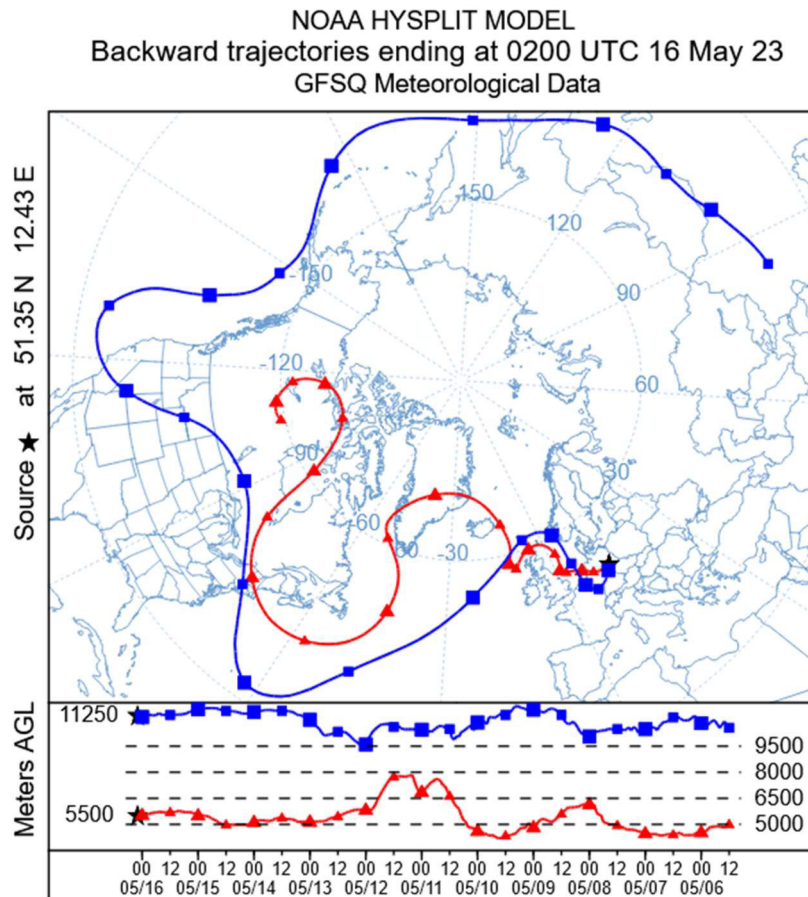
Figure A: Back trajectory analysis for Leipzig at 02:00 UTC on 16 May 2023.

L276: 'the aforemential Canadian wildfires' : not clear which wildfire is it referring to

➔ These simple back trajectory analysis does not allow the attribution to a certain wildfire source. Only a general source region can be identified. In general, the entire paragraph has been revised and the sentence mentioned in your comment has been omitted.

L288: '…reveals another aerosol layer": this layer is also called "another ..layer" in L271. Better to name the layers to be 'Layer 1, 2,…' in each case to improve the clarity.

➔ Thank you for the hint. The layers have been numbered accordingly.

L307-309: This long sentence should better be rephrased to improve clarity.

➔ Indeed. The sentence and the whole paragraph have been reformulated to improve clarity and accuracy.

L310: Is it range-corrected signal in Figure 5(b)? Is the fluorescence signal rescaled? What is the definition of the unit 'MC*s-1'? Are they from analog channels or photon-counting channels?

➔ Yes, Fig. 5(b) showed the range-corrected signal. To improve the accuracy of the explanations, this plot was replaced by the background-corrected signal, which units are megahertz (MHz).

L315: only a part of the actual (strong) backscatter signal: (strong) ? this is confusing.

➔ This part has been rephrased. Thanks for the remark.

L316,317: please provide the reference of the overlap correction. Are the profiles shown in previous figures corrected from overlap? Why are they cut below 1 km?

➔ We computed backscatter coefficients from the ratio between the elastic and Raman channel, which in principle does not suffer from overlap effects. Because our system has a large telescope and narrow field-of-view, the full overlap is reached only at about 2.5 km. We can extend the observational capabilities to the ground but in the first kilometer some interchannel-overlap and deadtime effects may introduce artifacts into the retrieved aerosol information. For this reason, we cut the signal at 1.2 km routinely.
The overlap function is based on Raytracing calculations. The correction is relevant for the extinction coefficient. The study focuses rather on the backscatter coefficient. This aspect has been removed from the paragraph, to keep the description as clear and accurate as possible focusing more in the sensitivity of the system.

L324: dynamical ➔ dynamic

➔ The word has been corrected. Thanks.

L324-325: …much smaller, and because of that, the… ➔ …much smaller. Consequently, the…

➔ Thank you for the hint. The sentence has been modified.

L332: 'UTLS' defined twice

➔ Thank you for the hint. The second definition was removed.

Figure 6: please add the profiles of temperature and relative humidity with respect to ice.

➔ The profiles of temperature and relative humidity obtained from radiosonde measurements at Lindenberg were added as Fig. 6(e).

L346: "water content can quench fluorescence scattering": this sentence is not clear enough. How does water content influence fluorescence capacity, by suppressing fluorescence emission or enhancing elastic scattering?

➔ As also explained in the reply to the other reviewer: To mention quenching here, makes no sense. The low fluorescence capacity of cloud particles is due to their strong elastic backscattering, while their fluorescence backscattering is low, as pure water does not fluoresce. The corresponding part of the sentence has therefore been corrected. For further details, please refer to the reply to the other reviewer.

L349: separate → differentiate

→ Change has been applied

L352,353: 'Furthermore, the smoke layer rose…the cloud top rose first, and later, become scattered and seemed to dissolve. All these facts indicate that the smoke particles may have triggered the cloud formation by serving as INPs'. The link between smoke particles serving as INPs and the movements of clouds and smoke layers is not clearly explained here. In addition, as clouds became thinner and thinner, the smoke layers seemed to get thicker, is it related to ice crystal formation?

→ The link between the temporal evolution of the smoke layer and the cirrus clouds could be that smoke particles are serving as INPs. The important observation is here, that the presence of cirrus clouds coincides temporally and spatially with the presence of smoke particles. From the time at which the smoke layer was located exclusively at higher altitudes than the altitude at which the cloud formation took place, ice nucleation appears to cease and cirrus clouds no longer form. If the ice nucleation would not be related to the smoke particles but to background aerosol only, one would not expect this stop in ice nucleation and cloud evolution. Therefore, this observation suggests that the smoke particles act as INPs in this case.

→ To comment on the thickening of the smoke layer over time would be rather hypothetical. Of course, looking at the lower fluorescence backscatter coefficient at the time, where the smoke layer seemed to be partly inside the cloud (before 23:30 UTC), one could speculate that ice nucleation and subsequent falling of the ice crystals may have reduced the optical thickness of the smoke layer at that time. However, it could also simply be a transport-related thickening of the smoke layer. So, the answer to your question is: We can't tell…

L354: "the clouds even became scattered and seemed to dissolve": why do ice clouds dissolve at such temperature?

→ Thank you for your comment. You are right, "dissolve" is probably the wrong vocabulary here. As we are monitoring with lidar the temporal evolution of the atmospheric column over a fixed location, the absence of cirrus clouds at the later time steps does not mean that the cirrus clouds observed at previous time steps did really dissolve, but that no cloud formation happened anymore at the later time steps. So, the following formulation is probably better: "The clouds even became scattered and the ice nucleation and cloud formation seemed to stop."

→ The change was applied to the manuscript.

L369: on 29 May → in the night of 29 May

→ Change has been applied

L371: 21 to 22 UTC → 21:00 to 22:00 UTC

→ Change has been applied

L371: please indicate the two nucleation sections in Figure 6(d)

→ Horizontal dashed lines were added to Fig. 6(d) to indicate the nucleation sections.

L375: 'Above, the high fluorescence capacity…': In the context, 'Above' does not specify what you are referring to.

➔ It was changed to "At around 12 km height, …"

L376-382: A general comment regarding the description of figures: when presenting values in a vertical profile, please include their corresponding vertical levels rather than relying only on descriptive phrases like 'down to the upper boundary of the lower cloud part' or 'at the top of the lower part of the cirrus clouds'…

➔ The vertical levels were either added or used to replace the descriptive phrases.

L382-383: If this difference in fluorescence backscattering can be explained by fluorescence quenching, then why did it occur specifically at the cloud top of the upper cirrus part, rather than elsewhere?

➔ First of all, fluorescence quenching is only one possibility. The height level of the reduction in the fluorescence backscatter coefficient seems to coincide with the upper nucleation zone, where ice crystals are forming. But whether this would increase fluorescence quenching is hypothetical.
➔ However, the removal of smoke particles (that served as INP) from this altitude due to the falling of ice crystals seems to be a more plausible explanation as it is also stated in the revised version of the manuscript.

L390: 'ranged from (at) the minimal values of…to…'

➔ We meant that the values of 10^{-6} to 10^{-5} are minimal compared to other regions and cases. We replaced minimal by "very low" to make this clearer.

L392--394: ' In this case,…again up to 1.4*10-5Mm-1sr-1 ." --- the fluroescent layer at around 7 km seemed not in contact with cirrus clouds after 21:00, and there was no fluorescence between 7.5 km and 8.5 km. Therefore, I am not convinced that this layer was due to the accumulation of smoke particles at cloud base.

➔ We disagree in this point. In our opinion, the missing contact of the fluorescing layer to the cirrus cloud does not argue against the hypothesis of accumulation of smoke particles at the cloud base. With lidar measurements, we only get a "snapshot" at a certain time and location. We do not know what happened to a certain air mass some minutes before we observed it with the lidar. The sublimation of the ice crystals may have happened some time before, so that the remaining smoke particles were later observed a bit below the cloud base. Furthermore, the vertical profile of the relative humidity over ice from the radiosonde at Lindenberg in Fig. 6(e) supports our hypothesis of scavenging and subsequent accumulation of smoke due to sublimation at the cloud base. Around 8 km altitude, RH_ice drops below 100% and decreases to values of 50-60% at around 7 km. In such humidity conditions, falling ice crystals will sublimate, leaving only remaining smoke particles, that had served as their INPs before. And this is also supported by our lidar observations. The fluorescence capacity plot in Fig. 6(c) reveals such strongly fluorescing layers not only at 7 km height between 21:00 and 22:00 UTC, but also between 01:15 UTC and 02:00 UTC at around 8 km height.

Figure 6(e) is not described.

➔ Figure 6(e) was replaced by a figure showing temperature and relative humidity over ice from a radiosonde at Lindenberg at 18 UTC on 29 May 2023.

Figure 7: please indicate the contour of cloud area, since the gray colors marked the area where cloud particles and smoke particles co-exist, as well as the boundaries of clouds.

➔ The top of the cirrus cloud layer has been marked in black in Fig. 7.

Figure 7 caption: Height-time distributions of the particle backscatter coefficient at 532 nm in the night of 29 - 30 May 2023. Height-time bins with a high fluorescence backscatter coefficient (> 2.5 ×10−5 Mm−1 sr−1) are colored in gray.

➔ Change has been applied

L406-407: 'in such cases with low but relevant aerosol presence in the cloud surroundings, especially at the cloud top': too long, with lots of ambiguity, therefore not clear. It would be better to put it in a general way: in such cases where aerosols/smoke particles are identified as INP…

➔ The important point in this statement is that the fluorescence lidar may be used to provide INP information also in cases with **low aerosol load**. Therefore, this information cannot be omitted here.

L421: newly-implemented ➔ newly implemented

➔ Change has been applied

L422: the 2023 summer wildfire season ➔ the summer wildfire season of 2023

➔ Change has been applied

The conclusion part could be re-organized to make it more fluent, compact and direct. Unnecessary sentences like 'back trajectory calculations suggested…' could be distracting (and it has been mentioned before), therefore, reduce the readability.

➔ The statement regarding the back trajectory calculations was removed.

Invisible aerosol layers: improved lidar detection capabilities by means of laser-induced aerosol fluorescence

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Abstract. One of the most powerful instruments for studying aerosol particles and their interactions with the environment is atmospheric lidar. In recent years, fluorescence lidar has emerged as a useful tool for identifying aerosol particles due to its link with biological content. Since 2022, this technique has been implemented in Leipzig, Germany. This paper describes the experimental setup and data analysis, with a special emphasis on the characterization of the new fluorescence channel centered at 466 nm. The new capabilities of the fluorescence lidar are examined and corroborated through several case studies. Most of the measurement cases considered are from the spring and summer of 2023, when large amounts of biomass-burning aerosol from the huge forest fires in Canada were transported to Europe. The fluorescence of the observed aerosol layers is characterized. For wildfire smoke, the fluorescence capacity was typically in the range of 2–7×10^{-4}, which aligns well with the values reported in the literature, with slightly larger values. The key aspects of this study are the capabilities of the fluorescence lidar technique, which can potentially improve not only the typing but even the detection of aerosol particles. In several measurement cases with an apparently low aerosol load, the fluorescence channel clearly revealed the presence of aerosol layers that were not detectable with the traditional elastic-backscatter channels. This capability is discussed in detail and linked to the fact that fluorescence backscattering is related to aerosol particles only. A second potential of the fluorescence technique is the distinction between non-activated aerosol particles and hydrometeors, given water's inability to exhibit fluorescence. A smoke-cirrus case study suggests an influence of the aerosol layer on cloud formation, as it seems to affect the elastic backscatter coefficient within the cloud passing time. These mentioned applications promise huge advancements towards a more detailed view of the aerosol-cloud interaction problem.

