

Anonymous Referee #1

We are grateful to Referee for careful reading the manuscript and suggestions. In the process of revision we tried to follow his recommendations.

The paper is well written and describes a new approach of an interference-filter-based aerosol fluorescence lidar with five channels. An aerosol fluorescence lidar is rather helpful to clearly identify pollen, wildfire smoke, and other fluorescing aerosol components and to separate them from mineral dust (that does only weakly fluoresce). I have only minor remarks. The second reviewer provided a good list of comments to the authors so I can be short.

line 71: What is the reason for fluorescence in the case of urban haze? Why is there a difference to wildfire smoke? What about pollen in this context? A few more words would be helpful.

The sources of the fluorescence in the urban haze are not completely understood at a moment. Organic aerosols could be important contributors to the fluorescence signal. As well, organic carbon fraction is probably responsible for strong fluorescence of the smoke. We added corresponding phrase in Introduction section.

line 76: The Adam et al. (2021, ACPD, Part 2) paper was reject and thus should not be cited. Adam et al., Part 1, is available (published).

We removed Part 2 from references.

line 148: At favorable conditions, aging of smoke particles is completed within 2 days. So, the particles were probably aged. However, smoke from North America is much older (10 days), and then may show different properties.

This the goal of our future studies, to see how the aging may influence the fluorescence properties. We removed word “fresh” from title.

line 185: With only one wavelength (355 nm) there is no (good) way to categorized smoke based on lidar ratio. 40 or 60 sr was found for smoke as well as for urban haze. Again, please use alternative citation. Adam et al. (2021) is not a good reference.

Yes, it is important to have 532 nm channel, to categorize the smoke. Unfortunately, we could not do it in the present configuration of the lidar. Reference for Adam 2021 is removed.

lines 197-200: Maybe one should mention that the presented aerosol typing is not optimum. Optimum would be dual-wavelength (355, 532nm) depolarization and lidar ratio observations TOGETHER with the fluorescence observations as well as with humidity observations.

We have added this comment to conclusion

lines 289-290: This is a valuable message of the work. Fluorescence observations at wavelengths < 532 nm are sufficient to distinguish fluorescing urban haze from wildfire smoke. That means, three wavelength lidar observations can be combined with fluorescence lidar observations.

We are in the process of preparation of this kind of the system.

Anonymous Referee #2

We are very grateful to Referee for careful reading the manuscript and for numerous suggestions made in the text. In the process of revision we tried to follow his recommendations.

“The authors present multi-spectral fluorescence backscatter profiles measured with a lidar. The study is of high importance to the lidar community as the fluorescence topic is still not sufficiently explored with lidar systems and the aerosol intensive properties measured here will most probably be used in future aerosol classification studies. Therefore, it is important that the values and respective uncertainties are well reported. I would recommend the publication of this paper after some revisions regarding the following points:”

1. *“Ratios of the fluorescence backscattering are calculated over a broad spectral region (438-614nm) but the authors do not specify how they treat the atmospheric attenuation in the different wavelengths. These spectral effects are not simplified by dividing the two backscatter coefficients and are certainly not negligible. The aerosol extinction cross-section at 614nm is 0.5 times less compared to 438nm for an Angstrom exponent of 2 (typical for biomass burning). Likewise, the molecular extinction cross-section at 614nm is 0.35 times less than the 438nm one. This introduces optical-depth dependent effects that become more and more important as the beam goes deeper in the atmosphere. For instance, the backscatter ratio among 472 and 614nm will be biased by ~4% above an aerosol layer with 0.1 AOD and ~12% above an aerosol layer with 0.3 AOD without including the molecular contribution at all. The authors must include a correction for the atmospheric attenuation in their technique (if not already applied), at least for molecules and for typical Angstrom exponent values. They should also address the error introduced when aerosol with different Angstrom exponent values are present. In Fig. 11 we may probably already see such an effect in the ratios B_{472}/B_{513} and B_{472}/B_{560} as the backscatter (and therefore the extinction) increases with humidity.”*

