Reviewer 1

First of all, we would like to thank the reviewer for careful reading of manuscript and for numerous useful comments. The manuscript was revised in accordance with recommendations.

This study presents a novel method to separate smoke and urban aerosols from their mixtures using a 355-nm fluorescence lidar with five discreet channels, and to retrieve the mass concentration of smoke. The authors assume that the total measured fluorescence backscattering at a given wavelength can be linearly divided into the contributions from smoke and urban aerosol. Specifically, these contributions are calculated by multiplying fluorescence backscattering of pure smoke or pure urban aerosol (serving as reference values) by their respective coefficients, denoted as a and b, which are determined using the least squares method (Eq. (1)). This validity of this approach is verified by applying the fluorescence capacity of pure smoke and pure urban aerosol to calculate their respective backscattering at 355 nm, and then reconstructing the elastic backscatter at 355 nm. Then, using the derived backscatter of smoke and urban aerosol at 355 nm along with several parameters (particle density, lidar ratio, and extinction-to-volume conversion factor), the mass concentration of smoke is finally obtained via the POLIPHON method. The urban aerosol mass concentration is not retrieved due to the lack of a reliable and stable conversion factor.

Several case studies are presented, including long-range transported smoke from north America and southern Russia, as well as an extreme pollution event, demonstrating the feasibility of the proposed method. Using multi-channel fluorescence lidar further advances aerosol classification and enables the quantification of smoke mass concentration from aerosol mixtures. Overall, the method is sound and the manuscript is well-written. The authors have made significant contributions to the development of discreet-channel spectrally resolved fluorescence lidar and its application in aerosol studies. The manuscript can be accepted after minor revision. Some minor comments are given below.

- *One concern is that the method is generally applicable only under relatively dry atmospheric conditions with RH below 70%, as high humidity can include hygroscopic growth and fluorescence quenching. A noted by the authors, this may lead to an underestimation of the derived smoke backscatter and, consequently, the smoke mass concentration, which is currently difficult to assess accurately. While the hygroscopic growth effect can potentially be quantified, could the authors consider quantitively investigating the impact of fluorescence quenching as well (is it feasible?), in order to extend the applicability of the proposed method to a wider range of atmospheric conditions? It would be also be beneficial to specify the applicable RH range in the abstract.*

  Reviewer is right, the hygroscopic growth is one of the factors, limiting application of fluorescence technique for aerosol characterization. At a moment, we are not able to correct the fluorescence quenching and development of approach for such correction is the subject of our current studies. We mention this issue in Conclusion.

- *In this study, the pre-required fluorescence capacity G for pure smoke and pure urban aerosol is calculated on a case-by-case basis, meaning that these values are case-specific and may vary depending on factors such as the smoke source, aging level, chemical composition, and so on. Is it possible to provide an analysis (either in this paper or in future work) that discusses typical G values that could be used for general classifications of smoke types (e.g., from north America and southern Russia, nearly-fresh partial-aged, and fully-aged, …)? Furthermore, is it feasible to obtain G values for pure smoke and pure urban aerosol for each case (in some cases, values from adjacent days may need to be used)? Are the G values for pure smoke highly variable from case to case?*

  The choice of fluorescence spectra for pure smoke and urban aerosol is important. We should recall that for separating the fluorescence contributions of these particles we use normalized spectra, so the absolute values of fluorescence capacity are not relevant. However, we still need them to calculate the corresponding backscattering coefficients. In our recent paper (Veselovskii et al., 2025, https://doi.org/10.5194/acp-25-1603-2025), we analyzed the range of variations in the fluorescence capacity of smoke during 2023. The variation is strong when the entire troposphere is considered, but it is smaller in the lower troposphere. For the period from August to October 2024, in particular, the values varied within approximately $(5–6) \times 10^{-6}$ nm^{-1}. Moreover, in most episodes we can identify a smoke layer above the PBL and derive the capacity for a specific case. For urban particles, the situation is more difficult because background aerosol is often mixed with smoke within the PBL. Therefore, we have to rely on values from other episodes that are free of smoke. In this study we provide mass concentrations only for smoke. To characterize the urban mass concentration, additional studies are needed.

- *The manuscript presents a large number of figures and values across multiple cases. It would be helpful to include a summary table at the end of the result section, listing all key information, including the parameter values for each case.*
  We added a Table with particle parameters in the Appendix. Besides, in the process of revision we introduced modifications to the Results section.

Technical suggestions:

- *L32, for spectral-resolved approach, a recently-published paper (Huang et al., 2025, doi: 1021/acs.est.5c00028) is also relevant.*
  Added

- *L47-48, for unaffected spectral shape, Liu et al. (2022) is also relevant (doi: 10.1109/TGRS.2022.3166191).*
  Added

- *L110, add ‘2024’ after ‘in September’*
  Done

- *L137, ‘only for smoke’, it would be better to mention aging level of the smoke particles*
  We changed it for “aged smoke”, because fresh smoke was not observed during the 2023-2025 period.

- *Caption of Fig. 2., ‘five-day’ -> ‘six-day’*
  Done

- *L176, ‘2000 m’, as seen from Fig. 2, it is more likely linked to the green curve (i.e., 3500 m).*
  We changed it to “2000-3500 m” because smoke occurred within this range.