1 Introduction

A crucial player in the atmospheric system are aerosol particles, given their role in various processes that ultimately shape the Earth's energy and hydrological budgets. Firstly, aerosol particles scatter and absorb radiation, affecting the energy balance on a global scale. By serving as cloud condensation nuclei (CCN) or ice nucleating particles (INPs), these particles can impact the microphysical properties of ~~water~~ clouds (Liu et al., 2014), making them more or less reflective depending on the aerosol situation~~(Twomey, 1974, 1977; Twomey et al., 1984).~~ . In the case of liquid-water clouds, this effect has

been largely studied (Twomey, 1974, 1977; Twomey et al., 1984; Quaas et al., 2020), for ice clouds it is a rather new topic (Patnaude and Diao, 2020; Maciel et al., 2023). Because the microphysical properties of a cloud play a major role in its development and the formation of precipitation, aerosol conditions can further affect the extension and lifetime of cloud events and therefore the global albedo (Albrecht, 1989; Stevens and Feingold, 2009). Highly absorbing aerosol particles might even affect clouds via the so-called semi-direct effect, which can manifest, e.g., in the evaporation of cloud droplets due to an aerosol-heated environment (Hansen et al., 1997). Aerosol effects on the ice phase of cloud formation only complicate the picture.

Multiple efforts have been made to analyze the role of aerosol particles as INPs in mixed-phase clouds via heterogeneous freezing and the global effect (Lohmann, 2017). As for cirrus clouds, recent studies suggest that heterogeneous freezing in cirrus clouds, particularly via smoke particles, needs to be explored in more detail (Ansmann et al., 2021; Veselovskii et al., 2022a; Mamouri et al., 2023; Ansmann et al., 2024a, b). To improve our understanding of these complex aerosol-cloud interaction processes, reliable detection and characterization of atmospheric aerosol particles are essential.

Multi-wavelength polarization lidars are powerful tools to detect and characterize aerosol particles. After decades of study, several classification schemes are available in the literature (Floutsi et al., 2023; Groß et al., 2013; Burton et al., 2012), mostly relying on intensive (i.e., concentration-independent) optical properties such as the lidar ratio, particle depolarization ratio and Ångström exponent. However, although clear signatures can be expected for some particle types (e.g., low particle depolarization and low lidar ratios for marine aerosol particles), some limitations remain. Distinguishing between stratospheric smoke and volcanic sulfates or separating between tropospheric smoke and urban pollution remain difficult tasks, as their typical ranges of values for particle depolarization and lidar ratios partially overlap. Additional information, such as the fluorescence of atmospheric aerosol particles, may be required to address these typing difficulties (Veselovskii et al., 2022b).

Laser-induced fluorescence is a ~~known process and~~ well-established technique, serving as the basis of several remote-sensing applications ~~are based on it~~. Fluorescence lidars have been around for a while, but their application has mostly focused on water composition (Palmer et al., 2013; Cadondon et al., 2020) and vegetation (Edner et al., 1994). In the ~~atmosphere~~ domain of atmospheric research, efforts have mostly gone towards the detection of single molecules (Mcllrath, 1980; Wang et al., 2021). To investigate the fluorescence of atmospheric aerosol particles, the experiments have been mostly performed through in situ probing (Pinnick et al., 2004; Pan, 2015; Zhang et al., 2019; Kawana et al., 2021). As an example, Pan (2015) analyzed the fluorescence of aerosol particles by measurements with an ultraviolet aerodynamic particle sizer (UV-APS) and reported a spectral range for atmospheric fluorescence of 400 to 650 nm, when excited at 355 nm. A first hint towards the observation of atmospheric aerosol fluorescence with lidar came in 2005, when Immler et al. (2005) observed an inelastic backscatter signal in the water vapor Raman channel (i.e., at 407 nm) that was not produced by Raman scattering. They attributed it to the laser-induced fluorescence emission from biomass-burning (BB) aerosol particles and already linked the aerosol fluorescence to organic compounds. ~~Pan (2015) analyzed the fluorescence of aerosol particles by measurements with an ultraviolet aerodynamic particle sizer (UV-APS) and reported a spectral range for atmospheric fluorescence of 400 to 650 nm, when excited at 355 nm. Later,~~ In the following, the first atmospheric fluorescence lidars were set up~~, but most were small and dedicated to fluorescence measurements only. Rao et al. (2018) and Li et al. (2019) used Nd:YAG lasers at 266 nm and 355 nm, respectively, and studied the backscattered light in one elastic-backscatter (at the excitation wavelength)~~

and one fluorescence channel. Both instruments looked only at the boundary layer. Saito et al. (2018) studied the fluorescence of atmospheric pollen with a lidar at 355 nm excitation wavelength and found a similar spectral range of 400 to 600 nm for the fluorescence emission. Sugimoto et al. (2012) constructed a lidar spectrometer and studied the fluorescence of Asian dust and pollution aerosols in the lower troposphere. The first advanced multi-channel atmospheric lidar system with fluorescence technology was implemented at Lindenberg observatory of the German Meteorological Service (DWD). For the first time, Reichardt et al. (2018) used a Initially, Reichardt (2014) observed atmospheric aerosol fluorescence with a lidar spectrometer that was dedicated to measurements of atmospheric water content. He established the fluorescence capacity as a new intensive aerosol property/parameter, which is defined as the ratio of the fluorescence backscatter coefficient to an elastic particle backscatter coefficient. Later, Reichardt et al. (2018) implemented a second spectrometer to measure the laser-induced fluorescence of aerosol particles in the middle and upper troposphere. They characterized the fluorescence of mineral dust and BB aerosol and underlined the capabilities of fluorescence measurements to study aerosol-cloud coexistence by enabling the observation of aerosol particles even inside clouds. Veselovskii et al. (2020, 2021) extended the concept to Saito et al. (2018) studied the spectral fluorescence of atmospheric pollen with lidar and reported a spectral range of 400 to 600 nm for the fluorescence emission, when excited at 355 nm.

Parallel to the developments in spectrometric fluorescence measurements, a further, more easily accessible approach to measure aerosol fluorescence with a single broadband/discrete lidar channel was pursued. Rao et al. (2018) and Li et al. (2019) used Nd:YAG lasers at 266 nm and 355 nm, respectively, and studied the backscattered light in one elastic-backscatter (at the excitation wavelength) and one fluorescence channel. Both instruments were dedicated to aerosol fluorescence measurements and looked only at the boundary layer. Veselovskii et al. (2020, 2021) extended the concept by adding a practical approach, in which only a single broadband fluorescence channel was added into a into an existing multi-wavelength lidar system at Lille, France. They also described a retrieval scheme for the computation of the fluorescence backscatter coefficient out of the signal ratio between the fluorescence and the nitrogen channels. Their observations also confirmed the potential of the fluorescence lidar technique to study aerosol particles inside clouds (Veselovskii et al., 2022a). Veselovskii et al. (2022b) showed that fluorescence measurements can improve the aerosol classification with lidar. They proposed, for the first time, a simple classification scheme that combines the linear depolarization ratio with the fluorescence capacity (Reichardt, 2014), which is defined as the ratio of the fluorescence backscatter coefficient to an elastic particle backscatter coefficient (e.g., at 532 nm as used in this study). With this approach, they were able to discriminate between smoke, mineral dust, pollen and urban aerosol, as pollen and smoke showed significantly higher values of fluorescence capacity than urban aerosol and mineral dust. Reichardt et al. (2023) described a procedure for absolute calibration of spectrometric fluorescence measurements and proposed a method to correct for the systematic fluorescence error in water vapor measurements with Raman lidar, which is significant for dry and strongly fluorescent aerosol layers. They also emphasized that the spectrum's shape is closely related to the aerosol type. Veselovskii et al. (2023) presented an approach to measure rough fluorescence spectra with a lidar with five discrete broadband fluorescence channels at Moscow, Russia. They reported advancements in aerosol typing with this approach compared to a single fluorescence channel. Smoke and urban aerosol particles could be discriminated even at high relative humidity and in the presence of hygroscopic growth.

In this work, we explore the observational capabilities of an atmospheric fluorescence lidar utilizing measurements performed in Leipzig, Germany, with an upgraded system since 2022. A detailed description of the new experimental setup is provided in Sec. 2. The analysis of several measurement case studies is presented in Sec. 3. Our findings corroborate the results obtained by previous studies on the capabilities of fluorescence lidars and deepen the discussion in the field of aerosol studies utilizing fluorescence lidar observations. We discuss a unique new capability that is special to this measurement approach. Because it is sensitive to particles only, a fluorescence channel can potentially improve not only the typing but even the detection of aerosol particles. Sec. 3.2.3 provides an in-depth analysis of the reasons for this increased sensitivity of the fluorescence channel to aerosol particles. An exceptional smoke-cirrus interaction case presented in Sec. 3.2.4 highlights the importance of the ability to detect thin aerosol layers in the upper troposphere and lower stratosphere (UTLS) region for the investigation of cirrus cloud formation. Furthermore, it corroborates and expands the initial work on the detection of fluorescence signals inside ice clouds. The paper concludes in Sec. 4.

2 Experimental Setup

2.1 Implementation of a fluorescence channel in the MARTHA lidar system

The Multi-wavelength Atmospheric Raman lidar for Temperature, Humidity and Aerosol profiling (MARTHA) is a lab-based lidar system at the Leibniz Institute for Tropospheric Research (TROPOS) in Leipzig. It emits electromagnetic radiation at three wavelengths (355 nm, 532 nm and 1064 nm) with an overall pulse energy of about 1.2 J at a repetition rate of 30 Hz and collects the backscattered radiation with a large main mirror, which measures 80 cm in diameter. A detailed description of the MARTHA system is given in Mattis et al. (2002), Schmidt et al. (2013) and Jimenez et al. (2019).

To measure the laser-induced fluorescence of atmospheric aerosol, the MARTHA lidar system was upgraded by adding a discrete fluorescence channel into the receiving unit in 2022. To facilitate comparability, the spectral range of the channel was set in the same wavelength range as in Veselovskii et al. (2020). A 44 nm-wide interference filter from Alluxa centered at 466 nm is used to select a part of the fluorescence spectrum of fluorescing aerosol particles. Because of the similar features, a first comparison of the results obtained in Lille, France, and Leipzig, Germany, is possible.

The backscattered fluorescence intensity depends on the aerosol situation, but in general, it is much weaker than elastic backscatter signals. Therefore, the signal-to-noise ratio in the new channel must be as high as possible. The features that made the MARTHA system suitable to detect fluorescence are its large telescope area and the high-power laser. The second and third harmonic generation setups allowed an increase in the laser energy at 355 nm, sending 6 ns pulses with an energy of about 350 mJ. The new setup of the MARTHA far-range (FR) receiver, including the fluorescence channel, is displayed in Fig. 1. The new detection channel was placed in the branch with the lower wavelengths. Therefore, the first long-pass beam splitter (BS1) was replaced to ensure the complete reflection of the intended fluorescence spectral band. A second beam splitter (BS2) was added. It transmits the shorter wavelengths to the elastic-backscatter and Raman channels related to the UV laser emission at 355 nm and reflects the longer wavelengths towards the fluorescence channel. As the fluorescence signal can be 4–5 orders of magnitude weaker than elastic backscattering (Veselovskii et al., 2020), sufficient suppression of the elastic returns in the
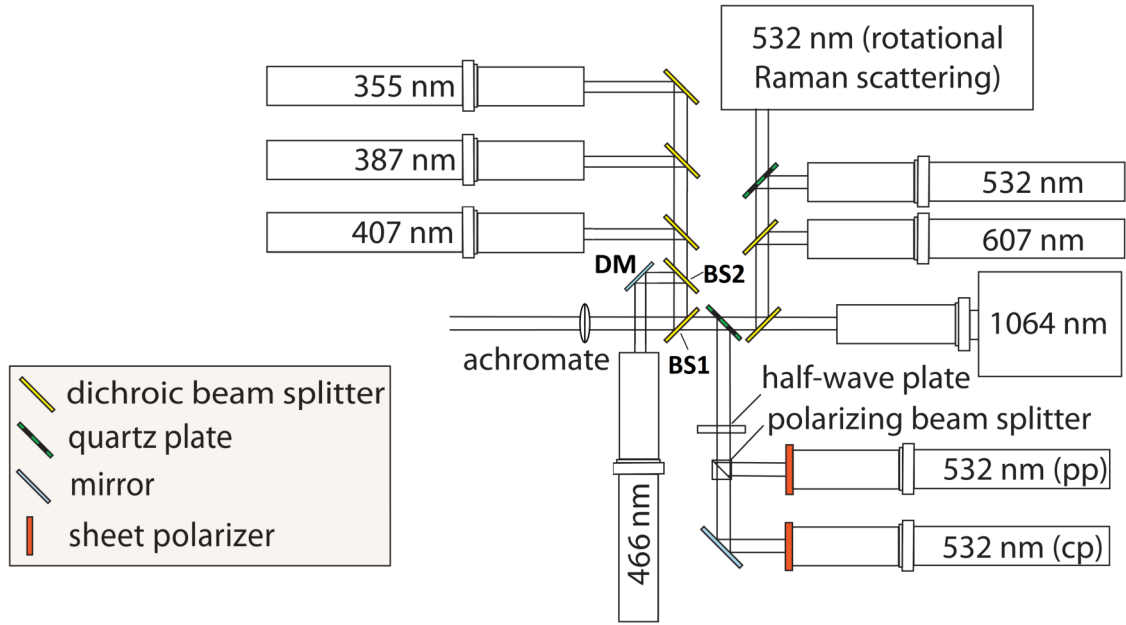
4

Figure 1. Setup of the far-range receiving unit of the MARTHA system after implementing the fluorescence channel (graphic adapted from Schmidt (2014)). DM: dielectric mirror; BS: beam splitter.

new channel was essential to measure fluorescence. The two new beam splitters received customized coatings from Laseroptik GmbH to guarantee high suppression of the elastic-backscatter (and Raman) lines ~~and only a little~~ with minimal loss of fluorescence return. Two interference filters were placed in tandem to suppress the elastic components further.