Referee is right, the correction of aerosol and Rayleigh differential extinction is important. Such correction is performed in our algorithm and the details are given in publication of Veselovskii et al. (2020). The atmospheric transmission of fluorescence signal is calculated at the wavelengths corresponding to the center of the filter transmission band. The error due to the neglect of spectral dependence of the Rayleigh optical depth (OD) inside the filter transmission band is the largest for the shortwave channel (438 nm). However, our computations show that at height of 4000 m this error of fluorescence backscattering coefficient calculation is below 4%. This residual error is not corrected at this stage of research, but it will be done in the next version of algorithm. For correction of aerosol differential extinction, we make an assumption about the extinction Angstrom exponents (EAE). In particular, for the aged smoke the EAE (355-532 nm) is about 1.0 (Hu et al., 2022), though corresponding backscattering Angstrom exponent is higher, about 2.0. All results presented in manuscript were calculated assuming $EAE=1.0$

To quantify the uncertainty, related to the choice of EAE, in revised manuscript we added Fig.4c. The aerosol OD at 355 nm for period considered exceeded 0.55 at 3000 m height. Fig.4c shows profiles of fluorescence backscattering B_{472} and B_{614} calculated for values of EAE $A=0.5, 1.0, 1.5$. Aerosol provides the largest effect to B_{614} , and the difference between values obtained with $A=1.0$ and 1.5 at 3000 m is about 6.5%. For B_{472} this difference is lower, about 5%. However, for the ratios of fluorescence backscattering, the influence of aerosol is lower: for both B_{472}/B_{513} and B_{472}/B_{614} ratios corresponding difference is below 2.0%. Thus observed decrease of

B_{472}/B_{513} and B_{472}/B_{560} in Fig. 11 can not be explained by the aerosol differential extinction. Corresponding comments are added to the revised manuscript.

2. Protective windows for the telescopes are usually deployed in lidar systems. Their spectral reflectance, that also depends on the incidence angle must be taken into account when calculating backscatter ratios over such a broad region. Are there protective windows installed in this system? Is their effect measured and subtracted, or at least included in the calibration with the lamp? The authors must address any such potential issues and if present, correct for them or include them in the error calculation.

Our system uses silica protective window. Reflection coefficient (from two sides for normal incidence) changes from 7.2% to 7% inside 400 – 615 nm spectral range, which we neglected. Reviewer is right that angular dependence of reflection may influence the calibration, so the lamp was located at 4 m distance from telescope entrance and only small central part of lamp radiation (about 50 mm diameter) was used. Thus the incident angle was below 1 dg and angular dependence could be neglected. Different parts of telescope entrance were tested to exclude spatial variation of the channels transmission.

3.--Uncertainties are not sufficiently addressed in the manuscript. There are some error bars in the figures but they are only there for some of the lines. The authors should include them for all. It is also not clear whether these correspond to random noise errors from the signal analysis or from some other source uncertainty as there is no relevant discussion. The authors should comment on how they performed the error estimation.

Estimation of uncertainties is definitely important. For calculation the fluorescence backscattering we use the nitrogen Raman scattering cross section and the values presented in different publications demonstrate difference. Analysis of the reason of this difference is out of the scope of this study and we always used the value from the most recent publication of Venable et al. (2011).

In the manuscript we consider the following uncertainties of the fluorescence backscattering calculation:

- Systematical errors of lamp calibration (ϵ_{cal}). We always used the same lamp. The calibration coefficients obtained during regular calibration procedure, demonstrated variation up to 15% in 614 nm channel, and for the rest of the channels this variation was below 10%.
- Residual error of correction of aerosol and Raleigh differential extinction (ϵ_A) which, for results presented, we estimate to be below 7% for all channels.
- Statistical errors (ϵ_{St}), which were calculated from lidar signals.

For profiles of backscattering coefficient we show the uncertainty $\sqrt{\epsilon_A^2 + \epsilon_{St}^2}$ (systematical error of calibration is not included). When calculating the spectra of fluorescence, we average fluorescence backscattering in the height range of several hundred meters so statistical errors become negligible and we show $\sqrt{\epsilon_A^2 + \epsilon_{Cal}^2}$.

Corresponding comments are added to the revised manuscript.

Minor comments are also included directly inline in the manuscript. The use of English can be in general improved. I would recommend the authors to go through the document again and correct minor grammatical/phrasing issues.

We included suggestions of Referee in the text in the revised manuscript.