- *L 181, add the launch time of radiosonde after ‘radiosonde’.*
  Done

- *L289, in the caption of Fig. 10, the period 02:30-03:30 was not the appearance of smoke plume (according to Fig. 8-9). Please confirm.*
  Sorry, it was local time. Corrected for 23:30-00:30.

- *L297, ‘accurately’->‘well’*
  Done

Reviewer 2

First of all, we would like to thank the reviewer for useful comments and for rising important questions. The manuscript was revised in accordance with recommendations.

The manuscript presents a useful method to discriminate smoke and urban aerosols using a five-channel fluorescence lidar. The authors apply a linear separation of fluorescence signals and apply the POLIPHON algorithm to convert retrieved smoke fractions into mass concentrations. The technique is presented through multiple case studies with emphasis on smoke within the planetary boundary layer (PBL). Overall, the approach appears very promising, innovative and relevant for the discrimination of smoke/urban aerosols.
My recommendation is that the manuscript can be published after a few minor revisions. The dataset and overall concept are strong. The methodology and equations are well presented. A few minor points need further elaboration (clarify small methodological details and provide brief sensitivity checks) before publication.

Minor comments:

- *One minor suggestion that may worth clarification is the selection of the fluorescence threshold. It would be helpful to know how this threshold was chosen and how sensitive the smoke/urban separation is to that specific choice.*
  Formally, the algorithm contains no threshold. However, for low signals, the ratios of fluorescence backscattering in different channels become oscillatory. In data analysis, this manifests as oscillations in the retrieved fluorescence components. Normally, in the lower troposphere, we limit our consideration to values where $B_{513} > 0.5$ $Tm^{-1}sr^{-1}nm^{-1}$. Corresponding comment has been added to the manuscript.

- *Another aspect that may deserve further discussion is the stability of the reference spectra under varying humidity conditions. In particular, it would be useful to know whether any sensitivity tests were performed for high-RH cases or if they will be part of another article maybe. Since humidity can influence both backscatter and fluorescence, it would also be helpful to comment on the expected bias in the retrieved smoke mass under humid conditions. Even a brief estimate or qualitative discussion would strengthen the interpretation.*
  Humidity is an important factor in fluorescence measurements. The hygroscopic growth of particles may lead to fluorescence quenching. However, it affects all fluorescence channels equally. As shown in Veselovskii et al. (2025a), even near the cloud base, the ratio of fluorescence backscattering in different channels remains stable. Therefore, we did not observe any effect of relative humidity (RH) on the separation of fluorescence contributions from smoke and urban particles. Nevertheless, RH does affect the calculation of the aerosol backscattering coefficients for individual components. At this stage, we do not consider the retrieval of smoke mass concentration under high RH conditions; this is the subject of our future research. One criterion indicating that RH has no effect is that the aerosol backscattering coefficient β_{355} is well reconstructed by the sum $\beta_{513}^{U} + \beta_{513}^{S}$.

- *The linear separation approach is interesting, but a short justification of why it remains robust in mixed aerosol conditions would improve the methodology section. This is especially relevant where smoke and urban aerosols may strongly overlap.*
  We consider the aerosol as an external mixture, and the linear separation of component contributions is the best we can currently do. At this point, we lack a solid framework for analyzing the interaction between urban aerosol and smoke. In the revised manuscript we mention, that aerosol mixture us assumed to be external.

  Moreover, the definition of the background or urban aerosol state could maybe be described in slightly more detail. It would also be interesting to indicate how sensitive the results are to that definition.
  Our definition of the background urban aerosol comprises a wide range of particle types, including sulfates, nitrates, secondary organic components, black carbon, and others. Among these, the organic components are the primary contributors to fluorescence. Consequently, any change in the proportion between organic and inorganic fractions leads to a variation in the

overall fluorescence capacity of the aerosol mixture. However, it does not provide significant influence to retrieval of smoke mass concentration. Recall, that in our algorithm, we use only shape of fluorescence spectra, which (in contrast to fluorescence capacity) for urban aerosol does not change significantly. In the revised manuscript we added a comment about composition of background aerosol.

- *Finally, for the case studies, a short statement on how representative the selected events are of the full dataset would improve the overall context. This would help to understand whether the examples shown are typical or more exceptional.*

  The episodes considered in this study are representative. In our previous publication, we analyzed variations in fluorescence spectra throughout 2023 and showed that the main spectral features are preserved across seasons. For the present study, we selected episodes to demonstrate different scenarios of smoke and urban aerosol mixing within the PBL. Individual spectra do not vary significantly from one episode to another, so the examples presented are typical. We added corresponding comment to Conclusion.

Technical suggestions:

1. *Line 125: Omit typo ":".*

   Done
2. *L183-183: I suggest changing the time label "23:00-24:00" to "23:00-00:00".*

   Done
3. *Figure 13: In the caption add "00:15-01:15 UTC"*

   Done
4. *Figure 21: In the caption and Fig. 21a, I suggest changing the time label "24:00" to "00:00".*

   Done