2.2 Analytical scheme of the fluorescence backscatter coefficient

The lidar system was operated manually and only ~~if~~ when no rain was expected. Complete ~~nights were~~ night measurements have been collected since 2022 and were analyzed with a focus on the fluorescing properties of the observed aerosol layers. A second important step is the derivation of ~~the~~ new products. The procedure is described as follows: The aerosol fluorescence backscatter coefficient was obtained by forming the ratio of the fluorescence (P_F) to the nitrogen Raman (P_R) signal, ~~similarly as in~~ similar to Veselovskii et al. (2020). Both signals can be described in terms of the lidar equation:

$$P_F = \beta_F T_L T_F C_F \tag{1}$$

$$P_R = \beta_R T_L T_R C_R. \tag{2}$$

T_L is the atmospheric transmission at the emitted laser wavelength. T_R and T_F denote the atmospheric transmission at the Raman and fluorescence wavelength ranges, respectively, and C_R and C_F represent the corresponding lidar calibration constants. By dividing Eq. (1) by Eq. (2), the following expression for the aerosol fluorescence backscatter coefficient β_F can

be derived:

$$\boldsymbol{\beta}_\mathrm{F} = \frac{\boldsymbol{P}_\mathrm{F}}{\boldsymbol{P}_\mathrm{R}} \frac{\boldsymbol{T}_\mathrm{R}}{\boldsymbol{T}_\mathrm{F}} \frac{C_\mathrm{R}}{C_\mathrm{F}} \boldsymbol{\beta}_\mathrm{R}. \tag{3}$$

The Raman backscattering $\boldsymbol{\beta}_\mathrm{R}$ is computed using the following expression in terms of the Rayleigh molecular backscatter coefficient ($\boldsymbol{\beta}_\mathrm{mol}$):

$$\boldsymbol{\beta}_\mathrm{R} = D_\mathrm{R} \boldsymbol{N}_{\mathrm{N}_2} = 0.78 D_\mathrm{R} \boldsymbol{N}_\mathrm{mol} = 0.78 \frac{D_\mathrm{R}}{D_\mathrm{mol}} \boldsymbol{\beta}_\mathrm{mol}, \tag{4}$$

with $\boldsymbol{N}_{\mathrm{N}_2}$ and $\boldsymbol{N}_\mathrm{mol}$ the number density of nitrogen and air molecules, respectively. $D_\mathrm{R}/D_\mathrm{mol}$ accounts for the Raman to Rayleigh backscatter differential-cross-section ratio. These cross sections were determined theoretically using Eqs. (20) and (14) in Adam (2009), resulting in theoretical values of $D_\mathrm{R}^* = 2.7344 \times 10^{-34}\,\mathrm{m}^2\,\mathrm{sr}^{-1}$ and $D_\mathrm{mol} = 3.10875 \times 10^{-31}\,\mathrm{m}^2\,\mathrm{sr}^{-1}$.

2.3 Technical considerations for the calibration of the fluorescence channel

To derive the particle fluorescence backscatter coefficient from Eq. (3), the traditional method, using a particle-free reference height, cannot be applied, due to the unknown fluorescence response of the background aerosol. Instead, a characterization of the channel's system efficiencies is needed. The contribution of each component in the respective detection path was carefully determined to infer the overall efficiencies and build the lidar-constant ratio $C_\mathrm{R}/C_\mathrm{F}$.

The first point to consider is the bandwidth of the interference filters. For the 387 nm nitrogen Raman channel, with a bandwidth of 2.7 nm, only 95 % of the theoretical Raman cross section can reach the detector, reducing the actual cross section at the detector to $D_\mathrm{R} = 0.95 \times D_\mathrm{R}^* = 2.59768 \times 10^{-34}\,\mathrm{m}^2\,\mathrm{sr}^{-1}$. This value is then used in Eq. (4) together with the molecular backscatter computed based on the temperature and pressure profiles provided by the Global Data Assimilation System (GDAS) (Rodell et al., 2004) to derive the Raman backscatter coefficient. As the laser power, pulse length, and telescope area are the same for both detection channels, the lidar-constant ratio $C_\mathrm{R}/C_\mathrm{F}$ simplifies to the ratio of the channel efficiencies. This ratio comprises the transmittances or reflectances of the optical elements (such as beam splitters, mirrors, interference filters and neutral-density filters) and the detection efficiencies of the detectors. The transmittances and reflectances of the optical components are collected in Tab. 1. As the neutral-density (ND) filters are eventually changed depending on the atmospheric and system conditions, only one exemplary set of ND filters, which is representative of most of the cases studied in this manuscript, was chosen for Tab. 1. When determining the ND filter transmission, the spectral dependence provided by the manufacturer (Thorlabs) was considered. The detection efficiencies of the photomultiplier tubes (PMTs) are split into electrical gain and detector efficiency. The ratio of the electrical gains was obtained by swapping the detectors in the nitrogen Raman and fluorescence channels and building the ratio of the mean signals measured by both detectors for each channel. This test yielded a PMT gain ratio ($\eta_\mathrm{gain,R}/\eta_\mathrm{gain,F}$) of 1.4155, indicating a higher gain of the nitrogen Raman channel's PMT. As for the detector surface, the so-called quantum efficiency accounts for the amount of photoelectrons generated by the cathode divided by the number of incident photons. This efficiency depends on the photon wavelength (Wright, 2017). The spectrally resolved quantum-efficiency data provided by Hamamatsu were considered to assess the PMT type used in the MARTHA system. The maximum efficiency of the detectors is about 35 %, and the values at the wavelength ranges of the two lidar channels were

Table 1. Transmittances $\mathcal{T}$ or reflectances $\mathcal{R}$ of all optical elements in the $387\,\mathrm{nm}$ and $466\,\mathrm{nm}$ FR channels. PMT quantum efficiencies, PMT gain ratio and overall ratio of the lidar calibration constants in both channels.

FR channel	Nitrogen Raman (387 nm)	Fluorescence (466 nm)
Common beam splitter (BS2)	$\mathcal{T}_1 = 97.1\%$	$\mathcal{R}_1 = 98\%$
Further (unique) optical elements	beam splitter $407\,\mathrm{nm}$: $\mathcal{T}_2 = 94.5\%$ beam splitter $387\,\mathrm{nm}$: $\mathcal{R}_2 = 95\%$	dielectric mirror (DM) $\mathcal{R}_3 = 99.75\%$
Interference filters	$\mathcal{T}_3 = 70\%$	$\mathcal{T}_4 = 92.5\%$ ($\mathcal{T}_4{}^2$ because of 2 filters)
Neutral-density filters (example)	$\mathrm{OD} = 1.3$ $\mathcal{T}_{\mathrm{ND}} \approx 0.0213$	no neutral-density filters $\mathcal{T}_{\mathrm{ND}} = 1$
Product $\prod\limits_{i=1}^{N} \mathcal{T}_i \prod\limits_{j=1}^{M} \mathcal{R}_j$	$\mathcal{R}_2\,\mathcal{T}_1\,\mathcal{T}_2\,\mathcal{T}_3\,\mathcal{T}_{\mathrm{ND}} = 0.01299$	$\mathcal{R}_1\,\mathcal{R}_3\,\mathcal{T}_4{}^2 = 0.8364$
PMT quantum efficiency PMT gain ratio	34.66 % $\eta_{\mathrm{gain,R}}/\eta_{\mathrm{gain,F}} = 1.4155$	25.13 %
Overall ratio of the lidar calibration constants	$C_{\mathrm{R}}/C_{\mathrm{F}} \approx 0.0303$	

determined by interpolation from the provided data points and averaging over the filter width of the interference filter in the fluorescence channel. This resulted in values of $\eta_{\mathrm{qe,R}} = 34.66\%$ and $\eta_{\mathrm{qe,F}} = 25.13\%$ for the quantum efficiencies of the used PMT type in the nitrogen Raman (386–388 nm) and fluorescence channel (444–488 nm), respectively, and we finally obtained a ratio of $\eta_{\mathrm{qe,R}}/\eta_{\mathrm{qe,F}} = 1.379$.

After these considerations, the ratio of the lidar calibration constants can be calculated from the efficiency ratios of the optical elements, detector gain and spectral response as follows:

$$\frac{C_{\mathrm{R}}}{C_{\mathrm{F}}} = \frac{\mathcal{R}_2\,\mathcal{T}_1\,\mathcal{T}_2\,\mathcal{T}_3\,\mathcal{T}_{\mathrm{ND}}}{\mathcal{R}_1\,\mathcal{R}_3\,\mathcal{T}_4{}^2}\,\frac{\eta_{\mathrm{gain,R}}}{\eta_{\mathrm{gain,F}}}\,\frac{\eta_{\mathrm{qe,R}}}{\eta_{\mathrm{qe,F}}}. \tag{5}$$

For the set of ND filters considered in Tab. 1 (OD 1.3 in the nitrogen Raman and no ND filters in the fluorescence channel) it results in a value of $C_{\mathrm{R}}/C_{\mathrm{F}} \approx 0.0303$.

The remaining unknown in Eq. (3) is the ratio $\boldsymbol{T}_{\mathrm{R}}/\boldsymbol{T}_{\mathrm{F}}$ of the atmospheric transmissions (ground to target) at the Raman and fluorescence wavelengths, respectively. The molecular part ($\boldsymbol{T}_{\mathrm{R}}/\boldsymbol{T}_{\mathrm{F}}|_{\mathrm{mol}}$) is calculated straightforwardly from the extinction and backscatter coefficients; the aerosol contribution to the transmission ratio ($\boldsymbol{T}_{\mathrm{R}}/\boldsymbol{T}_{\mathrm{F}}|_{\mathrm{par}}$) requires previous knowledge about

the aerosol backscatter coefficient. For the profile-based analysis, the aerosol optical properties are determined with the traditional Raman technique (Ansmann et al., 1990, 1992). The particle backscatter coefficient at high temporal resolution was obtained via a constant-based approach, in which a previous profile-based retrieval is needed to calculate the lidar constants, which are then used to compute high-resolution products out of the elastic-backscatter and Raman signals (Baars et al., 2017). In general, the particle atmospheric transmission differs little at the two wavelengths, making the effect on the fluorescence backscatter coefficient small, partially because only about 80 nm separate the central wavelengths of the Raman and fluorescence channels. For the cases with low and medium aerosol loads (see Sec. 3.2), the error in case of non-consideration of the differential transmission was in the range of 2–6 %. In case of an unusually high aerosol optical depth, like on the 4 July 2023 (see Sec. 3.1), the error was in the order of 10 %. Thus, the differential particle transmission at the two wavelengths was considered to guarantee the quality of the fluorescence backscatter coefficient, even above strongly backscattering aerosol layers. But still, the assumption of an appropriate Ångström exponent is necessary, which imposes an uncertainty of ± 1–7 % on the determined $T_\mathrm{R}/T_\mathrm{F}|_\mathrm{par}$, depending on the optical thickness of the present aerosol layers.

The data set acquired in Leipzig was then analyzed in a semi-automatic manner, setting the calibration constants and the reference height (particle-free) manually for each case. The fluorescence capacity G_F,

$$G_\mathrm{F} = \frac{\beta_\mathrm{F}}{\beta_{532}}, \tag{6}$$

was calculated as the ratio of the fluorescence backscatter coefficient (β_F) to the elastic particle backscatter coefficient at 532 nm (β_{532}). To encourage comparability with different setups (fluorescence lidars which do not deploy the second-harmonic generation wavelength and/or use a spectrometric approach or broadband fluorescence channels with different interference filter bandwidth), the general and season-mean values are also provided as spectral fluorescence capacity with respect to the third harmonic generation wavelength 355 nm (G_F^{355}),

$$G_\mathrm{F}^{355} = \frac{\beta_\mathrm{F}}{\beta_{355}\, d_\mathrm{IF}}, \tag{7}$$

where $d_\mathrm{IF} = 44\,\mathrm{nm}$ is the bandwidth of the interference filter in the fluorescence channel. Furthermore, data from the collocated portable Raman lidar Polly$^\mathrm{XT}$ (Engelmann et al., 2016) at TROPOS were used to provide quality-assured depolarization profiles.

3 Observational results

Due to the broad bandwidth of the fluorescence channel and the low intensity of the fluorescence signal, measurements were only possible during the night. At daytime, scattered solar radiation would cause too much noise in the fluorescence channel. As the MARTHA system is operated manually, the number of measurements remains limited. Since August 2022, about 50 measurements have been performed, providing more than 250 hours of atmospheric fluorescence observations. Typical atmospheric values of the fluorescence backscatter coefficient and fluorescence capacity, that were obtained at Leipzig during the time period from August 2022 to October 2023, are presented in the next paragraph.

In general, β_F ranged between $1 \times 10^{-5}\,\mathrm{Mm}^{-1}\,\mathrm{sr}^{-1}$ for background aerosol and more than $1 \times 10^{-3}\,\mathrm{Mm}^{-1}\,\mathrm{sr}^{-1}$ for optically extraordinarily thick wildfire smoke layers. Correspondingly, G_F ~~varied from 1×10^{-5} for background aerosol~~ (G_E^{355}) varied from $\sim 10^{-6}$ ~~for clouds)~~ ($\sim 10^{-8}\,\mathrm{nm}^{-1}$) for clouds and 1×10^{-5} ($1 \times 10^{-7}\,\mathrm{nm}^{-1}$) for background aerosol to 1.3×10^{-3} ($1.3 \times 10^{-5}\,\mathrm{nm}^{-1}$), whereas most of the measurement points were in the range of 5×10^{-5} to 7×10^{-4} ($6 \times 10^{-7}\,\mathrm{nm}^{-1}$ to $9 \times 10^{-6}\,\mathrm{nm}^{-1}$). I.e., the fluorescence backscatter coefficient was about four orders of magnitude lower than the elastic ones, which agrees with the findings by Veselovskii et al. (2020).

In the following, four interesting case studies are presented in several subsections. In Sec. 3.1, the fluorescence properties of wildfire smoke are discussed by analyzing an optically and geometrically thick smoke layer on 4 July 2023. In Sec. 3.2, we first demonstrate the ability of the fluorescence lidar technique to detect optically thin aerosol layers by presenting two case studies (Sec. 3.2.1 and Sec. 3.2.2). Subsequently, we discuss the reasons for the increased sensitivity of the fluorescence channel to aerosol particles in Sec. 3.2.3. Finally, we underline the importance of this new capability by presenting a striking smoke-cirrus interaction case study in Sec. 3.2.4.

3.1 Fluorescence of wildfire smoke – 4 July 2023

In the spring and summer of 2023, huge wildfires raged across Canada, with unusual intensity in the provinces of Alberta and British Columbia. With the prevailing westerly winds, large amounts of biomass-burning aerosol were transported towards Europe. As a result, we frequently observed wildfire smoke layers over Leipzig from mid-May to mid-July 2023.

