# Review to "Invisible aerosol layers: improved lidar detection capabilities by means of laser-induced aerosol fluorescence " by Benedikt Gast et al.

## 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.

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.

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.

## Specific comment

- L20: A logic issue with 'By serving as cloud condensation nuclei (CCN) or ice nucleating particles (INPs)…, the microphysical properties of 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.
- L43: In the atmosphere → In the domain of atmospheric research

- L51-52: “but most were small…” do you mean these lidars were with simple configuration? And please add reference.
- L111: “and only a little loss of fluorescence return” → “with minimal loss of the fluorescence return”
- L114: only if no rain is expected → only when no rain is expected
- L114: Complete nights were collected since 2022 … → Complete night measurements have been collected since 2022…
- L116: the derivation of ~~the~~ new products
- L117: similarly as → similarly to
- 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.
- L122: “and the $C_R$ and $C_F$ represent the corresponding lidar calibration constants.”
- L210: …polluted troposphere, with overall aerosol optical depth (AOD) of around 0.8 at 532 nm.
- 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.
- Figure 2: what are the white points in lidar quicklooks?
- 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.
- L217-220: Please indicate the vertical range which you are describing. Such lidar ratios were detected in tropospheric layer or UTLS layer?
- L243: 22 UTC…. indicated aerosol presence→ 22:00 UTC…were presented in the atmosphere. Just for logic consistency, since a cloud layer also existed.
- L245: This already illustrates ~~already~~ that with measurements…
- L252: the 532 nm backscatter coefficient is ~~already~~ only slightly enhanced
- 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.
- 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 $Mm^{-1}sr^{-1}$, 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?

- 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 %.
- L269: (again in agreement with AERONET: 0.1 at 500 nm)→ which is consistent with AERONET data showing 0.1 at 500 nm
- L264: Another example of such "unnoticeable" layers is the night of (~~from~~) 15-16 May 2023. Similar error in L266.
- 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 ?

- 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?
- L275: Does the back trajectory reveal any difference in transport time or pathway between the two layers?
- L276: 'the aforementional Canadian wildfires' : not clear which wildfire is it referring to
- 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.
- L307-309: This long sentence should better be rephrased to improve clarity.
- 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?
- L315: only a part of the actual (strong) backscatter signal: (strong) ? this is confusing.
- 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?
- L324: dynamical → dynamic
- L324-325: …much smaller, and because of that, the…→ …much smaller. Consequently, the…
- L332: 'UTLS' defined twice
- Figure 6: please add the profiles of temperature and relative humidity with respect to ice.
- L346: "water content can quench fluorescence scattering": this sentence is not clear enough. How does water content influence fluorescence capacity, by supressing fluorescence emission or enhancing elastic scattering?
- L349: separate → differentiate
- 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?

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

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

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

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

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

- 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’…

- 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?

- L390: ‘ranged from ~~(at)~~ the minimal values of…to…’

- L392--394: ‘ In this case,…again up to $1.4*10^{-5}Mm^{-1}sr^{-1}$ .” --- 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.

- Figure 6(e) is not described.

- 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.
- 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 (> $2.5 \times 10^{-5}$ $Mm^{-1}$ $sr^{-1}$) are colored in gray.
- 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…

- L421: newly-implemented → newly implemented
- L422: the 2023 summer wildfire season → the summer wildfire season of 2023
- 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.