As a first example, the fluorescence of an optically thick plume of wildfire smoke on 4–5 July 2023 shall be characterized. Figure 2 displays the height-time distributions of the particle backscatter coefficient at 532 nm, the fluorescence backscatter coefficient and the particle depolarization ratio at 532 nm for this night. Figure 2(a) shows a highly polluted troposphere~~, with~~ an overall aerosol optical depth (AOD) of around 0.8 at 532 nm~~)~~. This agrees well with data from the Aerosol Robotic Network (AERONET), where AOD values of around 0.75–0.8 were retrieved at 500 nm ~~and~~ at 17:00 UTC on 4 July 2023. An optically thick aerosol layer extended from 3.4 to 5.8 km height. To determine its optical properties, a 1-hour time period was considered for temporal averaging. Figure 2(d) shows the vertical profiles of the fluorescence and elastic backscatter coefficients, together with the fluorescence capacity averaged over the time period from 21:00 to 22:00 UTC. At the optically thickest part, β_{532} reached values of up to $5\,\mathrm{Mm}^{-1}\,\mathrm{sr}^{-1}$. The 532 nm AOD of the whole layer amounted to around 0.48.

The optical properties of this aerosol layer, ranging from 3.4 to 5.8 km height, were then used to determine the aerosol type. The fact that the lidar ratio at 532 nm (60 sr) was significantly larger than the one at 355 nm (38 sr) and the high backscatter-related Ångström exponent (1.66) are characteristic for aged BB aerosol (Müller et al., 2005; Ansmann et al., 2009; Ohneiser et al., 2021, 2022; Hu et al., 2022; Janicka et al., 2023). Furthermore, these retrieved lidar ratio values are in the same range as reported for aged wildfire smoke in previous studies (e.g., Murayama et al., 2004; Ansmann et al., 2009; Haarig et al., 2018; Hu et al., 2019). The low particle depolarization ratio ($\delta_{532} \leq 0.07$, cf. Fig. 2(c)) in the layer from 3.4 to 5.8 km height points to a spherical shape of the particles, which is also typical for aged wildfire smoke in the middle free troposphere (Haarig et al., 2018). Thus, it can be concluded that this tropospheric aerosol layer consisted of aged BB aerosol particles.
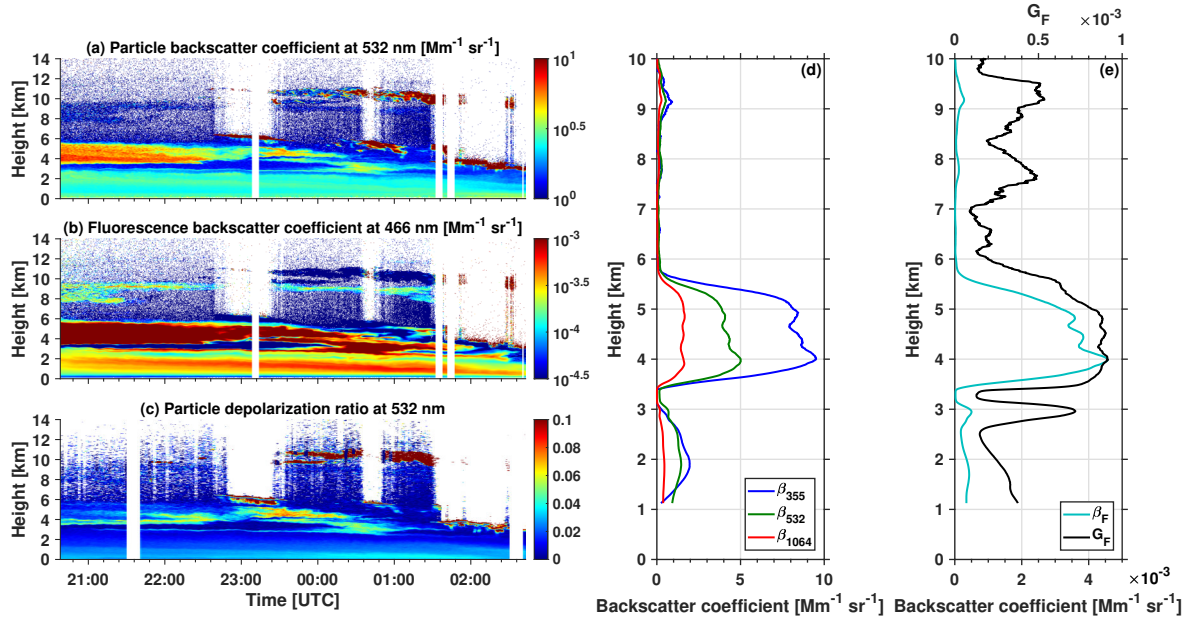
Figure 2. Height-time distributions of (a) particle backscatter coefficient at 532 nm and (b) fluorescence backscatter coefficient (β_F) measured with the MARTHA system and (c) particle depolarization ratio at 532 nm from Polly$^\mathrm{XT}$ on 4–5 July 2023. ~~(d)~~ Vertical profiles of ~~β_F and~~ (d) the elastic backscatter coefficients and (e) β_F together with the fluorescence capacity (G_F) from 21:00 to 22:00 UTC on 4 July 2023.

250 Figs. 2(b) and (d) show a very high fluorescence backscatter coefficient ($\beta_\mathrm{F} \approx 2.75 \times 10^{-3}\,\mathrm{Mm}^{-1}\,\mathrm{sr}^{-1}$) for this smoke layer and a corresponding fluorescence capacity of $G_\mathrm{F} \approx 7.8 \times 10^{-4}$. In other words, smoke shows very high values of fluorescence capacity compared to other particle types and can thus be clearly identified through this new quantity. These values ~~and the features~~ witnessed in our observations ~~corroborate~~ agree with the findings by ~~Reichardt et al. (2018) and~~ Veselovskii et al. (2020).

255 Considering the entire 2023 wildfire season, the fluorescence capacity G_F (spectral fluorescence capacity G_F^{355}) of smoke varied from ~~1~~ 1×10^{-4} to 13×10^{-4} ($1.5 \times 10^{-6}\,\mathrm{nm}^{-1}$ to $13 \times 10^{-6}\,\mathrm{nm}^{-1}$). Thereby, values of $2\text{–}7 \times 10^{-4}$ ($2\text{–}9 \times 10^{-6}\,\mathrm{nm}^{-1}$) were observed most frequently, which agrees with the results of Hu et al. (2022) and Veselovskii et al. (2022a), who reported values of G_F in the range of $1\text{–}4.5 \times 10^{-4}$ for their observations at Lille, France. The observed values of G_F^{355} are also in a range similar to the spectral fluorescence capacities of BB aerosol that were reported by Reichardt et al. (2018), although for a
260 broader wavelength range (455–530 nm). The particle depolarization ratio at 532 nm was low (below 0.07) for most (95 %) of the investigated smoke layers.

Figure 3. Height-time distributions of (a) range-corrected lidar signal at 1064 nm, (b) fluorescence backscatter coefficient (β_F) and (c) fluorescence capacity measured with the MARTHA system on 21 September 2022. ~~(d)~~ Vertical profiles of ~~β_F and (d)~~ the elastic backscatter coefficients and (e) β_F together with the fluorescence capacity (G_F) from 19:04 to 21:04 UTC on 21 September 2022.

3.2 Detection of optically thin aerosol layers with the fluorescence channel

Besides its relevance for aerosol type identification, our results suggest an additional capability of a fluorescence lidar: to detect optically thin aerosol layers. In several measurements with the new fluorescence channel, an enhanced fluorescence signal
265 revealed the presence of aerosol layers that went unnoticed when employing only the elastic-backscatter detection channels. Three exemplary measurement cases are discussed in the following sections.

3.2.1 Hidden smoke layers – 21 September 2022

Figure 3 shows the height-time distributions of the range-corrected lidar signal at 1064 nm (a), the fluorescence backscatter coefficient (b) and the fluorescence capacity (c). According to the elastic backscatter signal in Fig. 3(a), the upper troposphere
270 appears to be rather aerosol-free. Only the polluted boundary layer ~~,~~ and some thin layers up to 4 km height indicated aerosol presence, and a thin cloud was visible at around 4 km height from 21:00 to 22~~UTC indicated aerosol presence~~:00 UTC. However, an enhanced fluorescence backscatter coefficient in Fig. 3(b) reveals several other fluorescing aerosol structures throughout the middle and upper troposphere (at around 5, 6.5, 9 and 9.75 km height). This already illustrates ~~already~~ that with measurements of aerosol fluorescence, thin aerosol layers can be identified more easily from lidar quicklooks and therefore chosen
275 for detailed analysis. Looking at the vertical profiles, this measurement case appears even more impressive. Figure 3(d) displays

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the time-averaged vertical profiles of the fluorescence and elastic backscatter coefficients together with the fluorescence capacity. The profiles were averaged over the 2-hour time period from 19:04 to 21:04 UTC to exclude the cloud, which was present at around 4 km height from that point onwards. The lowest (3.3 km) and most fluorescent ($\beta_F \approx 2.5 \times 10^{-5}\,\mathrm{Mm}^{-1}\,\mathrm{sr}^{-1}$) layer above the boundary layer still shows clearly enhanced elastic backscatter coefficients at all three wavelengths. In the mid-level layers at around 5 and 6.5 km height, the 532 nm and 1064 nm backscatter coefficients are only slightly enhanced compared to the background. For the layer at 6.5 km height, their corresponding maxima are at higher altitudes than the distinct maximum in the fluorescence backscatter coefficient. The 355 nm backscatter coefficient even fails to resolve the aerosol layer at 6.5 km. However, all elastic-backscatter detection channels reach their limits with the two high layers at 9 and 9.75 km altitude. While the 355 nm backscatter coefficient is completely noisy in this altitude range, β_{532} and β_{1064} do show maxima in the altitude range of the increased β_F. But these maxima are difficult to distinguish from the background, which is likewise already quite noisy. Thus, it is unlikely that these two higher layers would have been detected as aerosol layers without the additional fluorescence information, especially because the particle depolarization ratio (not shown) is also quite low, around 2 %.

The overall AOD of this measurement case was around 0.13 at 532 nm, which is consistent with AERONET data showing 0.1 at 500 nm, whereas the majority of the aerosol was found in the boundary layer (AOD ≈ 0.1). The smoke layers above the boundary layer only added up to an AOD of around 0.03. The two thinnest layers at around 9 and 9.75 km height even had an AOD of only 0.002 each at 532 nm.

3.2.2 A thin smoke layer in the UTLS – 15 May 2023

Another example of such "unnoticeable" layers is the night of 15-16 May 2023. Figure 4(a-c) displays the height-time distributions of the range-corrected lidar signal at 532 nm, the fluorescence backscatter coefficient and the particle depolarization ratio at 532 nm. The vertical profiles of the backscatter coefficients together with the fluorescence capacity for the period of 01:15 – 02:15 UTC are shown in Fig. 4(d). This measurement case is characterized by pronounced fluorescent aerosol layers (532 nm AOD ≈ 0.05), ranging from 4 to 6.7 km height. The high fluorescence capacity and lidar ratio values allow us to identify the aerosol particles present as wildfire smoke. For further discussion, we consider a height-constant and rather homogeneous layer, ranging from 4.6 to 6.1 km (layer 1). Layer 1 shows a mean fluorescence capacity of around 5.6×10^{-4} and lidar ratios of around $40\,\mathrm{sr}$ at 355 nm and $70\,\mathrm{sr}$ at 532 nm. The particle depolarization ratio at 532 nm is low (around 1.7 %), indicating a well-advanced aging process of the smoke particles. At around 11 km height, the range-corrected signal at 532 nm in Fig. 4(a) shows another highly fluorescent smoke layer ($G_F \approx 6.5 \times 10^{-4}$), which we will refer to as layer 2 in the following. This higher value of the fluorescence capacity indicates a more efficient fluorescence emission in layer 2 than in layer 1. The reason for this remains unclear. On the one hand, this could be purer smoke, while layer 1 could also contain a small proportion of another less fluorescent aerosol type. On the other hand, the BB aerosol in both layers could differ in chemical composition and optical properties due to different fire sources
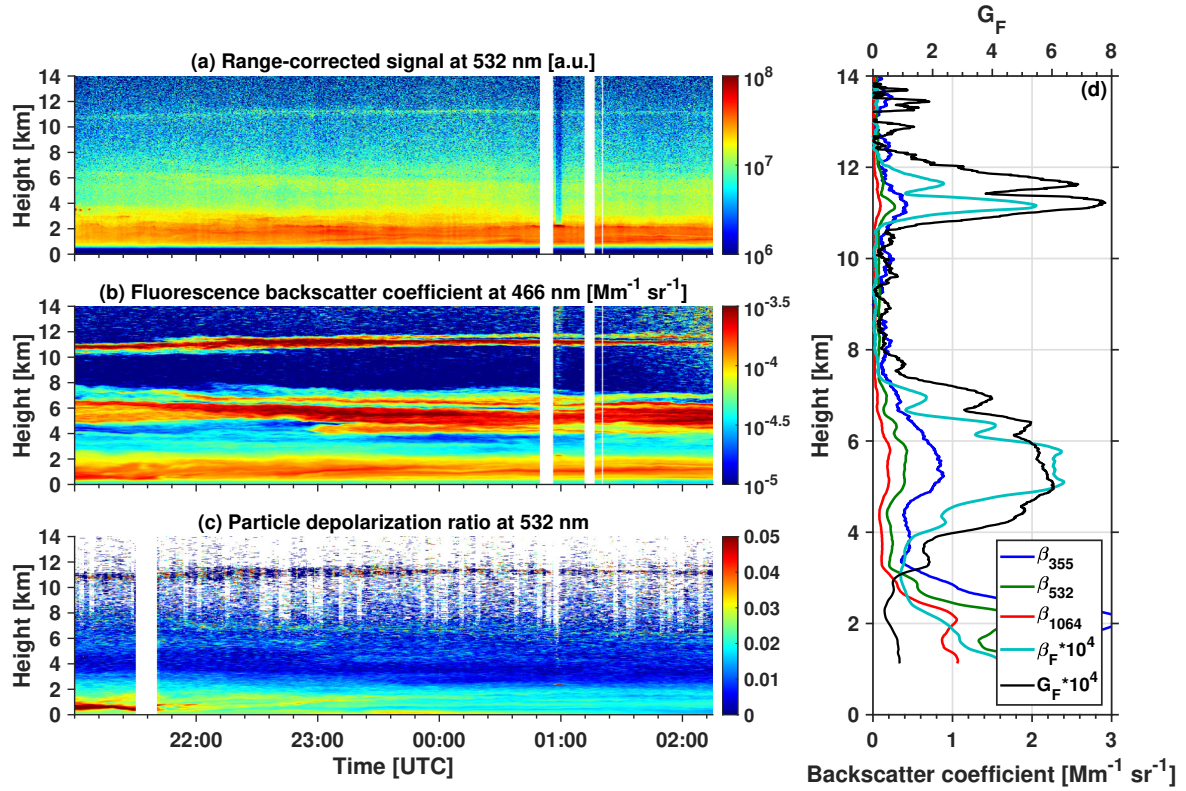
Figure 4. Height-time distributions of (a) range-corrected lidar signal at 532 nm and (b) fluorescence backscatter coefficient (β_F) measured with the MARTHA system and (c) particle depolarization ratio at 532 nm from Polly^{XT} on 15–16 May 2023. (d) Vertical profiles of β_F and the elastic backscatter coefficients together with the fluorescence capacity (G_F) from 01:15 to 02:15 UTC on 16 May 2023.

310 ~~pathways and pointing~~ and transport mechanisms. Backward trajectory analyses point generally to the same source region (the northern part of the North American continent) for both altitudes. ~~Thus, the aforementioned Canadian wildfires can be assumed as the corresponding smoke sources. Hence, the difference in the fluorescence capacities can be attributed to the distinct environmental conditions during atmospheric transport. At the upper smoke layer~~ However, this does not exclude the possibility of slightly different fire sources. The optical properties of the smoke particles differ slightly between the two

315 layers. The lidar ratios in layer 2 (55 sr at 355 nm and 75 sr at 532 nm) are slightly higher than the lidar ratios in layer 1. Furthermore, in layer 2, the particle depolarization ratio was slightly enhanced ($\delta_{532} \approx 6.5\%$) compared to ~~the lower layer~~ . ~~This indicates~~ layer 1, indicating a more irregular shape of the ~~particles~~ smoke particles in layer 2 (although in general terms this is still almost spherical)~~, meaning that the aging process had not yet progressed as far as in the layer at around 6~~. The particle size seems to play a minor role for depolarization in this case, as the backscatter-related Ångström exponent for the

320 355 and 532 ~~km height, where the aerosol particles may have swollen to spherical shapes due to the adsorption of water vapor from the ambient air (Haarig et al., 2018; Ansmann et al., 2021). With this hygroscopic growth, the elastic backscattering may~~

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~~have enhanced, which may have decreased the fluorescence capacity (Veselovskii et al., 2023). If a liquid shell has developed around the smoke particles, it may even have reduced the fluorescence emission of the smoke particles, an effect also known as fluorescence quenching (e.g., Lakowicz, 2006). Due to the lower humidity and temperature at higher altitudes, the aging~~

325 ~~may have proceeded more slowly in the upper aerosol layer , so that the smoke particles maintained a more irregular shape (Knopf et al., 2018; Ansmann et al., 2021) anda higher fluorescence capacity~~ nm wavelength pair (1.75 for layer 1 and 1.55 for layer 2) indicates only slightly larger particles in layer 2.

All in all, these differences in aerosol optical properties suggest different fire sources and/or transport mechanisms for both smoke layers. The observed difference in fluorescence capacity is therefore expectable, but its cause remains unclear.

330 Furthermore, Fig. 4(b) reveals another aerosol layer (layer 3) with enhanced fluorescence ~~directly above~~ at around 11.7 km~~.~~, directly above layer 2. The complete structure of ~~this layer~~ layer 3 remains unnoticed in the range-corrected signal in Fig. 4(a). This impression is confirmed by the vertical profiles in Fig. 4(d). The thin ~~aerosol layer at around 11.7 km~~ layer 3 cannot be distinguished from the background noise in β_{355}. β_{532} and β_{1064} exhibit a slight increase, although this increase is only very weakly pronounced at 1064 nm. So, only the 532 nm backscatter coefficient shows a clear peak for this layer ~~.~~3. At a closer

335 look, also the time-height distribution of the particle depolarization ratio at 532 nm in Fig. 4(c) indicates ~~this layer~~ layer 3 by slightly increased values at this altitude. But again, it would have been hard to recognize this layer from the elastic backscattering products alone, without having a clearer picture of the aerosol situation from the fluorescence channel. This underlines the potential of the fluorescence lidar technique beyond aerosol characterization. Fluorescence backscatter can be used for the detection of aerosol layers in scenarios where concentrations are below the lower detection limit of the elastic-backscatter

340 channels.

3.2.3 On the capabilities of a dedicated (aerosol) fluorescence channel

The ~~example cases presented above have clearly shown~~ measurement cases presented demonstrate the advantages of adding fluorescence observations to the analysis. ~~In the following , these enhanced capabilities are discussed~~ The following paragraph discusses the enhanced capabilities of a fluorescence backscatter channel compared to the classical elastic backscatter channel.

345 The three elastic-backscatter channels rely on the principle of elastic backscattering of the emitted laser radiation, which occurs at both air molecules and aerosol particles. ~~As a result, the received backscattered signal intensity is the sum of the radiation scattered by air molecules and aerosol particles. Figure 5(a) illustrates this context by showing the molecular (blue curve), particle (magenta curve) and total (molecular + particle, green curve) backscatter coefficient at 532 nm for the time period from 01:15 to 02:15 UTC on 16 May 2023. Vertical profiles of (a) particle (magenta), molecular (blue) and total (green)~~

350 ~~backscatter coefficients at 532 nm and fluorescence backscatter coefficient (black) and (b) range-corrected signals at 532 nm (green) and 466 nm (black) on 16 May 2023 for the time period from 01:15 to 02:15 UTC. Due to the strong reduction of the air density with height, the molecular scattering intensity steadily~~ Because of the strong decrease of air and aerosol density, the scattering intensity strongly decreases with height. ~~Aerosol particles are usually most concentrated close to the ground and tend to decrease with height as well, although this is not necessarily always the case. The resulting wide range of values,~~

355 ~~together with the inherent signal reduction~~ In addition, the backscatter detected by the lidar further decreases with height due

to the solid angle of the telescope and atmospheric extinction. As a result, the lidar-signal detection needs to cover a wide dynamic range. Elastic signals are usually attenuated in the detection unit to keep them in a manageable range for a single channel. This maximizes the vertical coverage, but at the expense of sensitivity to aerosol particle changes.

A fluorescence channel is dedicated to aerosol particles only. Thus, it can help to increase the sensitivity to aerosol particles by eliminating the molecular component. Furthermore, the fluorescence return does not only scale to the number of particles but also to its cross-section, which is directly related to the fluorescence capacity of the aerosol particles. I.e., a smoke layer will contrast more to the background than a dust layer because of the higher ability of smoke particles to fluoresce. This feature enhances the capabilities of such a channel to detect smoke particles in the atmosphere.

Figure 5(a) illustrates this context by showing the molecular, particle and total (molecular + particle) backscatter coefficient at 532 nm for a measurement on 16 May 2023. The contrast between the smoke layer (4–7 km) and the background aerosol (8–10 km) in the fluorescence backscatter is significantly more pronounced than in the elastic particle backscatter coefficient. The enhanced detection ability in the case of fluorescing aerosol particles becomes evident when comparing the observed lidar signals (background corrected), as depicted in Fig. 5(b) for this measurement case. Especially the aerosol layers at around 11 km stand out much clearer from the background in the fluorescence signal than in the elastic signal.
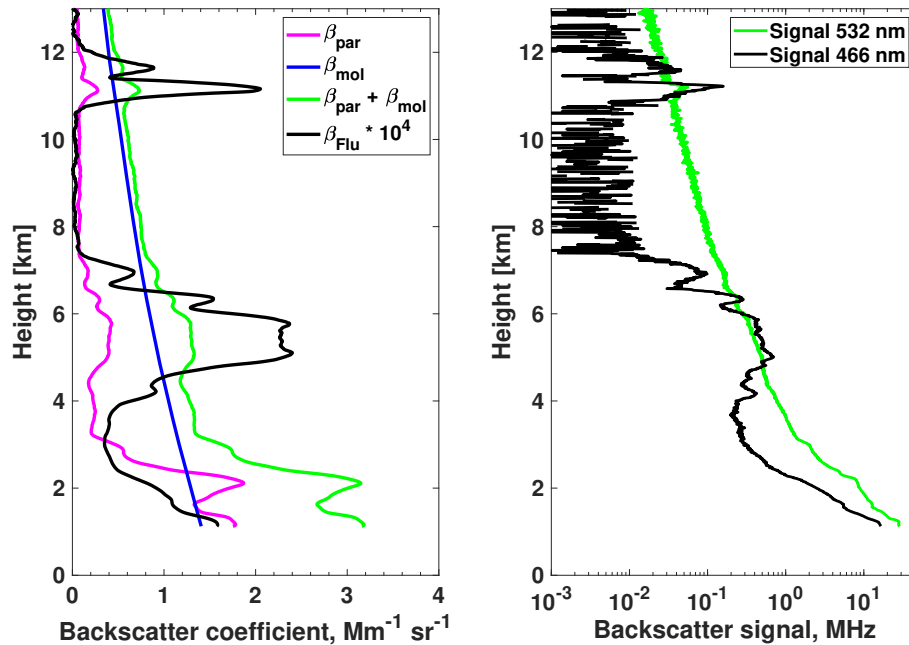
Figure 5. Vertical profiles of (a) particle (magenta), molecular (blue) and total (green) backscatter coefficients at 532 nm and fluorescence backscatter coefficient (black) and (b) background-corrected signals at 532 nm (green) and 466 nm (black) on 16 May 2023 for the time period from 01:15 to 02:15 UTC.

3.2.4 Atmospheric implication: Smoke-cirrus interaction – 29 May 2023

Now, after discussing the possibility of detecting such thin aerosol layers, the question of their relevance in atmospheric re-
390 search arises. Because of their low optical thicknesses (of typically ≤ 0.01), such aerosol layers might not have a relevant radiative effect, but they may impact cloud formation, e.g., by serving as INPs. In both cases presented above (Sec. 3.2.1 and 3.2.2), the measurements of the fluorescence backscatter coefficient revealed thin wildfire smoke layers at rather high altitudes around the tropopause. This altitude range, also referred to as ~~upper troposphere and lower stratosphere (UTLS)~~ UTLS region, is a common site for the formation of cirrus clouds. ~~And indeed, several~~ However, the ability and relevance of
395 smoke particles to act as INPs is an open question in the literature. Although a few available observations showed enhanced immersion-mode INP concentrations inside of BB aerosol plumes (Barry et al., 2021; McCluskey et al., 2014), wildfire smoke is considered to be a rather inefficient INP at temperatures above -30 °C compared to other aerosol types such as dust (e.g., Barry et al., 2021; Knopf et al., 2018). Thus, BB aerosol is, in general, not considered a relevant INP source in mixed-phase cloud processes. Likewise, in situ assessments have suggested that BB aerosol particles rarely freeze to form cirrus clouds
400 (Froyd et al., 2009, 2010). However, the authors could not exclude the INP ability of BB particles due to temperature limitations in their experimental setup. Recent lidar-based studies discussed the potential of smoke particles to promote freezing via

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deposition and provided evidence of BB aerosol acting as the main INP source in cirrus clouds observed at Limassol, Cyprus (Mamouri et al., 2023) and in the Arctic (Ansmann et al., 2024a, b). Simulations considering gravity waves further explain how heterogeneous freezing overtakes the main role, consuming quickly the water vapor and reducing supersaturation, hampering

405 in this way homogeneous freezing (Ansmann et al., 2024a). Thus, investigations of possible smoke-cirrus interactions in the UTLS region are an important topic for future studies.

Also, several of our measurement cases during the 2023 wildfire season showed cirrus clouds directly below such thin smoke layers. One example (29-30 May 2023) is displayed in Fig. 6 and will be discussed in the following.

The range-corrected lidar signal at 532 nm in Fig. 6(a) shows cirrus clouds, that extended from 7 to 11.5 km at the beginning

410 of the measurement. Above, enhanced values of the fluorescence backscatter coefficient (see Fig. 6(b)) reveal the presence of a smoke layer at 10.5 to 12 km height, that was not visible in the elastic-backscatter lidar signal over large parts of the observation period. Only at the end of this measurement (around 02:00 UTC), when the clouds became thinner and more scattered, the aerosol layer could be anticipated vaguely from weak signatures in the range-corrected signal (cf. Fig. 6(a)).

The time-height plot of the fluorescence capacity in Fig. 6(c) reveals another feature of fluorescence backscattering that is

415 exclusive to aerosol particles. Pure water does not fluoresce, and, as our measurements from 2022–2023 showed, hydrometeors such as cloud droplets and ice crystals, exhibit the lowest values of fluorescence capacity. In combination, elastic-backscatter and fluorescence channels can unambiguously differentiate aerosol particles and hydrometeors that coexist within the same air volume. This feature has also been pointed out in previous studies (Reichardt et al., 2018; Veselovskii et al., 2022a) and opens a new door into aerosol and cloud detection.

420 Due to this characteristic, the fluorescence capacity clearly shows the positions of the aerosol and cloud layers relative to each other in one plot. Remarkably, the upper boundary of the cirrus clouds coincides with the lower boundary of the fluorescing smoke layer for large parts of the observation period. The elastic-backscatter signal in Fig. 6(a) clearly shows pronounced virga structures (i.e., stripes of falling ice crystals). Such an arrangement has already been reported in the literature for smoke layers and cirrus clouds observed over the eastern Mediterranean and in the Arctic (Mamouri et al., 2023; Ansmann et al., 2024b).

425 This case reveals another feature of fluorescence backscattering that is Height-time distributions of (a) range-corrected lidar signal at 532 nm, (b) fluorescence backscatter coefficient (β_F) and (c) fluorescence capacity measured with the MARTHA system on 29 - 30 May 2023. Vertical profiles of β_F and the elastic backscatter coefficient at 532 nm together with the fluorescence capacity (G_F) from (d) 21:00 to 22:00 UTC on 29 May 2023 and (e) 0:15 to 1:15 UTC on 30 May 2023. exclusive to aerosol particles. Water content can quench the fluorescence scattering, and, as our measurements from 2022–2023 showed,

430 hydrometeors such as droplets and ice crystals exhibit the lowest values of fluorescence capacity. This feature has also been pointed out in previous studies (Reichardt et al., 2018; Veselovskii et al., 2022a) and opens a new door into aerosol and cloud detection. In combination, elastic-backscatter and fluorescence channels can unambiguously separate aerosol particles and hydrometeors that are coexisting within the same air volume.The arrangement of the cloud and the aerosol layer in this case indicates that the ice nucleation happened at the cloud top, from where the freshly formed ice crystals were falling down,

435 thus producing the aforementioned falling stripesAt the beginning of the measurement in the night of 29 May, parts of the cirrus clouds were even embedded in the smoke layer. Furthermore, the smoke layer slowly rose in altitude towards the end

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of the measurement. At the same time, the cloud top rose first, and later, the clouds even became scattered and ~~seemed to dissolve~~the ice nucleation and cloud formation seemed to stop. All these facts indicate that the smoke particles may have triggered the cloud formation by serving as INPs.~~However, the ability and relevance of smoke particles to act as INPs is an open question in the literature. Although a few available observations showed enhanced immersion-mode INP concentrations inside of BB aerosol plumes (Barry et al., 2021; McCluskey et al., 2014), wildfire smoke is considered to be a rather inefficient INP at temperatures above -30 °C compared to other aerosol types such as dust (e.g., Barry et al., 2021; Knopf et al., 2018). Thus, BB aerosol is, in general, not considered a relevant INP source in mixed-phase cloud processes. Likewise, in situ assessments have suggested that BB aerosol particles rarely freeze to form cirrus clouds (Froyd et al., 2009, 2010). However, the authors could not exclude the INP ability of BB particles due to temperature limitations in their experimental setup. Recent lidar-based studies discussed the potential of smoke particles to promote freezing via deposition and provided evidence of BB aerosol acting as the main INP source in cirrus clouds observed at Limassol, Cyprus (Mamouri et al., 2023) and in the Arctic (Ansmann et al., 2024a, b). Simulations considering gravity waves further explain how heterogeneous freezing overtakes the main role, consuming quickly the water vapor and reducing supersaturation, hampering in this way homogeneous freezing (Ansmann et al., 2024a). Investigations of possible smoke-cirrus interactions in the UTLS region will be an important topic for future studies.~~

~~At the beginning of the measurement on 29 May, parts of the cirrus clouds were even embedded in the smoke layer. Figure 6(d)displays~~ This hypothesis is further supported by Figs. 6(d-e), which display the vertical profiles of the elastic and fluorescence backscatter coefficients, together with the fluorescence capacity from 21~~:~~00 to 22~~UTC. ~~:00 UTC and the temperature and relative humidity over ice from a radiosonde launched at the nearest station Lindenberg (150 km away) at 18 UTC on 29 May 2023. From the high elastic backscatter coefficients, two nucleation sections can be identified. There is a lower part with ice crystals falling from about ~~10.25~~10.5 km (N1 in Fig. 6(d)), and an upper part where ice crystals start falling from 11.75 km (N2). These falling ranges somehow coincide with the aerosol layers observed with the fluorescence channel. Cloud-top temperatures were ~~estimated from radiosondes launched at the nearest station Lindenberg (150 km away)~~ also estimated from the radiosonde at Lindenberg and ranged from ~~-55~~ -58 to -51 °C, a temperature range in which deposition ice nucleation is particularly efficient ~~(Ansmann et al., 2024a). Above, the~~ (Ansmann et al., 2024b). The arrangement of the cloud and the aerosol layer in this case indicates that ice nucleation happened at the cloud top, from where the freshly formed ice crystals were falling down, thus producing the aforementioned falling stripes.

At around 12 km height, the high fluorescence capacity (up to $G_F = 7.5 \times 10^{-4}$ at the maximum) indicates the smoke layer. However, the fluorescence backscatter coefficient shows enhanced values over a wider altitude range, even down to ~~the upper boundary of the lower cloud part~~9 km altitude, which supports the hypothesis that wildfire smoke particles triggered the ice cloud formation. An interesting feature in Fig. 6(d) is the reduction of the fluorescence backscatter at the cloud top of the upper cirrus part ($\beta_F \approx 2.9 \times 10^{-5}\,\mathrm{Mm^{-1}\,sr^{-1}}$) compared to the higher values above this upper cloud layer ($\beta_F \approx 5 \times 10^{-5}\,\mathrm{Mm^{-1}\,sr^{-1}}$) and at the top of the lower part of the cirrus cloud at around 10.9 km height ($\beta_F \approx 6.4 \times 10^{-5}\,\mathrm{Mm^{-1}\,sr^{-1}}$). A possible reason for this reduction could be fluorescence quenching (Lakowicz, 2006) by the ice crystals ~~inside~~ within the upper cloud layer. ~~There is static quenching (the formation of a non-fluorescing complex from the fluorophore and a~~
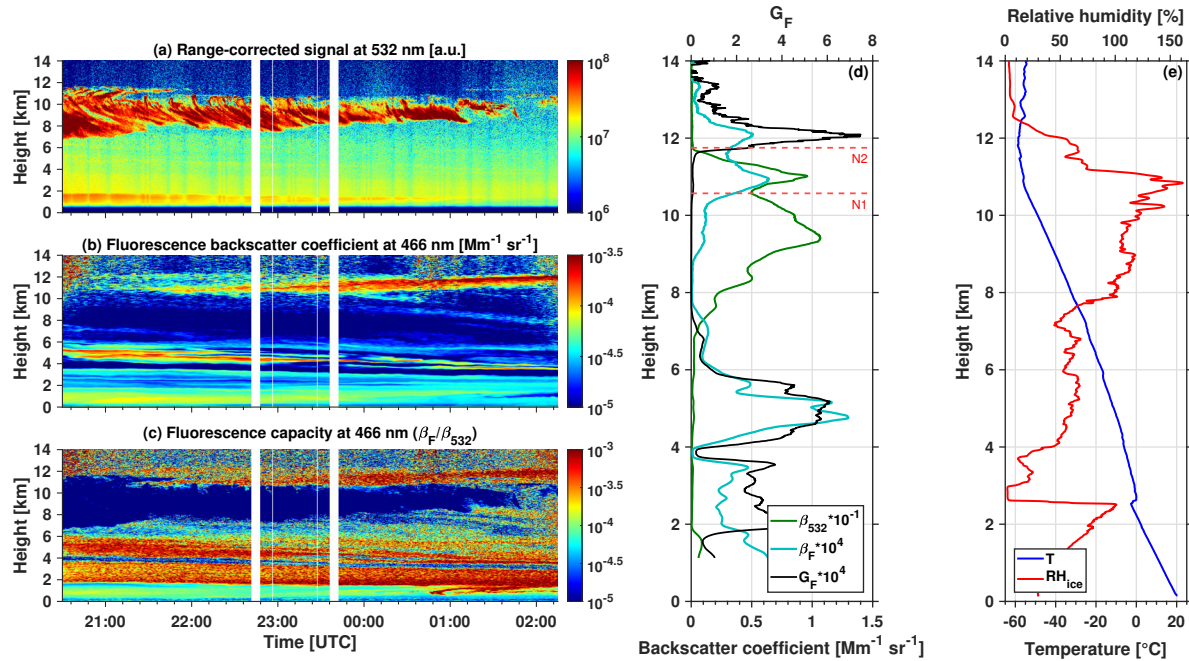
Figure 6. Height-time distributions of (a) range-corrected lidar signal at 532 nm, (b) fluorescence backscatter coefficient (β_F) and (c) fluorescence capacity measured with the MARTHA system on 29 - 30 May 2023. (d) Vertical profiles of β_F and the elastic backscatter coefficient at 532 nm together with the fluorescence capacity (G_F) from 21:00 to 22:00 UTC on 29 May 2023. (e) Vertical profiles of temperature and relative humidity over ice from a radiosounding at Lindenberg at 18 UTC on 29 May 2023.

~~quencher), dynamic quenching (the collision of the fluorophore with a quenching molecule) and resonance energy transfer (Lakowicz, 2006).~~ Water is known to act as a fluorescence quencher for organic fluorophores (e.g., Stryer, 1966; Dobretsov et al., 2014). ~~However, it has to be stated that these studies considered fluorophores in aqueous solutions and not enclosed in~~

475 ~~ice crystals . Besides, studies reported that fluorescence quenching by water is strongest for red-emitting fluorophores, where it can reduce the fluorescence emission by up to a factor of 3 (Maillard et al., 2021). For shorter absorption and emission wavelengths the effect is much weaker (Fürstenberg, 2017; Maillard et al., 2021)~~However, a more plausible explanation would be that some smoke particles acted as INPs and subsequent falling of the ice crystals (indicated by the virga) reduced the number of smoke particles over time. This hypothesis is supported by the fact that the altitude of the reduced β_F coincides with

480 the upper nucleation zone N2.

Another reduction of the fluorescence backscatter coefficient was observed inside the ice virga. In the middle part of the cloud, between 8 and 10 km, β_F ranged at ~~minimal~~ very low values of about 1×10^{-6} to $1 \times 10^{-5}\,\mathrm{Mm}^{-1}\mathrm{sr}^{-1}$. Aerosol scavenging arises as a possible explanation. I.e., ~~after ice nucleation, the freshly formed ice crystals fell down and~~ the falling ice crystals collected most of the aerosol particles (impaction), reducing the aerosol load in the cloud layer. In this case, one would expect

485 an accumulation of smoke particles at or directly below the cloud base. Indeed, near the cloud base at around 7 km height, the
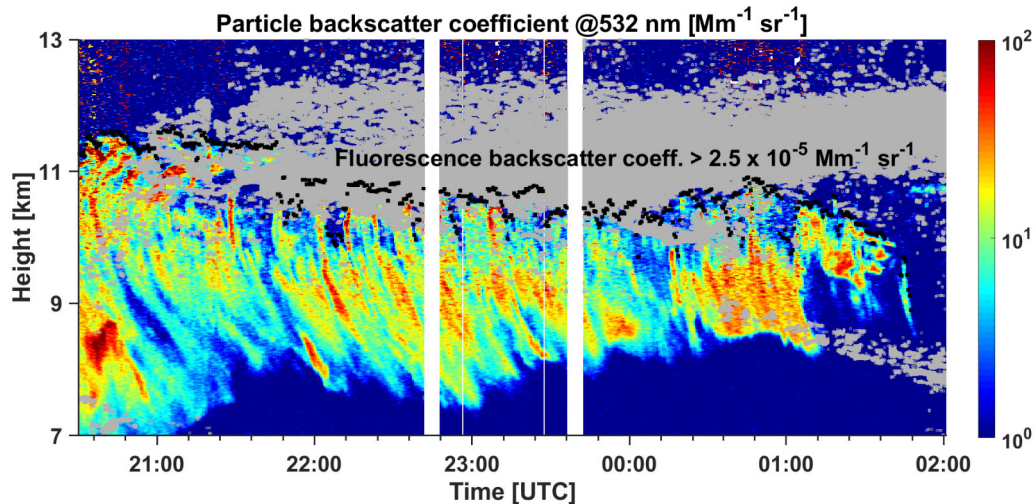
Figure 7. Height-time distributions of the particle backscatter coefficient at 532 nm ~~during~~ in the night ~~from~~ of 29 - 30 May 2023. Height-time bins with a ~~large~~ high fluorescence backscatter coefficient ($> 2.5 \times 10^{-5}$ Mm^{-1} sr^{-1}) are colored in gray. The top of the ice cloud layer was marked in black.

fluorescence backscatter increases again up to 1.4×10^{-5} Mm^{-1} sr^{-1}. A similar situation with a smoke layer directly below the cloud base is visible around 8 km from 01:15 to 02:00 UTC in Fig. 6(c), further supporting the scavenging hypothesis. However, both reductions discussed here could also be due to different aerosol loads and characteristics at the different altitudes. The situation is, in any case, complex, and further investigations of similar cases are needed to characterize aerosol
490 particles inside clouds by fluorescence observations.

In summary, our measurement results suggest two possible interaction pathways between the observed smoke layer and the cirrus clouds: fluorescence quenching and heterogeneous ice nucleation combined with aerosol scavenging. For further illustration, Fig. 7 shows the elastic and fluorescence backscatter coefficients together in one plot. The height-time bins with
495 pronounced aerosol fluorescence (in gray color) along with the elastic backscattering at 532 nm clearly show a major aerosol-cloud interplay. Just before ~~1~~01:00 UTC, an interesting situation arose. The smoke particles were deeply embedded in the cloud, exhibiting two layers: one around 10 km and one between 8 and 9 km, accompanied by a significant increase in the elastic backscatter coefficient.

500 A further potential application of fluorescence lidar is to provide INP information in such cases with low but relevant aerosol presence in the cloud surroundings, especially at the cloud top. A conversion from the unambiguous fluorescence backscatter coefficient to an INP number concentration (N_{INP}) is desirable. An approach applying conversion factors, which link the fluorescence backscatter coefficient with the previously inverted microphysical properties of the fluorescing aerosol particles from

multi-wavelength lidar data, was suggested by Veselovskii et al. (2022a). In the case of a low aerosol load or inside a cloud
layer, the resulting mean conversion factors, together with the fluorescence backscatter coefficient, can then be used to derive
the aerosol surface area concentration, which is needed as input to the INP parameterization (Veselovskii et al., 2022a). An
alternative approach would be to determine N_{INP} directly from ice crystal number information provided by lidar-radar synergy
and find a conversion factor between β_{F} and N_{INP}. Such a factor σ_{F} would be in the form of $\sigma_{\text{F}} = N_{\text{INP}}/\beta_{\text{F}}$ and could be
used for cirrus cloud scenes with comparable temperature and humidity. Preliminary assessments of INP concentrations via
the POLIPHON method (Ansmann et al., 2012; Mamouri and Ansmann, 2014) and ice crystal number concentrations from
lidar-radar synergy (Bühl et al., 2019) suggest a conversion factor in the range of 3–8 $\times 10^4 \, \text{Mm} \, \text{sr} \, \text{L}^{-1}$.

A reliable conversion to link the fluorescence backscatter coefficient to ice nuclei concentrations would be beneficial to inves-
tigate aerosol-cloud interactions, especially in those situations with low aerosol amounts. Further aerosol-cloud cases will be
investigated in the future to evaluate this potential application of fluorescence backscatter information specifically.

4   Conclusions

In this study, we presented the ~~newly-implemented~~ newly implemented fluorescence channel in the lidar system MARTHA,
located at TROPOS, Leipzig, Germany. Some of the first measurements performed with the upgraded system were during the
~~2023~~ summer wildfire season ~~.~~ of 2023. The fluorescence capacity of wildfire smoke mainly ranged between 2×10^{-4} and
7×10^{-4}, thus confirming previously reported values in the literature (Hu et al., 2022; Veselovskii et al., 2022a).

Special care was put into the characterization of the fluorescence lidar, where each component along the optical path was
considered to determine the system efficiency constants needed to derive the new fluorescence parameters. The detection of
optically thin aerosol layers that are only recognizable in the fluorescence signal can significantly improve the detection capa-
bilities of a lidar, which could be critical for low-particle-concentration situations. The enhanced sensitivity results from the
fact that laser-induced fluorescence emission originates exclusively from aerosol particles, while air molecules and hydrome-
teors are excluded from this scattering process. Furthermore, as our observations showed, the new dedicated "particle" channel
enables an unambiguous differentiation between coexisting unactivated aerosol particles and hydrometeors within clouds.

Because of their strong fluorescence and rather low depolarization, the aerosol layers presented in the case studies could
be identified as biomass-burning aerosol. ~~The analysis of backward trajectory calculations suggested that the discussed layers
consisted of smoke particles from Canadian wildfires.~~ The measurements show that such optically thin smoke layers are not
so rare in the UTLS region. This suggests that the atmosphere over Europe might be more polluted than previously thought,
especially during the summer wildfire season. Those thin layers might not have a strong direct radiative impact, but at these
altitudes, smoke particles could become an additional INP source in an otherwise relatively clean atmosphere. Investigating
such aerosol layers with a fluorescence lidar, combined with advanced remote-sensing techniques to assess cloud microphysics,
could provide more clarity about the relevance of heterogeneous freezing of smoke particles in cirrus cloud formation com-
pared to homogeneous nucleation onto small droplets from background particles. Several observations of cirrus clouds directly

below thin biomass-burning aerosol layers suggest that these might be the primary INP source, indicating that heterogeneous freezing is the dominant process. To thoroughly explore this potential aerosol-cloud effect, a larger data set would be beneficial and might provide stronger evidence and more detailed insights into this hypothesis.

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Further instrumental upgrades are currently ongoing in the MARTHA system. A new powerful laser, together with a 32-channel spectrometer, will extend the observational sharpness and aim to provide state-of-the-art information about aerosol and clouds from the ground up to the stratosphere.

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References

Adam, M.: Notes on temperature-dependent lidar equations, Journal of Atmospheric and Oceanic Technology, 26, 1021–1039, https://doi.org/10.1175/2008JTECHA1206.1, 2009.

Albrecht, B. A.: Aerosols, Cloud Microphysics, and Fractional Cloudiness, Science, 245, 1227–1230, https://doi.org/10.1126/science.245.4923.1227, 1989.

Ansmann, A., Riebesell, M., and Weitkamp, C.: Measurement of atmospheric aerosol extinction profiles with a Raman lidar, Optics Letters, 15, 746–748, https://doi.org/10.1364/OL.15.000746, 1990.

Ansmann, A., Wandinger, U., Riebesell, M., Weitkamp, C., and Michaelis, W.: Independent measurement of extinction and backscatter profiles in cirrus clouds by using a combined Raman elastic-backscatter lidar, Applied Optics, 31, 7113–7131, https://doi.org/10.1364/AO.31.007113, 1992.

Ansmann, A., Baars, H., Tesche, M., Müller, D., Althausen, D., Engelmann, R., Pauliquevis, T., and Artaxo, P.: Dust and smoke transport from Africa to South America: Lidar profiling over Cape Verde and the Amazon rainforest, Geophysical Research Letters, 36, https://doi.org/https://doi.org/10.1029/2009GL037923, 2009.

Ansmann, A., Seifert, P., Tesche, M., and Wandinger, U.: Profiling of fine and coarse particle mass: case studies of Saharan dust and Eyjafjallajökull/Grimsvötn volcanic plumes, Atmospheric Chemistry and Physics, 12, 9399–9415, https://doi.org/10.5194/acp-12-9399-2012, 2012.

Ansmann, A., Ohneiser, K., Mamouri, R.-E., Knopf, D. A., Veselovskii, I., Baars, H., Engelmann, R., Foth, A., Jimenez, C., Seifert, P., and Barja, B.: Tropospheric and stratospheric wildfire smoke profiling with lidar: mass, surface area, CCN, and INP retrieval, Atmospheric Chemistry and Physics, 21, 9779–9807, https://doi.org/10.5194/acp-21-9779-2021, 2021.

Ansmann, A., Jimenez, C., Knopf, D. A., Roschke, J., Bühl, J., Ohneiser, K., and Engelmann, R.: Impact of wildfire smoke on Arctic cirrus formation, part 2: simulation of MOSAiC 2019–2020 cases, EGUsphere, 2024, 1–29, https://doi.org/10.5194/egusphere-2024-2009, 2024a.

Ansmann, A., Jimenez, C., Roschke, J., Bühl, J., Ohneiser, K., Engelmann, R., Radenz, M., Griesche, H., Hofer, J., Althausen, D., Knopf, D. A., Dahlke, S., Gaudek, T., Seifert, P., and Wandinger, U.: Impact of wildfire smoke on Arctic cirrus formation, part 1: analysis of MOSAiC 2019–2020 observations, EGUsphere, 2024, 1–29, https://doi.org/10.5194/egusphere-2024-2008, 2024b.

Baars, H., Seifert, P., Engelmann, R., and Wandinger, U.: Target categorization of aerosol and clouds by continuous multiwavelength-polarization lidar measurements, Atmospheric Measurement Techniques, 10, 3175–3201, https://doi.org/10.5194/amt-10-3175-2017, 2017.

Barry, K. R., Hill, T. C. J., Levin, E. J. T., Twohy, C. H., Moore, K. A., Weller, Z. D., Toohey, D. W., Reeves, M., Campos, T., Geiss, R., Schill, G. P., Fischer, E. V., Kreidenweis, S. M., and DeMott, P. J.: Observations of Ice Nucleating Particles in the Free Troposphere From Western US Wildfires, Journal of Geophysical Research: Atmospheres, 126, e2020JD033 752, https://doi.org/https://doi.org/10.1029/2020JD033752, e2020JD033752 2020JD033752, 2021.

Bühl, J., Seifert, P., Radenz, M., Baars, H., and Ansmann, A.: Ice crystal number concentration from lidar, cloud radar and radar wind profiler measurements, Atmospheric Measurement Techniques, 12, 6601–6617, https://doi.org/10.5194/amt-12-6601-2019, 2019.

Burton, S. P., Ferrare, R. A., Hostetler, C. A., Hair, J. W., Rogers, R. R., Obland, M. D., Butler, C. F., Cook, A. L., Harper, D. B., and Froyd, K. D.: Aerosol classification using airborne High Spectral Resolution Lidar measurements – methodology and examples, Atmospheric Measurement Techniques, 5, 73–98, https://doi.org/10.5194/amt-5-73-2012, 2012.

Cadondon, J. G., Napal, J. P. D., Abe, K., Lara, R. D., Vallar, E. A., Orbecido, A. H., Belo, L. P., and Galvez, M. C. D.: Characterization of water quality and fluorescence measurements of dissolved organic matter in Cabuyao river and its tributaries using excitation-emission matrix spectroscopy, Journal of Physics: Conference Series, 1593, 012 033, https://doi.org/10.1088/1742-6596/1593/1/012033, 2020.

Dobretsov, G. E., Syrejschikova, T. I., and Smolina, N. V.: On mechanisms of fluorescence quenching by water, Biophysics, 59, 183–188, https://doi.org/10.1134/S0006350914020079, 2014.

Edner, H., Johansson, J., Svanberg, S., and Wallinder, E.: Fluorescence lidar multicolor imaging of vegetation, Appl. Opt., 33, 2471–2479, https://doi.org/10.1364/AO.33.002471, 1994.

Engelmann, R., Kanitz, T., Baars, H., Heese, B., Althausen, D., Skupin, A., Wandinger, U., Komppula, M., Stachlewska, I. S., Amiridis, V., Marinou, E., Mattis, I., Linné, H., and Ansmann, A.: The automated multiwavelength Raman polarization and water-vapor lidar Polly^{XT}: the neXT generation, Atmospheric Measurement Techniques, 9, 1767–1784, https://doi.org/10.5194/amt-9-1767-2016, 2016.

Floutsi, A. A., Baars, H., Engelmann, R., Althausen, D., Ansmann, A., Bohlmann, S., Heese, B., Hofer, J., Kanitz, T., Haarig, M., Ohneiser, K., Radenz, M., Seifert, P., Skupin, A., Yin, Z., Abdullaev, S. F., Komppula, M., Filioglou, M., Giannakaki, E., Stachlewska, I. S., Janicka, L., Bortoli, D., Marinou, E., Amiridis, V., Gialitaki, A., Mamouri, R.-E., Barja, B., and Wandinger, U.: DeLiAn – a growing collection of depolarization ratio, lidar ratio and Ångström exponent for different aerosol types and mixtures from ground-based lidar observations, Atmospheric Measurement Techniques, 16, 2353–2379, https://doi.org/10.5194/amt-16-2353-2023, 2023.

Froyd, K. D., Murphy, D. M., Sanford, T. J., Thomson, D. S., Wilson, J. C., Pfister, L., and Lait, L.: Aerosol composition of the tropical upper troposphere, Atmospheric Chemistry and Physics, 9, 4363–4385, https://doi.org/10.5194/acp-9-4363-2009, 2009.

Froyd, K. D., Murphy, D. M., Lawson, P., Baumgardner, D., and Herman, R. L.: Aerosols that form subvisible cirrus at the tropical tropopause, Atmospheric Chemistry and Physics, 10, 209–218, https://doi.org/10.5194/acp-10-209-2010, 2010.

Fürstenberg, A.: Water in Biomolecular Fluorescence Spectroscopy and Imaging: Side Effects and Remedies, CHIMIA, 71, 26, https://doi.org/10.2533/chimia.2017.26, 2017.

Groß, S., Esselborn, M., Weinzierl, B., Wirth, M., Fix, A., and Petzold, A.: Aerosol classification by airborne high spectral resolution lidar observations, Atmospheric Chemistry and Physics, 13, 2487–2505, https://doi.org/10.5194/acp-13-2487-2013, 2013.

Haarig, M., Ansmann, A., Baars, H., Jimenez, C., Veselovskii, I., Engelmann, R., and Althausen, D.: Depolarization and lidar ratios at 355, 532, and 1064 nm and microphysical properties of aged tropospheric and stratospheric Canadian wildfire smoke, Atmospheric Chemistry and Physics, 18, 11 847–11 861, https://doi.org/10.5194/acp-18-11847-2018, 2018.

Hansen, J., Sato, M., and Ruedy, R.: Radiative forcing and climate response, Journal of Geophysical Research: Atmospheres, 102, 6831–6864, https://doi.org/https://doi.org/10.1029/96JD03436, 1997.

Hu, Q., Goloub, P., Veselovskii, I., Bravo-Aranda, J.-A., Popovici, I. E., Podvin, T., Haeffelin, M., Lopatin, A., Dubovik, O., Pietras, C., Huang, X., Torres, B., and Chen, C.: Long-range-transported Canadian smoke plumes in the lower stratosphere over northern France, Atmospheric Chemistry and Physics, 19, 1173–1193, https://doi.org/10.5194/acp-19-1173-2019, 2019.

Hu, Q., Goloub, P., Veselovskii, I., and Podvin, T.: The characterization of long-range transported North American biomass burning plumes: what can a multi-wavelength Mie–Raman-polarization-fluorescence lidar provide?, Atmospheric Chemistry and Physics, 22, 5399–5414, https://doi.org/10.5194/acp-22-5399-2022, 2022.

Immler, F., Engelbart, D., and Schrems, O.: Fluorescence from atmospheric aerosol detected by a lidar indicates biogenic particles in the lowermost stratosphere, Atmospheric Chemistry and Physics, 5, 345–355, https://doi.org/10.5194/acp-5-345-2005, 2005.

Janicka, L., Davuliene, L., Bycenkiene, S., and Stachlewska, I. S.: Long term observations of biomass burning aerosol over Warsaw by means of multiwavelength lidar, Opt. Express, 31, 33 150–33 174, https://doi.org/10.1364/OE.496794, 2023.

Jimenez, C., Ansmann, A., Engelmann, R., Haarig, M., Schmidt, J., and Wandinger, U.: Polarization lidar: an extended three-signal calibration approach, Atmospheric Measurement Techniques, 12, 1077–1093, https://doi.org/10.5194/amt-12-1077-2019, 2019.

Kawana, K., Matsumoto, K., Taketani, F., Miyakawa, T., and Kanaya, Y.: Fluorescent biological aerosol particles over the central Pacific Ocean: covariation with ocean surface biological activity indicators, Atmospheric Chemistry and Physics, 21, 15 969–15 983, https://doi.org/10.5194/acp-21-15969-2021, 2021.

Knopf, D. A., Alpert, P. A., and Wang, B.: The Role of Organic Aerosol in Atmospheric Ice Nucleation: A Review, ACS Earth and Space Chemistry, 2, 168–202, https://doi.org/10.1021/acsearthspacechem.7b00120, 2018.

Lakowicz, J. R.: Introduction to Fluorescence, in: Topics in Fluorescence Spectroscopy: Principles, edited by Lakowicz, J. R., chap. 1, pp. 1–26, Springer Science & Business Media, Boston, third edn., 2006.

Li, B., Chen, S., Zhang, Y., Chen, H., and Guo, P.: Fluorescent aerosol observation in the lower atmosphere with an integrated fluorescence-Mie lidar, Journal of Quantitative Spectroscopy and Radiative Transfer, 227, 211–218, https://doi.org/https://doi.org/10.1016/j.jqsrt.2019.02.019, 2019.

Liu, Y., Jia, R., Dai, T., Xie, Y., and Shi, G.: A review of aerosol optical properties and radiative effects, Journal of Meteorological Research, 28, 1003–1028, https://doi.org/10.1007/s13351-014-4045-z, 2014.

Lohmann, U.: Anthropogenic Aerosol Influences on Mixed-Phase Clouds, Current Climate Change Reports, 13, 32–44, https://doi.org/10.1007/s40641-017-0059-9, 2017.

Maciel, F. V., Diao, M., and Patnaude, R.: Examination of aerosol indirect effects during cirrus cloud evolution, Atmospheric Chemistry and Physics, 23, 1103–1129, https://doi.org/10.5194/acp-23-1103-2023, 2023.

Maillard, J., Klehs, K., Rumble, C., Vauthey, E., Heilemann, M., and Fürstenberg, A.: Universal quenching of common fluorescent probes by water and alcohols, Chem. Sci., 12, 1352–1362, https://doi.org/10.1039/D0SC05431C, 2021.

Mamouri, R. E. and Ansmann, A.: Fine and coarse dust separation with polarization lidar, Atmospheric Measurement Techniques, 7, 3717–3735, https://doi.org/10.5194/amt-7-3717-2014, 2014.

Mamouri, R.-E., Ansmann, A., Ohneiser, K., Knopf, D. A., Nisantzi, A., Bühl, J., Engelmann, R., Skupin, A., Seifert, P., Baars, H., Ene, D., Wandinger, U., and Hadjimitsis, D.: Wildfire smoke triggers cirrus formation: lidar observations over the eastern Mediterranean, Atmospheric Chemistry and Physics, 23, 14 097–14 114, https://doi.org/10.5194/acp-23-14097-2023, 2023.

Mattis, I., Ansmann, A., Althausen, D., Jaenisch, V., Wandinger, U., Müller, D., Arshinov, Y. F., Bobrovnikov, S. M., and Serikov, I. B.: Relative-humidity profiling in the troposphere with a Raman lidar, Appl. Opt., 41, 6451–6462, https://doi.org/10.1364/AO.41.006451, 2002.

McCluskey, C. S., DeMott, P. J., Prenni, A. J., Levin, E. J. T., McMeeking, G. R., Sullivan, A. P., Hill, T. C. J., Nakao, S., Carrico, C. M., and Kreidenweis, S. M.: Characteristics of atmospheric ice nucleating particles associated with biomass burning in the US: Prescribed burns and wildfires, Journal of Geophysical Research: Atmospheres, 119, 10 458–10 470, https://doi.org/https://doi.org/10.1002/2014JD021980, 2014.

Mcllrath, T. J.: Fluorescence Lidar, Optical Engineering, 19, 194 494, https://doi.org/10.1117/12.7972549, 1980.

Murayama, T., Müller, D., Wada, K., Shimizu, A., Sekiguchi, M., and Tsukamoto, T.: Characterization of Asian dust and Siberian smoke with multi-wavelength Raman lidar over Tokyo, Japan in spring 2003, Geophysical Research Letters, 31, https://doi.org/https://doi.org/10.1029/2004GL021105, 2004.

Müller, D., Mattis, I., Wandinger, U., Ansmann, A., Althausen, D., and Stohl, A.: Raman lidar observations of aged Siberian and Canadian forest fire smoke in the free troposphere over Germany in 2003: Microphysical particle characterization, Journal of Geophysical Research: Atmospheres, 110, https://doi.org/10.1029/2004jd005756, 2005.

Ohneiser, K., Ansmann, A., Chudnovsky, A., Engelmann, R., Ritter, C., Veselovskii, I., Baars, H., Gebauer, H., Griesche, H., Radenz, M., Hofer, J., Althausen, D., Dahlke, S., and Maturilli, M.: The unexpected smoke layer in the High Arctic winter stratosphere during MOSAiC 2019–2020, Atmospheric Chemistry and Physics, 21, 15 783–15 808, https://doi.org/10.5194/acp-21-15783-2021, 2021.

Ohneiser, K., Ansmann, A., Kaifler, B., Chudnovsky, A., Barja, B., Knopf, D. A., Kaifler, N., Baars, H., Seifert, P., Villanueva, D., Jimenez, C., Radenz, M., Engelmann, R., Veselovskii, I., and Zamorano, F.: Australian wildfire smoke in the stratosphere: the decay phase in 2020/2021 and impact on ozone depletion, Atmospheric Chemistry and Physics, 22, 7417–7442, https://doi.org/10.5194/acp-22-7417-2022, 2022.

Palmer, S. C., Pelevin, V. V., Goncharenko, I., Kovács, A. W., Zlinszky, A., Présing, M., Horváth, H., Nicolás-Perea, V., Balzter, H., and Tóth, V. R.: Ultraviolet Fluorescence LiDAR (UFL) as a Measurement Tool for Water Quality Parameters in Turbid Lake Conditions, Remote Sensing, 5, 4405–4422, https://doi.org/10.3390/rs5094405, 2013.

Pan, Y.-L.: Detection and characterization of biological and other organic-carbon aerosol particles in atmosphere using fluorescence, Journal of Quantitative Spectroscopy and Radiative Transfer, 150, 12–35, https://doi.org/https://doi.org/10.1016/j.jqsrt.2014.06.007, 2015.

Patnaude, R. and Diao, M.: Aerosol Indirect Effects on Cirrus Clouds Based on Global Aircraft Observations, Geophysical Research Letters, 47, e2019GL086 550, https://doi.org/https://doi.org/10.1029/2019GL086550, e2019GL086550 2019GL086550, 2020.

Pinnick, R. G., Hill, S. C., Pan, Y.-L., and Chang, R. K.: Fluorescence spectra of atmospheric aerosol at Adelphi, Maryland, USA: measurement and classification of single particles containing organic carbon, Atmospheric Environment, 38, 1657–1672, https://doi.org/10.1016/j.atmosenv.2003.11.017, 2004.

Quaas, J., Arola, A., Cairns, B., Christensen, M., Deneke, H., Ekman, A. M. L., Feingold, G., Fridlind, A., Gryspeerdt, E., Hasekamp, O., Li, Z., Lipponen, A., Ma, P.-L., Mülmenstädt, J., Nenes, A., Penner, J. E., Rosenfeld, D., Schrödner, R., Sinclair, K., Sourdeval, O., Stier, P., Tesche, M., van Diedenhoven, B., and Wendisch, M.: Constraining the Twomey effect from satellite observations: issues and perspectives, Atmospheric Chemistry and Physics, 20, 15 079–15 099, https://doi.org/10.5194/acp-20-15079-2020, 2020.

Rao, Z., He, T., Hua, D., Wang, Y., Wang, X., Chen, Y., and Le, J.: Preliminary measurements of fluorescent aerosol number concentrations using a laser-induced fluorescence lidar, Appl. Opt., 57, 7211–7215, https://doi.org/10.1364/AO.57.007211, 2018.

Reichardt, J.: Cloud and Aerosol Spectroscopy with Raman Lidar, Journal of Atmospheric and Oceanic Technology, 31, 1946 – 1963, https://doi.org/10.1175/JTECH-D-13-00188.1, 2014.

Reichardt, J., Leinweber, R., and Schwebe, A.: Fluorescing aerosols and clouds: investigations of co-existence, EPJ Web Conferences, 176, 05 010, https://doi.org/10.1051/epjconf/201817605010, The 28th International Laser Radar Conference (ILRC 28), 2018.

Reichardt, J., Behrendt, O., and Lauermann, F.: Spectrometric fluorescence and Raman lidar: absolute calibration of aerosol fluorescence spectra and fluorescence correction of humidity measurements, Atmospheric Measurement Techniques, 16, 1–13, https://doi.org/10.5194/amt-16-1-2023, 2023.

Rodell, M., Houser, P. R., Jambor, U., Gottschalck, J., Mitchell, K., Meng, C.-J., Arsenault, K., Cosgrove, B., Radakovich, J., Bosilovich, M., Entin, J. K., Walker, J. P., Lohmann, D., and Toll, D.: The Global Land Data Assimilation System, Bulletin of the American Meteorological Society, 85, 381 – 394, https://doi.org/10.1175/BAMS-85-3-381, 2004.

Saito, Y., Ichihara, K., Morishita, K., Uchiyama, K., Kobayashi, F., and Tomida, T.: Remote Detection of the Fluorescence Spectrum of Natural Pollens Floating in the Atmosphere Using a Laser-Induced-Fluorescence Spectrum (LIFS) Lidar, Remote Sensing, 10, 1533, https://doi.org/10.3390/rs10101533, 2018.

Schmidt, J.: Dual-field-of-view Raman lidar measurements of cloud microphysical properties: Investigation of aerosol-cloud interactions, Ph.D. thesis, Dissertation, Leipzig, Universität Leipzig, 2014, 2014.

Schmidt, J., Wandinger, U., and Malinka, A.: Dual-field-of-view Raman lidar measurements for the retrieval of cloud microphysical properties, Appl. Opt., 52, 2235–2247, https://doi.org/10.1364/AO.52.002235, 2013.

Stevens, B. and Feingold, G.: Untangling aerosol effects on clouds and precipitation in a buffered system, Nature, 461, 607–613, https://doi.org/10.1038/nature08281, 2009.

Stryer, L.: Excited-state proton-transfer reactions. A deuterium isotope effect on fluorescence, Journal of the American Chemical Society, 88, 5708–5712, 1966.

Sugimoto, N., Huang, Z., Nishizawa, T., Matsui, I., and Tatarov, B.: Fluorescence from atmospheric aerosols observed with a multi-channel lidar spectrometer, Opt. Express, 20, 20800–20807, https://doi.org/10.1364/OE.20.020800, 2012.

Twomey, S.: Pollution and the planetary albedo, Atmospheric Environment (1967), 8, 1251–1256, https://doi.org/https://doi.org/10.1016/0004-6981(74)90004-3, 1974.

Twomey, S.: The Influence of Pollution on the Shortwave Albedo of Clouds, Journal of Atmospheric Sciences, 34, 1149 – 1152, https://doi.org/10.1175/1520-0469(1977)034<1149:TIOPOT>2.0.CO;2, 1977.

Twomey, S. A., Piepgrass, M., and Wolfe, T. L.: An assessment of the impact of pollution on global cloud albedo, Tellus B, 36B, 356–366, https://doi.org/https://doi.org/10.1111/j.1600-0889.1984.tb00254.x, 1984.

Veselovskii, I., Hu, Q., Goloub, P., Podvin, T., Korenskiy, M., Pujol, O., Dubovik, O., and Lopatin, A.: Combined use of Mie–Raman and fluorescence lidar observations for improving aerosol characterization: feasibility experiment, Atmospheric Measurement Techniques, 13, 6691–6701, https://doi.org/10.5194/amt-13-6691-2020, 2020.

Veselovskii, I., Hu, Q., Goloub, P., Podvin, T., Choël, M., Visez, N., and Korenskiy, M.: Mie–Raman–fluorescence lidar observations of aerosols during pollen season in the north of France, Atmospheric Measurement Techniques, 14, 4773–4786, https://doi.org/10.5194/amt-14-4773-2021, 2021.

Veselovskii, I., Hu, Q., Ansmann, A., Goloub, P., Podvin, T., and Korenskiy, M.: Fluorescence lidar observations of wildfire smoke inside cirrus: a contribution to smoke–cirrus interaction research, Atmospheric Chemistry and Physics, 22, 5209–5221, https://doi.org/10.5194/acp-22-5209-2022, 2022a.

Veselovskii, I., Hu, Q., Goloub, P., Podvin, T., Barchunov, B., and Korenskii, M.: Combining Mie–Raman and fluorescence observations: a step forward in aerosol classification with lidar technology, Atmospheric Measurement Techniques, 15, 4881–4900, https://doi.org/10.5194/amt-15-4881-2022, 2022b.

Veselovskii, I., Kasianik, N., Korenskii, M., Hu, Q., Goloub, P., Podvin, T., and Liu, D.: Multiwavelength fluorescence lidar observations of smoke plumes, Atmospheric Measurement Techniques, 16, 2055–2065, https://doi.org/10.5194/amt-16-2055-2023, 2023.

Wang, Y., Hu, R., Xie, P., Chen, H., Wang, F., Liu, X., Liu, J., and Liu, W.: Measurement of tropospheric HO2 radical using fluorescence assay by gas expansion with low interferences, Journal of Environmental Sciences, 99, 40–50, https://doi.org/https://doi.org/10.1016/j.jes.2020.06.010, 2021.

Wright, A. G.: Why photomultipliers?, in: The Photomultiplier Handbook, Oxford University Press, ISBN 9780199565092, https://doi.org/10.1093/oso/9780199565092.003.0001, 2017.

Zhang, M., Klimach, T., Ma, N., Könemann, T., Pöhlker, C., Wang, Z., Kuhn, U., Scheck, N., Pöschl, U., Su, H., and Cheng, Y.: Size-Resolved Single-Particle Fluorescence Spectrometer for Real-Time Analysis of Bioaerosols: Laboratory Evaluation and Atmospheric Measurements, Environmental Science & Technology, 53, 13 257–13 264, https://doi.org/10.1021/acs.est.9b01862, pMID: 31589819, 2019.